

Turbulent transfer of sulphur dioxide to a wheat crop

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SUMMARY

Fluxes of sulphur dioxide to wheat were measured over two growing seasons. The deposition velocity, v_g , depended primarily on affinities of surfaces for SO_2 and varied from less than 0.1 to about 1.5 cm s^{-1} . Stomata were an important sink, and diurnal changes in stomatal resistance accounted for most of the variation in v_g when leaves were green and dry. When leaves were wet with dew, v_g was generally large, irrespective of the physiological state of vegetation. A resistance analogue of transfer to the canopy agrees well with published work and is used to estimate seasonal uptake of SO_2 by the crop, showing that 30% of the sulphur content of the crop at harvest was probably supplied by absorption of SO_2 through stomata.

1. INTRODUCTION

Natural processes which remove pollutants from the atmosphere have been studied in detail in recent years in search of a better understanding of the fate and effects of such materials. All gases emitted into the atmosphere from industrial sources share common physical processes for dispersion but many pollutant gases undergo complex chemical and physical transformation in the atmosphere (Kellogg *et al.* 1972). Products of these transformations and some of the primary pollutants may be returned to the earth's surface in rain (wet deposition) or they may reach the surface by turbulent transfer, a mechanism usually called dry deposition. Understanding the fate of these pollutants requires a knowledge of rates of deposition at the ground by both mechanisms.

Measurements of material returned to the ground in rain are extensive (Stevenson 1968; Eriksson 1963). Analyses of early records of wet deposition, in conjunction with emission data, provided some of the first evidence of the importance of dry deposition for removal of SO_2 from the atmosphere (Meetham 1950). However, only in the last few years have rates of dry deposition of SO_2 been measured directly (Garland *et al.* 1974; Shepherd 1974; Fowler and Unsworth 1974). Garland (1977), reviewing the relatively small number of measurements of dry deposition rate, concluded that the mean deposition velocity (v_g) for SO_2 to vegetation and soil was 0.8 cm s^{-1} . This or similar values have been applied extensively for the evaluation of regional or global atmospheric sulphur budgets (Garland 1974), the assessment of annual inputs of sulphur to various ecosystems (Braekke 1976) and for studies of long-range transport of sulphur compounds (Fisher 1975). As for other gases with sources or sinks at the earth's surface, fluxes of SO_2 to the ground depend on atmospheric transport and surface factors, and although these were separated in the early work, large uncertainties in the measurements combined with a small number of observations prevented further analysis of the results to reveal the sink strengths of the various components at the surface. In this paper an extensive set of micrometeorological measurements of SO_2 fluxes to wheat crops in two growing seasons is reported. Analyses of the variation of fluxes as the state of the atmosphere and of the crop changed show the range of values of v_g and the factors controlling sink strengths. A resistance analogue of the deposition process is then proposed and subsequent analysis enables prediction of rates of SO_2 deposition to grass or cereal crops in a wide range of atmospheric and surface conditions.

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2. THEORY

(a) *Micrometeorology*

Micrometeorological methods are particularly useful in studying the exchange of gases by crops because they enable mean fluxes over short periods (~ 1 hour) to be measured without disturbing the crop canopy. Thom (1975) reviewed the physical principles of the flux-gradient method employed here. Above horizontal uniform surfaces the flux density, F_s , of SO_2 to the ground may be determined from measurements of the vertical gradient of SO_2 concentration, χ , if the eddy diffusivity for SO_2 is known,

$$\text{i.e.} \quad F_s = -K_s d\chi/dz. \quad (1)$$

The two most common methods of determining K_s are the aerodynamic method assuming similarity in fully forced convection with the diffusivity for momentum, and from the energy balance assuming similarity with heat and water vapour diffusivities. Biscoe *et al.* (1975), working at the field sites we used, showed that fluxes of CO_2 determined by both methods from measurements over a barley crop were in agreement during the day, but that the energy balance method was inaccurate at night when net radiation was small. Fluxes of SO_2 at night may be significant because the gas is very reactive with most surfaces, so the aerodynamic method was used to determine K_s .

In all stabilities, the vertical gradient of windspeed (u) may be written in a generalized form

$$\partial u/\partial z = u_* \phi_M / \{k(z-d)\}, \quad (2)$$

where u_* = friction velocity, k = von Kármán's constant, d = zero plane displacement, and where ϕ_M is an empirical stability function with value unity in a neutral atmosphere. An analogous relationship can be written for the gradient of SO_2 concentration:

$$\partial \chi/\partial z = -\chi_* \phi / \{k(z-d)\}, \quad (3)$$

where ϕ is a stability correction for SO_2 transport and

$$\chi_* = F_s/u_*. \quad (4)$$

Hence substituting u_* from Eq. (2) and for χ_* from Eq. (3) the generalized form of the flux gradient equation for SO_2 transport is

$$F_s = -k^2(z-d)^2(\partial u/\partial z)(\partial \chi/\partial z)(\phi_M \phi)^{-1}. \quad (5)$$

It is commonly assumed that turbulent exchanges of heat, water vapour and any other entrained quantity are identical, so that $\phi = \phi_H$, the stability function for heat. Thom *et al.* (1975) called the term $(\phi_H \phi_M)^{-1}$ the stability factor, F , and, reviewing the literature, they concluded that the dependence of F on gradient Richardson number, Ri , where

$$Ri = \{gT^{-1}(\partial \theta/\partial z)(\partial u/\partial z)^{-2}\}(1+0.070/\beta), \quad (6)$$

was described empirically by

$$F = (1-5.2Ri)^2, \quad Ri \geq 0 \quad (7)$$

$$F = (1-16Ri)^{0.75}, \quad Ri < 0. \quad (8)$$

In Eq. (6) g is the acceleration due to gravity, T absolute temperature, θ potential temperature, β the Bowen ratio (sensible/latent heat flux). The term $1+0.070/\beta$ is an approximate correction for the influence of the buoyancy of moist air on Ri (Thom *et al.* 1975). In practice, small values of β over cereal crops in Britain are very seldom associated with strongly unstable or stable atmospheres (large $|Ri|$). Consequently, the errors in fluxes

incurred by ignoring this term seldom exceed 10%; in the following analysis the buoyancy of moist air was neglected. Over tall, aerodynamically rough crops such as forests there is evidence that the aerodynamic method described in Eqs. (5)–(8) systematically and substantially underestimates fluxes in unstable conditions and overestimates them in stable atmospheres (Thom *et al.* 1975). Over cereals in Britain there is reasonably good agreement between the aerodynamic method and independent methods of determining water vapour fluxes (Grant 1975) and CO₂ fluxes (Biscoe *et al.* 1975); in a recent detailed comparison of methods of flux determination over short grass in Canada, Saugier and Ripley (1978) showed that the aerodynamic method underestimated sensible heat fluxes by up to 20% in unstable conditions and overestimated them by up to 40% in moderately stable atmospheres. In the absence of unequivocal evidence from micro-meteorological experiments or theory, we have calculated fluxes on the basis of Eqs. (5)–(8), assuming one-dimensional transfer (Monteith 1973; Webb 1965).

(b) *Interpretation of fluxes – resistance analogues*

In a constant flux layer, integration of Eq. (1) with respect to z between heights z_1 and z_2 yields

$$F_s = \{\chi_s(z_2) - \chi_s(z_1)\} / \int_{z_1}^{z_2} K_s^{-1} dz, \quad (9)$$

a form analogous to Ohm's law: flux = potential difference/resistance (Monteith 1973; Thom 1975). In particular, if the lower limit of the integral is an imaginary surface in the crop where the SO₂ concentration is $\chi_s(0)$, Eq. (9) defines the aerodynamic resistance to turbulent transfer of SO₂ (Fig. 1). Within the canopy, Fig. 1 illustrates that there may be several sinks for SO₂, to be discussed later; assuming that SO₂ concentration within a sink

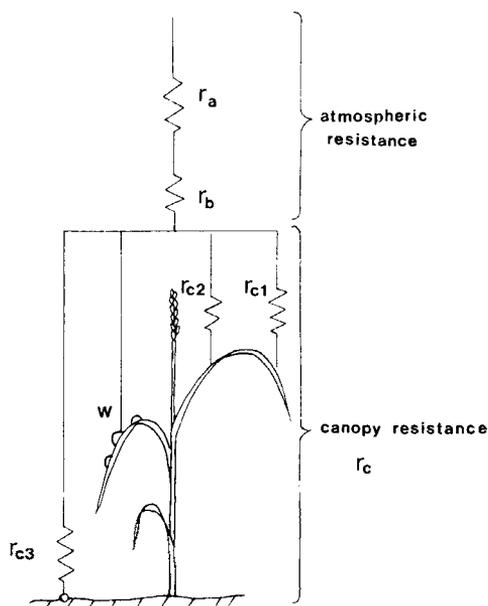


Figure 1. Resistance analogue of dry deposition of SO₂ to a wheat canopy, showing aerodynamic resistance, r_a , bluff-body resistance, r_b , and resistances in canopy, r_{c1} , to stomatal uptake, r_{c2} , to surface deposition, r_{c3} , to uptake by soil, and, w , to uptake by surface moisture.

is zero, the canopy resistance, r_c , is the resultant of all the resistances in parallel, and may be defined by

$$r_c = \{\chi_s(0) - 0\}/F_s. \quad (10)$$

Similarly, the total resistance to SO_2 transfer, r_t , may be defined by

$$r_t = \chi_s(z)/F_s. \quad (10a)$$

The reciprocal of r_t has dimensions of velocity and is often called the deposition velocity, v_g (Chamberlain 1955). However, the resistance analogy is more useful than v_g in analysing fluxes of pollutants when several sinks exert chemical and physiological control on fluxes.

The aerodynamic resistance to SO_2 transfer may be treated as the sum of two components in series (Fig. 1): the aerodynamic resistance to momentum transfer, r_a , and the bluff-body resistance, r_b (Chamberlain 1968; Thom 1975). The value of r_a is given (Monteith 1973) by

$$r_a = (ku_*)^{-1} \{\ln\{(z-d)/z_0\} + 5(z-d)/L\}, \quad (11)$$

where z_0 is the aerodynamic roughness length, L is the Monin-Obukhov length, and other symbols are as defined earlier. (Strictly Eq. (11) is valid only for small values of $(z-d)/L$, a condition satisfied in the present measurements; Thom and Oliver (1977) give an expression valid over a wider range.)

The resistance r_b arises because momentum transfer is more effective than mass transfer because of bluff-body forces. Chamberlain (1968) showed that for surfaces with bluff roughness elements

$$r_b = (Bu_*)^{-1} \quad (12)$$

and concluded (Chamberlain 1974) that for most crop canopies an appropriate value for B^{-1} was 7 for SO_2 .

Values of r_a and r_b may be calculated from wind and temperature profiles using Eqs. (11) and (12); values of r_t are calculated from Eq. (10a). If $r_t - r_a - r_b$ is not significantly different from zero, the surface is a perfect sink for SO_2 . However, for many surfaces a residual resistance $r_c = r_t - r_a - r_b$, exists, its value depending on the affinity of the surface for uptake of SO_2 . For simple surfaces such as water the value of r_c depends on the solubility of SO_2 in water and on subsequent chemical reactions between dissolved SO_2 and water, but for dry deposition of SO_2 on vegetation Fig. 1 shows that uptake may depend on diffusion through stomata (r_{c1}), reactions on leaf cuticle (r_{c2}), reactions with moisture on the crop (w) and transfer to soil (r_{c3}); thus r_c is the resultant of these resistances in parallel.

(c) Restrictions

In addition to the usual restrictions in flux-gradient analysis of fetch, uniformity of surface and averaging period, discussed by Webb (1965), there is an important restriction in the aerodynamic method for estimating fluxes of pollutant gases. Pollutant concentrations must be constant with time over the sampling period. This condition is not easily satisfied for pollutant gases since sampling must usually be done in a region where large ambient concentrations are common (i.e. near sources) to obtain the required precision in measurement. In these conditions errors may arise from storage or advection (Thom 1975). Unsworth and Fowler (1974) and Garland (1977) estimated the maximum errors in SO_2 flux measurements attributable to SO_2 concentrations varying with time. When the rate of change of SO_2 concentration is $20 \mu\text{g m}^{-3}\text{h}^{-1}$, typical of maximum values in Figs. 2-5, the error in SO_2 flux is about 5%, negligible in comparison with other sources of error.

3. SITE AND METHODS

In 1973 and 1974 measurements were made of fluxes of SO₂ to crops of winter wheat at Sutton Bonington in a predominantly agricultural area about 14 km SW of the city of Nottingham. Different sites, about 1.5 km apart, were used in the two years. Mean annual SO₂ concentration in this region is about 50 µg m⁻³. Sulphur dioxide concentrations were measured at 5 heights above the crop using apparatus described in detail by Garland (1974). The maximum measurement height was 2 m above the ground. Air was drawn at about 500 cm³s⁻¹ for periods of 60 to 120 minutes through heated filters which remove particles but not SO₂, then through glass bubblers containing a 1% hydrogen peroxide solution where SO₂ was trapped and oxidized to sulphate ions. The sulphate ion concentration in solution was determined by the method of Persson (1966). Air temperatures were measured at seven heights from 0.6 to 2.0 m above ground using a differential thermocouple system described by Biscoe *et al.* (1975). Windspeeds at six heights up to 2 m were measured with sensitive cup-anemometers mounted on horizontal arms on two separate masts to minimize mutual interference. The measurement programme was designed to determine SO₂ fluxes to the wheat crop in a wide range of atmospheric and surface conditions. In practice, 8 to 12 almost consecutive measurements (each taking 2 hours) were made over periods of 24 to 36 hours at various stages in the growth cycle of the crop.

The largest contribution to uncertainty in the SO₂ fluxes was usually the error in concentration gradient which ranged from 20 to 40% depending on concentration (Garland 1977). Biscoe *et al.* (1975) discussed uncertainties in wind and temperature profiles. Overall random errors in individual flux measurements may be up to 50% at times, but the consistency of sets of consecutive measurements (appendix 1) suggests that, in general, random errors were smaller than this. On the present evidence, unquantifiable systematic errors inherent in the aerodynamic method of flux determination are unlikely to be large for short crops in the British climate.

4. RESULTS

When the mechanism of SO₂ uptake at the surface is independent of SO₂ concentration then v_g is independent of concentration and, regarded as the flux per unit atmospheric concentration, it enables study of the effects of surface and atmospheric conditions on deposition rates without confounding effects of changes in SO₂ concentration. From a total of 84 estimates of v_g during 1973 and 1974 (tabulated in appendix 1) no relationship was found between v_g and SO₂ concentration in the range 10 to 150 µg m⁻³; thus v_g is an appropriate factor to describe SO₂ fluxes. Figs. 2 to 5 show changes in v_g and canopy resistance (r_c) with time on four separate occasions and demonstrate important factors determining fluxes. Each point represents a mean value for a two-hour sampling period, the mid-point of which is shown. Also shown are (i) windspeed (ii) SO₂ concentration 1 m above the zero plane (iii) the presence of dew from visual observation or from dew recorder measurements and (iv) whether or not the stomata of the crop were open.

(A). 8 to 9 May 1974 (Fig. 2). During this period of rapid crop growth, marked diurnal changes in deposition velocity and canopy resistance were observed. The soil water deficit was small, so that with solar irradiance exceeding 500 W m⁻² between 12 and 16 h (all times are GMT) on the 8th and 08 to 12 h on the 9th we assume that the stomata of the crop were open; during the hours of darkness the stomata would have been closed.

(B). 26 to 27 May 1974 (Fig. 3). The period from 11 h on the 26th until midnight shows variations in v_g and r_c similar to (A). Between 01 and 02 h on the 27th dew began to form and was widespread on the crop by 03 h. When dew occurred, v_g increased by a factor of

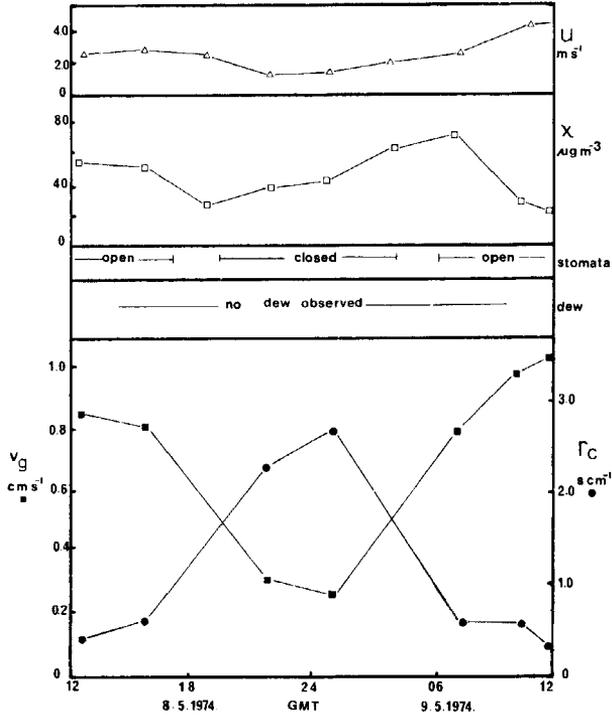


Figure 2. Diurnal variations of deposition velocity, v_g , canopy resistance, r_c , wind speed, u , and SO_2 concentration, χ . Durations of dew deposition and of estimated stomatal opening are also shown. All height-dependent parameters are referred to 1 m above the zero plane.

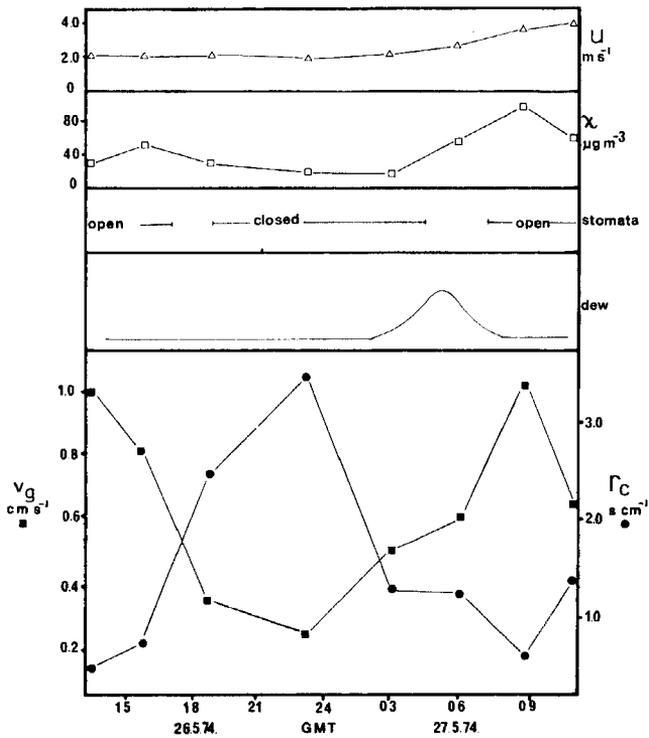


Figure 3. As Figure 2.

two. Between 10 and 16 h on the 26th and 18 and 13 h on the 27th solar irradiance exceeded 500 W m^{-2} ; with a soil water deficit of 50 mm, stomata were probably open.

(C). 18 to 19 June 1974 (Fig. 4). The figure shows the diurnal variation in v_g and r_c in the absence of dew. To obtain direct information about stomatal responses, stomatal resistances of individual leaves of the crop were measured during the period 06–09 h on the

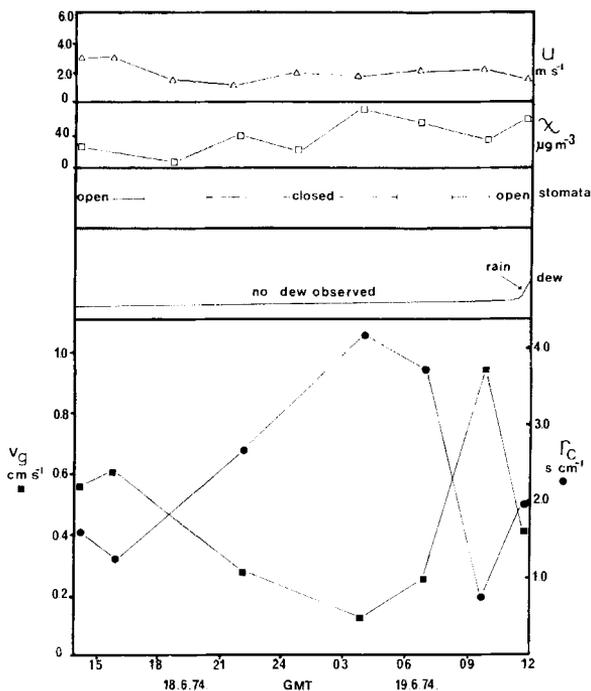


Figure 4. As Figure 2.

19th using a diffusion porometer. The period during which v_g increased from 0.1 to 0.9 cm s^{-1} coincided with the period during which stomata were opening. Over the 24 hours, wind-speed and SO₂ concentration both fluctuated about their means of 2 m s^{-1} and $35 \mu\text{g m}^{-3}$, respectively, but not in a manner consistent with changes in v_g or r_c .

(D). 24 to 25 July 1973 (Fig. 5). These measurements were obtained when the crop was senescent, with few green areas of foliage present. The stomata were therefore assumed to be closed (Meidner and Mansfield 1969). The diurnal change in v_g and r_c is in the opposite sense to that in Figs. 2 and 3. Dew was first observed on the crop soon after 18 h on the 24th and r_c decreased to zero. Uncertainties in the analysis of wind and temperature profiles for the runs at 19 and 22 h on the 24th, when Ri (calculated at 1 m above the zero plane) exceeded $+0.2$, make interpretation of changes in r_c and v_g uncertain but when canopy resistance increased from zero at midnight to 2.6 s cm^{-1} at 07 h, Ri was less than $+0.05$ so that this change may be interpreted with more confidence.

From these case studies, which are a small fraction of the data tabulated in appendix 1, it is clear that in quite similar atmospheric conditions (windspeed, SO₂ concentration and temperature gradient) the deposition velocity may vary between 0.1 and 1.5 cm s^{-1} . When leaf surfaces were dry, deposition velocities to the green crop with open stomata were generally a factor of four larger ($\sim 1.2 \text{ cm s}^{-1}$) than those either to the green crop at night (closed stomata) or to the senescent crop (both $\sim 0.3 \text{ cm s}^{-1}$). When there was dew on the crop, deposition velocities were generally large irrespective of the physiological condition of the vegetation.

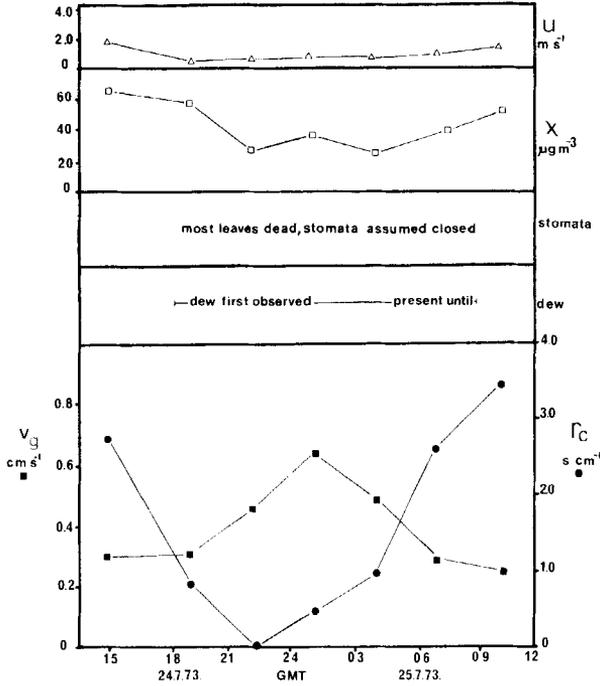


Figure 5. As Figure 2.

These generalizations indicate the dominance of surface conditions in controlling deposition. Thus, in Figs. 2 to 5, canopy resistance and deposition velocity are strongly inversely correlated.

Pooling data from 1973 and 1974 for the four-month period May–August, mean surface resistance was about 65% of mean total resistance ($r_t = 1/v_g$); as r_c was usually the major component of r_t the two are roughly linearly related (Fig. 6). Using the resistance analogue (Fig. 1) as a description of the deposition process the results may now be examined to evaluate component resistances and to assess their importance in a variety of conditions.

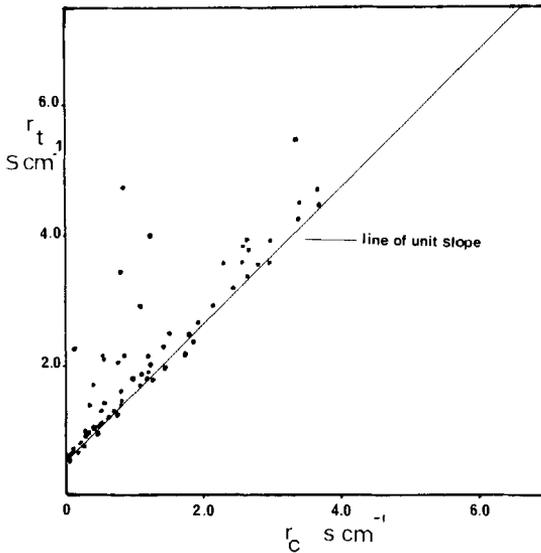


Figure 6. Variation of total resistance, r_t , to SO_2 transfer with canopy resistance, r_c . (Reference height is 1 m above zero plane.)

5. VARIATIONS IN RESISTANCES TO SO₂ DEPOSITION(a) *Dry canopy*

(i) *Aerodynamic resistances, $r_a + r_b$.* Values of the aerodynamic resistance $r_a + r_b$ were generally in the range 0.4–0.9 s cm⁻¹ when windspeeds at 1 m ranged from 5 to 1 m s⁻¹, and appendix 1 shows that $r_a + r_b$ was usually smaller than r_c apart from a few occasions at night. Consequently, there was usually little dependence of flux or v_g on windspeed.

(ii) *Canopy resistance, r_c .* The canopy resistance, r_c , may be regarded as three components acting in parallel (Fig. 1): resistance r_{c1} to uptake through stomata of all leaves in the canopy; r_{c2} to absorption on the surface of leaves and stems; and r_{c3} to absorption at the soil surface.

Soil surface component, r_{c3} . Shortly before harvest, all vegetation was yellow and senescent. The leaf area index had decreased from a maximum of 4.5 to 3.5 and soil was visible from above the crop. On such occasions, when leaves were dry, SO₂ gradients were below the detectable limit (0.5 μg m⁻³ m⁻¹) and so the canopy resistance was greater than 7 s cm⁻¹ (appendix 2). Thus the lower limit for the resistance r_{c3} at this time was 7 s cm⁻¹ assuming that the entire SO₂ flux was being absorbed by the soil (because the leaves had infinite resistance). It may be shown by taking typical values of atmospheric and surface resistances when the crop was green and putting $r_{c3} = 7$ s cm⁻¹, that fluxes of SO₂ to the soil during most of the season would not have exceeded 5 to 10% of the total flux. When leaves were dry and yellow during the two-week period preceding harvest, uptake by soil may have been the main deposition path.

Stomatal component, r_{c1} . Figs. 2 and 4 show that canopy resistance r_c varied diurnally with a minimum when stomata were likely to be open. The leaf surface absorption resistance, r_{c2} , will also vary diurnally as relative humidity changes within the canopy – Spedding (1969) showed a dependence of v_g to many surfaces on humidity – but r_{c2} would have its maximum value during the day. Consequently we conclude that r_{c1} was considerably smaller than r_{c2} during the day so that stomatal resistance determined the variation of canopy resistance.

Further evidence of the role of stomata in determining r_c was obtained using a porometer to measure stomatal resistance, r_s , to water vapour diffusion of leaves in the crop concurrently with SO₂ flux measurements. Measurements were made on the adaxial and abaxial surfaces of the tenth leaf from the ground on a number of plants. Fig. 7 shows the variations with time of the mean resistance of leaf 10 (obtained by combining resistances of upper and lower surfaces in parallel) and of r_c ; it is clear that variations of r_c and of r_s are positively correlated. A referee has pointed out that if the stomatal resistance of leaf 10 was representative of the mean stomatal resistance of leaves in the canopy (leaf area index \approx 4.5), it would be expected that $r_s/4.5 \approx r_c/1.8$, i.e. $r_c/r_s \approx 2.5$, where the factor 1.8 is the ratio of the diffusivity of water vapour in air to that for SO₂. For simultaneous points in Fig. 7, the mean value of r_c/r_s is about 2.7, supporting the contention that stomata were the main determinant of r_c . Such evidence is also consistent with the general observation that the proportion of total resistance attributable to canopy resistance increased with time throughout the season, because during senescence, stomata open for shorter periods and less widely than earlier in the season (Fig. 8).

For the green crop, minimum values of r_c during the day were between 0.5 and 1.0 s cm⁻¹, comparable with minimum canopy resistances for water vapour diffusion of an unstressed wheat crop in the same field in 1977 (Squire and Biscoe, personal communication). At night, in the absence of dew, r_c was generally about 3 s cm⁻¹, much smaller than typical night-time canopy resistance for water vapour transfer (Sestak *et al.* 1971). This suggests that r_{c2} , the dominant resistance at night, arises from mechanisms other than diffusion through stomata or cuticle. By assuming that at night r_{c2} was much less than r_{c1} it is possible

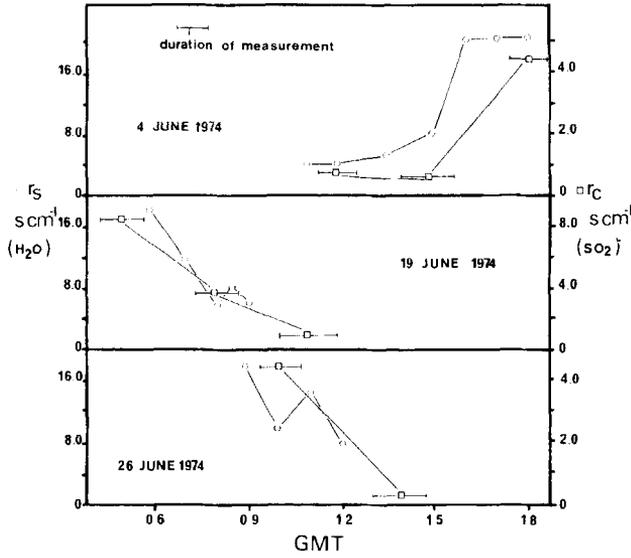


Figure 7. Diurnal variations of canopy resistance, r_c , to SO_2 transfer and of stomatal resistance, r_s , of single leaves in the canopy to water vapour diffusion, on three days.

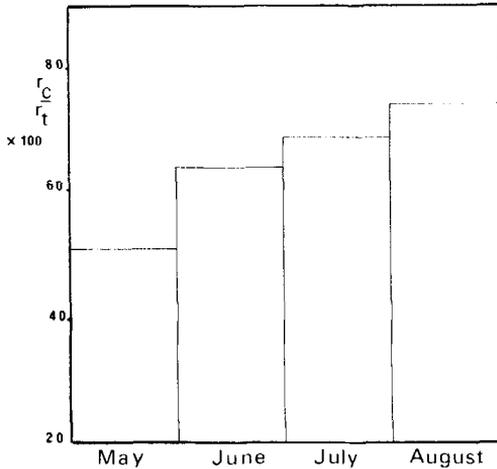


Figure 8. Monthly mean values of the percentage of the total resistance, r_t , to SO_2 transfer accounted for by canopy resistance, r_c .

to deduce mean values of r_{c2} and r_{c1} for the green canopy as follows. The mean of 24 estimates of canopy resistance, r_c , during the day (09–17 h) between 8 May and 30 June was $0.7 \pm 0.1 \text{ s cm}^{-1}$. During the same period the mean of 9 measurements of r_c when the canopy was dry at night (20–06 h) was $2.5 \pm 0.3 \text{ s cm}^{-1}$. Assuming that the latter value approximates r_{c2} and that the same value of r_{c2} is appropriate for daytime conditions it follows that $r_{c1} \approx 1.0 \text{ s cm}^{-1}$. If r_{c2} had doubled during the day as relative humidity in the canopy decreased, the estimate of r_{c1} would be 0.8 s cm^{-1} so that our conclusions are not very sensitive to assumptions about r_{c2} .

(b) *Wet canopy*

The influence of dew on canopy resistance may be dramatic, reducing its value in some cases to zero (Fig. 5). For many nights with clear skies and dew, windspeeds are small and extreme stability in the lowest few metres of the atmosphere is common (Ri is a stronger function of windspeed than of temperature (Eq. (6)). In such conditions turbulent transfer is strongly damped and local topographic effects may influence windspeeds. These effects, combined with stalling anemometers, preclude reliable flux measurements by the aerodynamic method (the night of 4/5 June 1974 is an example). However, on the evening of 26 May windspeed (at 2 m) did not fall below 1.8 m s^{-1} and Ri did not exceed $+0.05$, and thus the large changes in r_c are unlikely to be artefacts of the analysis. On the night of 24/25 July 1973 (Fig. 5) the runs at 17 and 21 h were in very stable conditions but all subsequent runs were made in suitable conditions for confident estimates of r_c , which increased from about 0.5 s cm^{-1} at midnight to 2.5 s cm^{-1} at 07 h in the presence of dew. Junge and Ryan (1958), Van den Heuvel and Mason (1963) and Brimblecome and Spedding (1972) showed that rates of uptake of SO₂ by water become limited by liquid phase resistances, w (Fig. 1), when the liquid pH falls to about 3.0. From the measurements on 24–25 July we may estimate as follows the pH of the dew film when canopy resistance begins to increase. Typical rates of dew formation are 20 to $30 \text{ g m}^{-2} \text{ h}^{-1}$ (Monteith 1957). On 24 July the period of dew formation was about three hours, because complete cloud cover developed by midnight. Taking the measured SO₂ flux at 18 h on the 24th of $0.2 \mu\text{g m}^{-2} \text{ s}^{-1}$ and assuming that 60 g m^{-2} of dew accumulated in the three cloudless hours and none thereafter, and that no neutralizing ions were present in solution, the pH of dew was about 3.0 by midnight. Thus changes of canopy resistance during the night of 24/25 July were consistent with the absorption of SO₂ in the dew short-circuiting other resistances in the canopy until the pH of the dew fell to about 3.0, whereupon resistance of the dew layer increased.

On many other occasions dew formation continued for 6 to 9 hours and because vertical mixing was suppressed, SO₂ concentrations above the crop decreased (Fig. 9). When there is continuing dewfall and SO₂ concentrations near the ground are small, sufficient acidification of the dew to prevent 'perfect sink' behaviour of the surface may be infrequent. In both seasons, when the crop was senescent, appendix 1 shows that measurable values of v_g were obtained only when dew was likely.

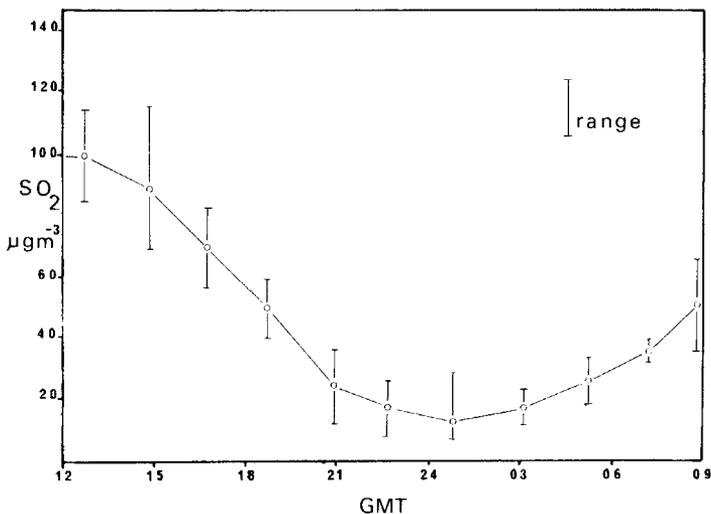


Figure 9. Mean diurnal variation of measured SO₂ concentration at 2 m over wheat on three days in 1973 when there was dew deposition on the crop. The ranges of the measurements are shown.

TABLE 1. PREDICTED AND MEASURED DEPOSITION VELOCITY, v_d , FOR SO₂ TO GRASS. A REFERENCE HEIGHT OF 1 m AT WHICH WINDSPEED IS 2.5 m s⁻¹ IS ASSUMED

Surface	Season	Height h (cm)	Assumed parameters		Surface resistance (s cm ⁻¹)	Calculated aerodynamic resistance (r_a+r_b)(s cm ⁻¹)	Deposition velocity		Ref.
			Roughness length z_0 (cm)	Zero plane displacement d (cm)			Calculated	Measured	
Short grass	Spring, early summer	1.5	0.2	1.0	0.70	1.3	0.50	0.55	(1)
Grass	Summer	13	1.7	9.0	0.70	0.65	0.74	0.80	(2)
Moorland	Summer	13	1.7	9.0	0.70	0.65	0.74	0.70	(3)
Grass	Autumn	13	1.7	9.0	2.5	0.65	0.32	0.32	(4)
Grass and bare soil	Autumn, winter	13	1.7	9.0	2.5	0.65	0.32	0.85	(5)

(1) Garland *et al.* (1974). Mean of 14 measurements.

(2) Shepherd (1974). Mean of 16 measurements.

(3) Holland *et al.* (1974). Mean of 28 measurements over moorland, principally cotton sedge.

(4) Shepherd (1974). Mean of 5 measurements in late October.

(5) Garland *et al.* (1973). Mean of 15 measurements over sparse short grass covering calcareous soil. The discrepancy between calculation and measurement may be because the soil was a good sink for SO₂.

6. COMPARISONS WITH OTHER STUDIES

There are several published measurements of SO₂ fluxes to grass, and as grasses and cereals are similar in leaf structure and physiology we may modify the resistance model for fluxes to wheat to make estimates of fluxes to grass.

For example Garland *et al.* (1974) reported measurements by day in spring over short grass (height $h \approx 1.5$ cm). Assuming roughness length $z_0 = 0.13h$, $d = 0.7h$ (Monteith 1973), a typical windspeed of 2.5 m s^{-1} , and neutral stability, integration of Eq. (2) and use of Eqs. (11) and (12) yield $r_a + r_b = 1.3 \text{ s cm}^{-1}$. Assuming that $r_c = 0.7 \text{ s cm}^{-1}$ (section 5) gives total resistance $r_t = 2.0 \text{ s cm}^{-1}$, i.e. $v_g = 0.5 \text{ cm s}^{-1}$. From 14 measurements of v_g Garland *et al.* reported a mean value of 0.55 cm s^{-1} , close to the estimated value. Table 1 summarizes this example and gives similar comparisons with other measurements.

The good agreement between prediction and measurement in Table 1 shows that this approach may be used to estimate rates of deposition on regions where grass and cereal surfaces predominate. The results of such an approach (Fowler 1978) are consistent with estimates of dry deposition using airborne sampling (Smith and Jeffrey 1975) and numerical modelling (Fisher 1975).

7. INTEGRATION: SEASONAL UPTAKE OF SULPHUR BY THE CROP

A knowledge of the components of canopy resistance enables fluxes of SO₂ onto leaf and stem surfaces and into stomata to be estimated if mean stomatal responses to environmental factors are known. When water is not limiting, light is the main factor influencing stomata in the field. Squire and Biscoe (personal communication) recently made a detailed study of stomatal and canopy resistances for water vapour transfer from a wheat crop at Sutton Bonington. Over the period May–July (when 80% of the dry matter of the crop accumulated) the mean daily canopy resistance to water vapour transfer, based on mean irradiance and on Squire and Biscoe's measurements, was about 0.8 s cm^{-1} . Since the ratio of diffusivities for SO₂ and water vapour is 1/1.8, the appropriate value of r_{c1} is 1.4 s cm^{-1} . We assume a mean value of r_{c2} of 2.5 s cm^{-1} and ignore r_{c3} during this period. Mean windspeed was 1.5 m s^{-1} and so $r_a + r_b = 0.5 \text{ s cm}^{-1}$. Concentrations of SO₂ averaged $50 \mu\text{g m}^{-3}$. Substituting the values of resistances in the network of Fig. 1 enables sulphur deposition on leaf surfaces and uptake through stomata to be estimated independently. Table 2 summarizes the results of this exercise and shows that about 30% of the sulphur content of the crop at harvest was probably supplied by absorption of SO₂ through stomata. The quantity of sulphur deposited on leaf surfaces was probably underestimated because the influence of dew and rain-wetted surfaces was ignored. When Raybould *et al.* (1977) allowed for the high deposition velocity of SO₂ to wet surfaces, their estimate of sulphur deposition on leaf surfaces during the growing season was doubled.

TABLE 2. ESTIMATIONS OF UPTAKE OF SULPHUR AS SO₂ BY A WHEAT CROP OVER THE PERIOD 1 MAY–31 JULY ASSUMING A MEAN SO₂ CONCENTRATION OF $50 \mu\text{g m}^{-3}$ AND MEAN WINDSPEED 1.5 m s^{-1} AT 1 m ABOVE THE ZERO PLANE. DEPOSITION TO SOIL IS NEGLECTED

	Aerodynamic	Resistance (s cm^{-1})			Sulphur deposition (g sulphur m^{-2})		
		$r_a + r_b$	Stomatal	Cuticular	Total	Cuticular	Stomatal
Day (12h)	0.5	r_{c1}	r_{c2}	r_t			
Night	0.5	1.4	2.5	1.4	0.26	0.46	0.72
		$\sim \infty$	2.5	3.0	0.33	0	0.33
				Total	0.59	0.46	1.05

Sulphur content of crop at harvest = $1.46 \text{ g sulphur m}^{-2}$.

Percentage atmospheric sulphur entering vegetation through stomata = $0.46 \times 100 / 1.46 = 32\%$.

8. CONCLUSIONS

Analysis of micrometeorological measurements reveals the factors controlling deposition rates for SO_2 . Deposition velocity, commonly used to describe fluxes of pollutants, is shown to depend primarily on surface affinity for SO_2 so that v_g may vary by a factor of five over short periods. During the day, stomata are an important sink for atmospheric SO_2 which may have supplied 30% of the sulphur in our crops at harvest.

When leaves are wet with dew, rates of SO_2 uptake from the atmosphere depend initially only on atmospheric concentration and on aerodynamic resistance. If canopy resistance is also small during and after rain, when aerodynamic resistance is often smaller than on nights with dew, fluxes of sulphur to rain-wetted vegetation may be substantial.

Sulphur dioxide is also deposited on leaf surfaces of the crop, a path available by night and day. Consequently, more sulphur may be deposited on external surfaces than inside the crop (Table 2).

Regional models of dry deposition could usefully include resistance analogues of surface processes to allow for effects of the weather on surface affinity for SO_2 . Local and short-term studies, especially where effects or subsequent fate of deposited sulphur are important, should find the resistance formulation more useful than deposition velocity in parametrizing surface fluxes.

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APPENDIX 1

Results of 84 measurements of SO₂ fluxes to wheat when SO₂ profiles were within limits of resolution. Wind speed, u , SO₂ concentration, χ , and deposition velocity, v_g , are referred to 1 m above zero plane. Symbols are as given in the text.

Date	Time (GMT)	u (ms ⁻¹)	χ ($\mu\text{g m}^{-3}$)	ϕ_H	u_* (ms ⁻¹)	r_a (scm ⁻¹)	r_b (scm ⁻¹)	SO ₂ flux (ng m ⁻² s ⁻¹)	v_g (cm s ⁻¹)	r_c (scm ⁻¹)
8.vi.73	1100	1.3	152.1	0.73	0.23	0.24	0.25	640	0.42	1.88
10.vi	1100	2.2	81.8	0.95	0.34	0.19	0.21	260	0.32	2.71
10.vi	1500	2.8	29.9	0.98	0.47	0.13	0.17	90	0.30	3.00
24.vi	1100	1.6	20.9	0.71	0.32	0.15	0.20	230	1.10	0.56
5.vii	1800	0.8	11.0	3.00	0.08	1.26	0.63	7	0.06	14.27
6.vii	0600	0.5	22.9	1.00	0.08	0.87	0.55	40	0.17	4.50
18.vii	1900	1.1	22.4	4.50	0.09	1.22	0.56	40	0.19	3.37
24.vii	1440	1.9	65.0	0.84	0.27	0.26	0.24	200	0.31	2.77
24.vii	1800	0.4	58.0	1.64	0.05	1.45	0.89	180	0.31	0.90
24.vii	2100	0.4	25.8	1.72	0.04	2.24	0.98	120	0.46	0.0
25.vii	0050	0.8	35.4	1.35	0.11	0.65	0.48	220	0.63	0.46
25.vii	0300	0.6	23.9	1.45	0.10	0.57	0.51	118	0.49	0.95
25.vii	0600	0.9	36.2	1.00	0.13	0.57	0.40	100	0.28	2.62
25.vii	0900	1.3	52.3	0.57	0.23	0.25	0.24	130	0.26	3.41
29.vii	1000	1.3	76.9	0.66	0.25	0.21	0.23	180	0.24	3.69
29.vii	1710	2.1	71.5	0.96	0.30	0.24	0.23	400	0.55	1.34
30.vii	0200	0.8	13.0	1.00	0.12	0.53	0.42	140	1.10	0.0
30.vii	0505	0.4	23.4	1.00	0.08	0.62	0.54	110	0.49	0.88
30.vii	0905	1.0	82.3	0.50	0.22	0.20	0.23	970	1.18	0.41
2.viii	0900	1.9	124.8	0.65	0.31	0.19	0.20	2090	1.68	0.20
2.viii	1603	2.1	102.0	0.92	0.31	0.22	0.22	760	0.74	0.90
2.viii	2005	1.3	16.6	1.06	0.22	0.27	0.28	160	0.99	0.45
3.viii	0907	1.6	61.3	1.00	0.25	0.25	0.26	420	0.69	0.94
7.viii	1900	2.5	70.8	1.07	0.30	0.27	0.23	270	0.38	2.17
7.viii	2310	2.7	23.3	1.02	0.32	0.26	0.20	130	0.56	1.33
8.viii	0205	2.0	20.2	1.12	0.23	0.38	0.28	89	0.44	1.60
8.viii	0500	1.8	56.7	1.00	0.23	0.33	0.27	175	0.31	2.63
12.viii	0400	0.4	34.7	1.22	0.06	1.22	0.70	164	0.47	0.19
12.viii	0700	2.1	36.3	0.86	0.33	0.19	0.21	285	0.78	0.87
8.v.74	1200	2.4	58.4	0.87	0.22	0.50	0.29	497	0.85	0.38
8.v	1500	2.7	52.7	1.00	0.26	0.39	0.25	432	0.82	0.58
8.v	2100	1.5	38.1	1.35	0.14	0.61	0.35	117	0.31	2.29
8.v	0000	1.5	46.7	1.35	0.13	0.70	0.37	124	0.27	2.68
9.v	0600	2.6	74.5	1.00	0.25	0.43	0.26	588	0.79	0.57
9.v	0900	4.6	29.3	1.00	0.43	0.25	0.18	290	0.99	0.57
9.v	1200	4.8	14.9	1.00	0.45	0.24	0.17	230	1.55	0.24
15.v	1130	3.4	23.2	0.87	0.37	0.26	0.20	397	1.71	0.22
18.v	1230	0.9	21.6	0.87	0.06	2.80	0.68	49	0.23	0.92
18.v	1530	1.0	38.7	1.00	0.07	2.00	0.59	100	0.25	1.28
22.v	0815	1.8	64.6	0.88	0.22	0.34	0.27	180	0.28	3.00
22.v	1100	4.8	56.1	0.95	0.56	0.15	0.15	702	1.25	0.50
22.v	1400	3.4	33.0	0.94	0.40	0.21	0.19	146	0.44	1.80
26.v	0940	2.0	31.1	0.70	0.27	0.25	0.23	456	1.46	0.20
26.v	1200	2.1	34.6	0.75	0.27	0.25	0.23	388	1.12	0.41
26.v	1500	2.1	48.1	0.77	0.27	0.25	0.24	385	0.80	0.76
26.v	1807	2.4	33.9	2.02	0.31	0.24	0.23	115	0.34	2.48
26.v	2240	1.8	16.1	1.31	0.17	0.50	0.31	38	0.24	3.43
27.v	0205	2.0	18.1	1.24	0.20	0.45	0.29	91	0.50	1.25
27.v	0507	2.6	54.9	1.00	0.33	0.24	0.21	326	0.59	1.23
27.v	0900	3.4	103.3	0.91	0.44	0.18	0.17	1080	1.05	0.60
27.v	1200	4.0	61.1	0.94	0.51	0.15	0.16	337	0.55	1.51
29.v	0500	1.8	51.8	0.87	0.23	0.32	0.27	856	1.65	0.01
29.v	0700	3.6	26.9	0.94	0.47	0.16	0.17	591	2.20	0.13
29.v	0900	3.7	25.2	0.91	0.49	0.15	0.16	777	3.08	0.01
30.v	1300	2.6	35.4	0.86	0.34	0.22	0.21	299	0.84	0.76
30.v	1600	3.1	24.0	1.02	0.38	0.21	0.19	214	0.89	0.82

Date	Time (GMT)	u (ms ⁻¹)	χ ($\mu\text{g m}^{-3}$)	ϕ_H	u_* (ms ⁻¹)	r_a (scm ⁻¹)	r_b (scm ⁻¹)	SO ₂ flux (ng m ⁻² s ⁻¹)	v_g (cm s ⁻¹)	r_c (scm ⁻¹)
30.v	1840	2.3	25.6	1.19	0.25	0.35	0.25	143	0.56	1.19
4.vi	1200	2.1	95.4	0.75	0.32	0.19	0.21	790	0.84	0.80
4.vi	1500	1.7	92.3	0.81	0.26	0.24	0.25	870	0.91	0.56
5.vi	0600	1.2	62.2	0.66	0.20	0.28	0.29	370	0.60	1.11
13.vi	1900	1.5	20.3	2.10	0.18	0.53	0.33	117	0.58	0.87
14.vi	0405	0.8	25.2	3.00	0.09	1.00	0.50	300	0.36	1.20
14.vi	0700	1.8	114.7	0.82	0.25	0.26	0.25	1500	1.32	0.25
14.vi	1000	2.7	52.5	0.87	0.37	0.19	0.19	668	1.27	0.41
18.vi	1315	3.0	31.5	0.91	0.43	0.15	0.18	163	0.52	1.61
18.vi	1600	2.9	20.6	0.94	0.40	0.18	0.19	126	0.61	1.27
18.vi	2200	1.2	38.1	1.38	0.13	0.47	0.35	109	0.29	2.68
19.vi	0700	2.2	53.2	1.03	0.33	0.20	0.21	129	0.24	3.71
19.vi	1000	2.3	36.0	1.06	0.36	0.18	0.20	340	0.94	0.74
19.vi	1255	1.2	57.9	1.00	0.15	0.52	0.36	214	0.37	1.82
20.vi	2200	0.9	14.4	1.29	0.18	0.23	0.30	58	0.41	1.97
21.vi	0100	0.7	20.2	1.29	0.10	0.29	0.34	94	0.46	1.52
21.vi	1000	2.2	26.1	0.95	0.40	0.14	0.19	285	1.09	0.59
26.vi	1300	3.2	13.7	0.87	0.54	0.12	0.16	468	3.40	0.02
8.vii	1900	1.8	23.3	1.09	0.24	0.31	0.26	450	1.94	0.00
9.vii	1100	4.2	54.8	0.98	0.62	0.11	0.14	1190	3.50	0.03
11.vii	1207	2.6	48.9	0.96	0.39	0.17	0.19	599	1.22	0.46
11.vii	2200	2.0	83.8	1.10	0.29	0.24	0.23	890	1.06	0.47
12.vii	0705	2.1	86.4	1.00	0.32	0.21	0.22	957	1.10	0.47
12.vii	1000	2.2	68.2	0.86	0.33	0.20	0.21	881	1.20	0.34
22.vii	2105	2.4	31.1	1.00	0.31	0.23	0.22	540	1.70	0.11
23.vii	0800	2.5	52.0	0.91	0.34	0.20	0.21	590	1.10	0.47
3.viii	1105	1.3	95.2	0.72	0.13	0.73	0.40	690	0.73	0.24
13.viii	1000	1.4	16.2	1.00	0.19	0.38	0.31	260	1.63	0.69

APPENDIX 2

Results of six measurements when the vertical gradient of SO₂ concentration was smaller than the resolution of the chemical analysis ($0.5 \mu\text{g m}^{-3}\text{m}^{-1}$). Upper limits of SO₂ flux densities are based on an assumed gradient of $0.5 \mu\text{g m}^{-3}\text{m}^{-1}$. Symbols are as given in the text.

Date (Aug. 1973)	Time (GMT)	u (ms ⁻¹)	χ ($\mu\text{g m}^{-3}$)	u_* (ms ⁻¹)	r_a+r_b (scm ⁻¹)	Maximum SO ₂ flux (ng m ⁻² s ⁻¹)	Maximum v_g (cm s ⁻¹)	Minimum r_c (scm ⁻¹)
7	1020	4.5	48.5	0.27	0.70	60	0.11	9.0
7	1300	5.3	45.6	0.31	0.70	60	0.14	7.2
7	1600	4.4	37.2	0.26	0.75	50	0.14	7.0
8	0900	2.5	41.0	0.16	1.20	30	0.08	12.5
8	1200	2.6	35.0	0.16	1.25	30	0.09	10.7
11	1000	1.9	110.0	0.12	1.60	20	0.02	45