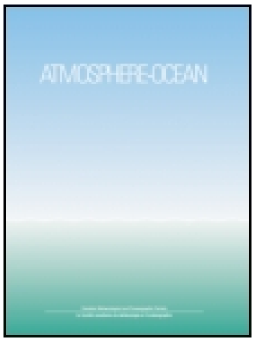

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Using measured variances to compute surface fluxes and dry deposition velocities: A comparison with measurements from three surface types

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Using Measured Variances to Compute Surface Fluxes and Dry Deposition Velocities: A Comparison With Measurements From Three Surface Types

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ABSTRACT *Fluxes of temperature, water vapour,* O_3, SO_2 *and* CO_2 *were estimated from the measurement of their variances, taken over a wetland region in northern Ontario (Canada) during the summer of 1990 and over a deciduous forest when it was fully leafed during the summer of 1988 and when it was leafless during the winter of 1990. A set of flux-variance relations was employed, including empirical forms of universal functions that could be adjusted with some constants. Results from the present study show that these constants needed to be adjusted with site-specific data in order to achieve a closer agreement between estimated and observed fluxes. Best estimates were obtained for the fluxes of temperature and water vapour and it was found that the flux estimates of* O_3, SO_2 *and* CO_2 *correlated better with water vapour than with temperature. For these trace gases the flux-variance method yielded estimates of dry deposition velocities that were either comparable with or larger than those obtained from a resistance analogue model. Both methods yielded values that overestimated the observed dry deposition velocities. The employment of the flux-variance method in an operational network would require the use of fast-response sensors and a practical method for reducing the noise level of the measured variances.*

RÉSUMÉ *On a estimé, à partir de la mesure de leurs variances, les flux de température, vapeur d'eau,* O_3, SO_2 *et* CO_2 *au-dessus d'une région de terres humides du nord de l'Ontario à l'été 1990, et au-dessus d'une forêt caduque mature durant l'été de 1988 et, sans feuilles, durant l'hiver 1990. On a utilisé un ensemble de rapports flux-variances en incluant des formes empiriques de fonctions universelles qui peuvent être ajustées à l'aide de constantes. Les résultats de l'étude montrent que ces constantes doivent être ajustées en utilisant des données spécifiques à la location afin d'avoir un accord qui approche les flux estimés et observés. Les flux de température et de vapeur d'eau ont produit les meilleurs estimés; les flux estimés de* O_3, *de* SO_2 *et de* CO_2 *ont une meilleure corrélation avec la vapeur d'eau qu'avec la température. Pour ces gaz trace, la méthode de variance des flux a produit des estimés de la velocité des dépositions sèches qui étaient soit comparable à ou plus grands que ceux obtenus d'un modèle analogue de la résistance. Les deux méthodes ont produit*

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des valeurs qui surestimaient les vélocités de déposition sèche. L'usage de la méthode de variance des flux dans un contexte opérationnel nécessiterait l'utilisation de capteurs à réponse rapide et un moyen pratique de réduite le niveau de bruit des variances mesurées.

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1 Introduction

A number of studies have shown possibilities for inferring dry deposition rates (or surface fluxes) from routinely measured meteorological and air quality data. Hicks et al. (1980) summarized a variety of such methods and ranked them. Highest among these was a method referred to as "indirect calculation of dry deposition rates", a term that was used to refer to the calculations of dry deposition velocity at a site, using the resistance analogue method (Hicks et al., 1987). This method has the advantage that it can employ easily measured atmospheric variables such as temperature and wind but suffers from the disadvantage that it requires information about surface characteristics that are not readily available. Numerical models have benefited from this approach (Walcek et al., 1986; Chang et al., 1987; Venkatram et al., 1988; Wesely and Lesht, 1989; Padro et al., 1991); now serious attempts are being made to employ it in a monitoring network (Hales et al., 1987; Meyers et al., 1991). Another method that was ranked high by Hicks et al. (1980) for its research and monitoring potentials is that of estimating fluxes (or deposition rates) from measurements of variances of scalar parameters. This method has the advantage that it does not require any information about surface characteristics. Its disadvantage, however, is that in order to measure fluctuations of scalar variables, it requires accurate fast-response sensors, which may not be practical in operational networks. Kanemasu et al. (1979) and Wesely (1983) describe some of the features of this method. More recently, Wesely (1988) investigated three methods for estimating fluxes from variance measurements and concluded that they are reliable for ozone, sulphur dioxide, temperature and water vapour, but are not practical for routine measurements because of the requirement for fast-response sensors. Weaver (1990) used a slightly different approach but limited his study to the computation of fluxes of temperature and water vapour using variances that were measured over grass, shrubs, bare soil and 1-m hummocks. He concluded that in general the flux estimates were reasonably accurate even over non-homogeneous surfaces and that the method could be used operationally if certain constants are adjusted with one-dimensional eddy correlation measurements of fluxes that are not expensive to obtain. De Bruin et al. (1991) employed a narrower scope of the variance method and limited the study only to temperature fluxes, yielding estimates that were in excellent agreement with the observations for thermally uniform terrain, but were larger than the observations for non-uniform terrain. Similarly, Lloyd et al. (1991), using the same method, obtained estimates of temperature fluxes that were in excellent agreement with observations that were collected over four different land use types.

Results from the above studies indicate the need to test the flux-variance relations with more observations of trace gases. In the present study we test these relations with large sets of data for temperature, water vapour, O_3, SO_2 and CO_2 over a deciduous forest when it was fully leafed and when it was leafless, and over a northern Ontario wetland region. We investigate the local dependence of some of the constants employed in the empirical universal functions of the flux-variance relations. Our primary objective is to test the quality of the estimated fluxes for computing dry deposition velocities. These estimates will be compared with those obtained from resistance analogue models.

2 Data

A variety of air quality and meteorological measurements were made during three separate field experiments over three different land use types. The first dataset (referred to as Borden 88) was collected during July and August of 1988 over a fully leafed deciduous forest located at the Canadian Forces Base Borden (44°19′N, 80°56′W). The types of variables that were measured are reported in Padro et al. (1991) and the methods of measurement are described in Shaw et al. (1988). Neumann et al. (1989) provide a description of the forest canopy, which consists primarily of mixed deciduous trees with an average height of 18 m. Measurements were made of concentrations (c), fluxes (F) and standard deviations (σ) of O_3 and CO_2. Other measurements included the sensible heat flux (H), latent heat flux (L_wE), momentum flux (for computing the friction velocity u_*), wind speed and direction, temperature (T or potential temperature θ), water vapour (q) and the standard deviations σ_T, σ_q, σ_w, σ_{O_3}, and σ_{CO_2}, where w denotes the vertical motion. The fluxes were measured using the eddy correlation technique. The second dataset (referred to as Borden 90) was also collected over the Borden forest but when the forest was leafless during the latter part of the winter of 1990. The same types of variables were measured as in the Borden 88 dataset except SO_2 instead of CO_2 measurements were made. This dataset is reported in Padro et al. (1992a, b), which also includes a description of the ground conditions that were sometimes dry, wet or snow covered.

The third dataset (referred to as Kinoje 90) was collected near Lake Kinoje (51°33′N, 81°49′W) in the Hudson Bay Lowlands, about 100 km west of Moosonee, Ontario, during the summer of 1990. The same types of measurements were made as in the Borden 88 dataset, except the surface was heterogeneous and consisted of wetland, dry areas, sphagnum, mosses, lichens, shrubs on hummocks, ponds and other land use types. More details, including methods of measurement, are reported in Chipanshi (1991).

The measurements were averaged over 30-min intervals. The data were quality controlled according to suggestions by Webb et al. (1980), Businger (1986) and Hicks et al. (1989). These led to datasets that were drastically smaller than the original ones. Figures 1 and 2 give an indication of the noise level in the measurements of the variances of the different variables. In Fig 1 the mean power spectra for selected half hours of temperature (nC_{TT}) and water vapour (nC_{qq}) indicate little noise, showing a $-5/3$ slope, with the exception of the night-time water vapour measurements of 9 April 1990 when the concentrations were very small. The frequency is denoted by n and the spectral coefficients of the variances are

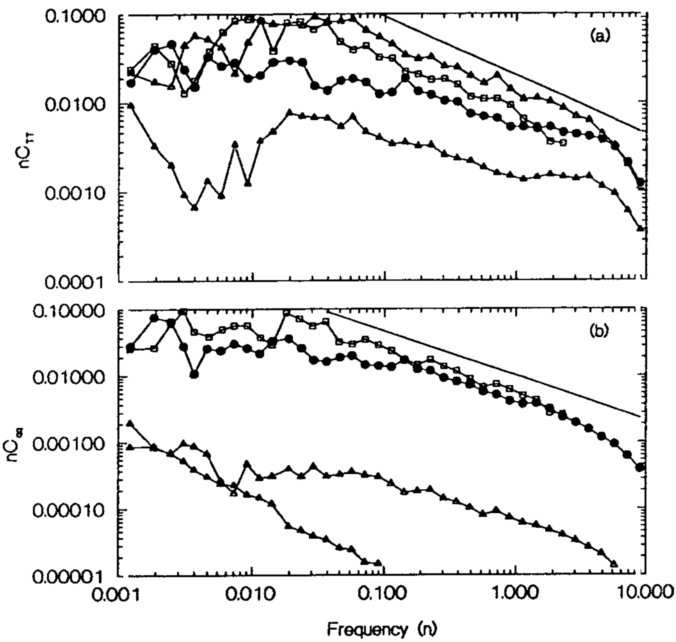

Fig. 1 Mean power spectra of (a) temperature and (b) water vapour for EDT periods: (i) 1200, 13 July 1990 (filled circles), (ii) 1300, 10 August 1988 (open squares), (iii) 1600, 8 April 1990 (open triangles), (iv) 0230, 9 April 1990 (filled triangles).

denoted by C_{TT} and C_{qq}. Similarly, the slope of the power spectra of carbon dioxide (nC_{CC}) show some agreement with the $-5/3$ slope (Fig. 2a). This is not true for the power spectra of ozone (nC_{OO}) and sulphur dioxide (nC_{SS}), which give a clear indication of white noise. The high-frequency ($n > 2$) roll-off was due to low-pass filters (nominal 10-Hz cutoff) on the instrumentation outputs. For carbon dioxide in 1988, the roll-off was observed at a lower frequency and reflects the response of the instrument itself as determined by sample cell volume and flow rate.

3 Theory

a *A Discussion of the Flux-Variance Equations*

A consistent set of equations relating the fluxes of scalar variables to their variances has been summarized by Wesely (1988). The equations can be derived from the Monin-Obukhov similarity theory (Monin and Obukhov, 1954; Panofsky and Dutton, 1984). Their theory states that the non-dimensional standard deviation of a scalar variable can be expressed as a universal function $\phi(Z/L)$ of the Monin-Obukhov stability parameter (Z/L). Attempts to derive these universal functions empirically from field measurements were made by Wyngaard et al. (1971) and Hicks (1981). Hicks (1981) and Weaver (1990) have shown that certain constants, which form part of the empirical functions, may assume different values depending upon the datasets from which they are computed. Thus, in practice, certain non-dimensional functions may not be as universal as expected from the ideal sim-

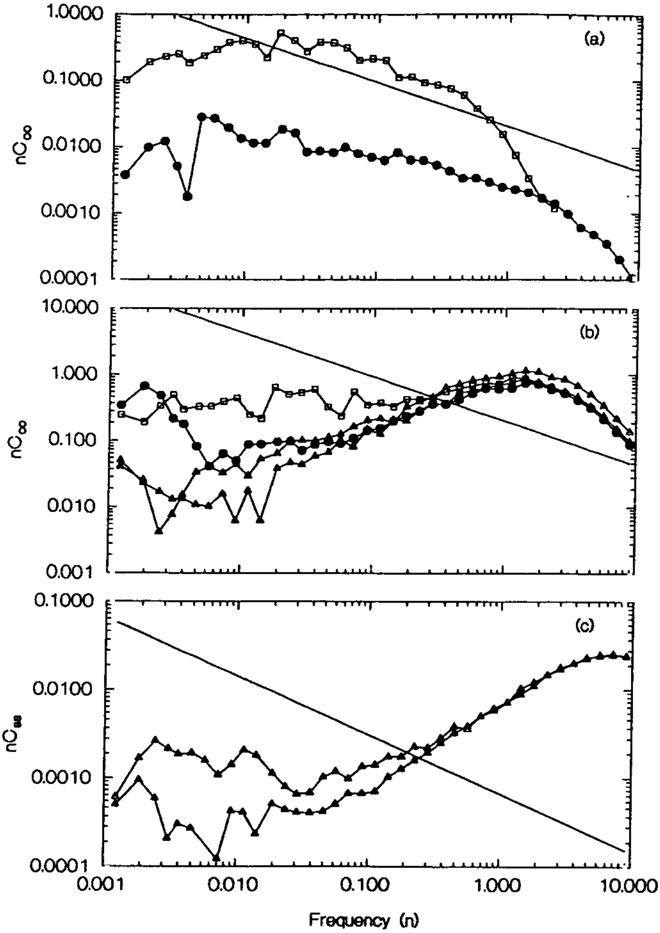

Fig. 2 Mean power spectra of the concentrations of (a) carbon dioxide, (b) ozone and (c) sulphur dioxide for the same EDT periods as in Fig. 1.

ilarity theory. Instead, they may vary from site to site and from one scalar variable to another. In the present study, we compute a variety of values for these constants by adjusting them with the observed fluxes, using Weaver's (1990) method. After including the constants in the universal functions, we solve the flux-variance equations for fluxes and compare them with observations. If we denote a scalar variable by s and rearrange some of the equations that appear in Wesely (1988), we obtain the following set:

$$|F_c| = |F_s| \frac{\sigma_c}{\sigma_s} \tag{1}$$

$$\frac{\sigma_s u_*}{|F_s|} = \phi(Z/L) \tag{2}$$

$$\begin{aligned}\phi(Z/L) &= a_1, \qquad Z/L \geqslant -(a_2/a_1)^3 \\ &= a_2(-Z/L)^{-1/3}, \qquad Z/L < -(a_2/a_1)^3\end{aligned} \tag{3}$$

$$L = \frac{-\rho c_p u_*^3 \theta}{\kappa g(H + L_w E/14)} \tag{4}$$

and u_* can be computed from the following empirical relation (Hicks, 1981):

$$\begin{aligned}u_* &= \sigma_w/1.3, \qquad Z/L > 0 \\ &= \sigma_w/[1.3(1-2.0Z/L)^{1/3}], \qquad Z/L < 0\end{aligned} \tag{5}$$

$$F_T = \frac{H}{\rho c_p}, \qquad F_q = E \tag{6}$$

where ρ is the air density, c_p is the specific heat of air at constant pressure, $\kappa(= 0.4)$ is the von Karman constant, g is the gravitational acceleration and a_1 and a_2 are constants associated with the universal function ϕ that will be determined using the method of Weaver (1990) and the existing datasets. Wesely (1988) selected $a_1 = 1.85$, a value that agrees with that of other studies (Wyngaard et al., 1971) and $a_2 = 1.25$, which differs from the more common value of 1.0 (Panofsky and Dutton, 1984) but agrees with the data of Wesely (1983), obtained over a different land use type. In (5), the values of 1.3 and 2.0 fit our datasets quite well, but they may not necessarily be ideal for all other datasets, as explained by Hicks (1981). In (3), the stability ranges of $(a_2/a_1)^3$ have been prescribed as in Weaver (1990).

We briefly discuss each of the above equations. The direction of the flux is not determined by this method and must be deduced by some other means. The basic assumption inherent in (1) can be understood from a simple definition of the linear correlation coefficient between the variables w and c (R_{wc}) and between w and s (R_{ws}). These yield

$$R_{wc} = \frac{\overline{w'c'}}{\sigma_w \sigma_c} = \frac{F_c}{\sigma_w \sigma_c} \tag{7}$$

$$R_{ws} = \frac{\overline{w's'}}{\sigma_w \sigma_s} = \frac{F_s}{\sigma_w \sigma_s} \tag{8}$$

where w', c' and s' denote deviations from the respective means of these variables. If we divide (7) by (8), we obtain

$$\frac{F_c}{F_s} = \frac{\sigma_c}{\sigma_s}\frac{R_{wc}}{R_{ws}}$$

If we make the assumption that c and s are transported by w in a similar manner, then $R_{wc} = R_{ws}$ and this explains the limitation of (1).

Equation (2) represents the non-dimensional standard deviation as a function of stability denoted by $\phi(Z/L)$ and it can be derived from similarity theory (Panofsky and Dutton, 1984). The form of the universal function $\phi(Z/L)$ is expressed in (3) and can be found in Wyngaard et al. (1971) and other studies. In the present study we demonstrate that the values of a_1 and a_2 need to be adjusted with local fluxes in order to achieve a better agreement between estimated and observed fluxes. Weaver (1990) made such adjustments in computing temperature and water vapour fluxes for stability ranges that varied in accordance with $(a_2/a_1)^3$. Theoretically, a_1 and a_2 are expected to be constant, provided the assumptions of the similarity theory are obeyed rigorously. This is seldom achieved in the real world owing to surface inhomogeneities, non-uniform sources and sinks and advection of turbulence from upstream sources.

Equation (4) is a well known definition of the Monin-Obukhov length L. It requires u_*, which can be computed from (5). Empirical forms for the stability-dependent function in (5) have been obtained by Merry and Panofsky (1976), Panofsky et al. (1977) and Hicks (1981). Hicks (1981) cautions against the use of (5) without ensuring that the values of 1.3 and 2.0 are in agreement with the datasets that are collected at specific sites. Alternatively, Weaver (1990) computed u_* from the measurements of wind speed and the application of the surface-layer wind equation.

b *Method of Computation*

We shall investigate (1) and (2) separately for computing fluxes. Equation (1) has the disadvantage that it requires the measurement of the flux of one variable to compute the flux of another. Equation (2) has the disadvantage that it requires prior knowledge of a_1 and a_2 and some means of computing L. For this reason we also solve the combined set of equations (1) to (6) for the fluxes, requiring knowledge of the variances only. All parameters, including L, a_1 and a_2 are solved simultaneously. The procedure for the solution requires an algebraic manipulation of (1)–(6) to yield the following expressions for L:

$$L = \frac{(\sigma_w/1.3)^2 \theta a_1}{\kappa g \left(\sigma_T + \dfrac{L_w}{14\rho c_p}\sigma_q\right)}, \quad \begin{array}{l}\text{stable case}\\ Z/L \geqslant -(a_2/a_1)^3 \text{ and } Z/L > 0\end{array} \tag{9}$$

and

$$L^2 - 2ZL - \left(\frac{Z^{1/3}(\sigma_w/1.3)^2 \theta a_2}{\kappa g \left(\sigma_T + \dfrac{L_w}{14\rho c_p}\sigma_q\right)}\right)^{3/2} = 0, \quad \begin{array}{l}\text{unstable case}\\ Z/L < -(a_2/a_1)^3 \text{ and } Z/L < 0\end{array} \tag{10}$$

A third expression for L, valid in the "near neutral" range, also exists. It has the form of a cubic polynomial and arises because of the possibility that part of the stability range in (5) may overlap part of the stability range in (3). We note that the solutions for L in (9) and (10) depend upon the input of parameters

$\theta, \sigma_T, \sigma_q, \sigma_w, a_1$ and a_2. Wesely assigned the value of 1.85 to a_1 and the value of 1.25 to a_2 and solved for the estimated fluxes using (1)–(6). He used the stability ranges $Z/L > -0.31$ and $Z/L < -0.31$, which agree with the computations of these ranges using $(a_2/a_1)^3$. Wesely (1988) used noise-filtered σs in (2) obtained by employing the relation $\sigma_c = \overline{c'T'}/\sigma_T$ where the temperature fluctuations T' are assumed to have a large signal-to-noise ratio in contrast to that of the concentration fluctuations c'. Alternatively, Weaver (1990) treated a_1 and a_2 as variables that were adjusted with the flux datasets. The values of a_1 and a_2 adjusted the empirical fit of $\phi(Z/L)$ to the observed local flux. Initial guesses were made for a_1 and a_2 to compute the initial fluxes (Eq. (2)), which were then regressed linearly against the measured fluxes from each of the datasets. If these initial fluxes appear to be poor estimates due to inadequate a_1 and a_2 values and large noise in the measurements of the scalar variable, then multiplying them by the regression slope would tend to yield improved fluxes. This process is repeated until successive values of a_1 and a_2 differ from previous ones by an arbitrarily small amount. The final values of a_1 and a_2 lead to the computation of the best estimates of the fluxes. In this sense, although the flux estimates are improved by the choices of a_1 and a_2, the noise in the σs is unchanged, unlike Wesely's (1988) method. It is noted that for each solution of a_1 and a_2, there is a new solution for L, thus yielding stability ranges in (3) that depend upon a_1 and a_2.

The accuracy of each of the equations in the set (1)–(6) was tested with observations. $|F_c|$ in (1) was estimated by using measured values of $|F_s|, \sigma_c$ and σ_s. Estimated $|F_s|$ in (2) was tested by using locally adjusted $\phi(Z/L)$, as explained above, and measurements of σ_s, u_*, H, E and θ. Similarly, the accuracy of u_*, estimated from (5) was tested with the input of measured values of L (using H and E) and σ_w.

4 Results and discussion

a *Estimated Fluxes*

Figures 3a–e illustrate some comparisons of the estimated fluxes computed from (1) with the observed fluxes from datasets Kinoje 90, Borden 90 and Borden 88. All the figures include small values that are clustered near the origin. We note that for the Kinoje site, (1) yields estimates of F_T (Fig. 3a) that agree well with the observations when F_q is a given input, yielding a correlation coefficient of 0.97 and an average error of 17% (Table 1). However, when F_{O_3} is a given input, although the scatter is not too large, the F_T estimates are much lower than the observed fluxes, yielding a systematic bias that bears a linear relationship to the observed fluxes. Although a linear correlation exists, the systematic error is large. Similarly, the corresponding results in Table 1 show a correlation coefficient of 0.59 but a large error of 479%. That there exists a correlation and the error is systematic gives some reason for optimism regarding the usefulness of (1). In Fig. 3b, which contains the Borden 90 datasets, the estimates of F_q appear somewhat larger than the observed fluxes when F_T is given but significantly smaller when F_{O_3} is given. When F_{O_3} is input in (1) to estimate either F_T or F_q, both Figs 3a and b show underestimated linear biases. These results indicate that the similarity theory

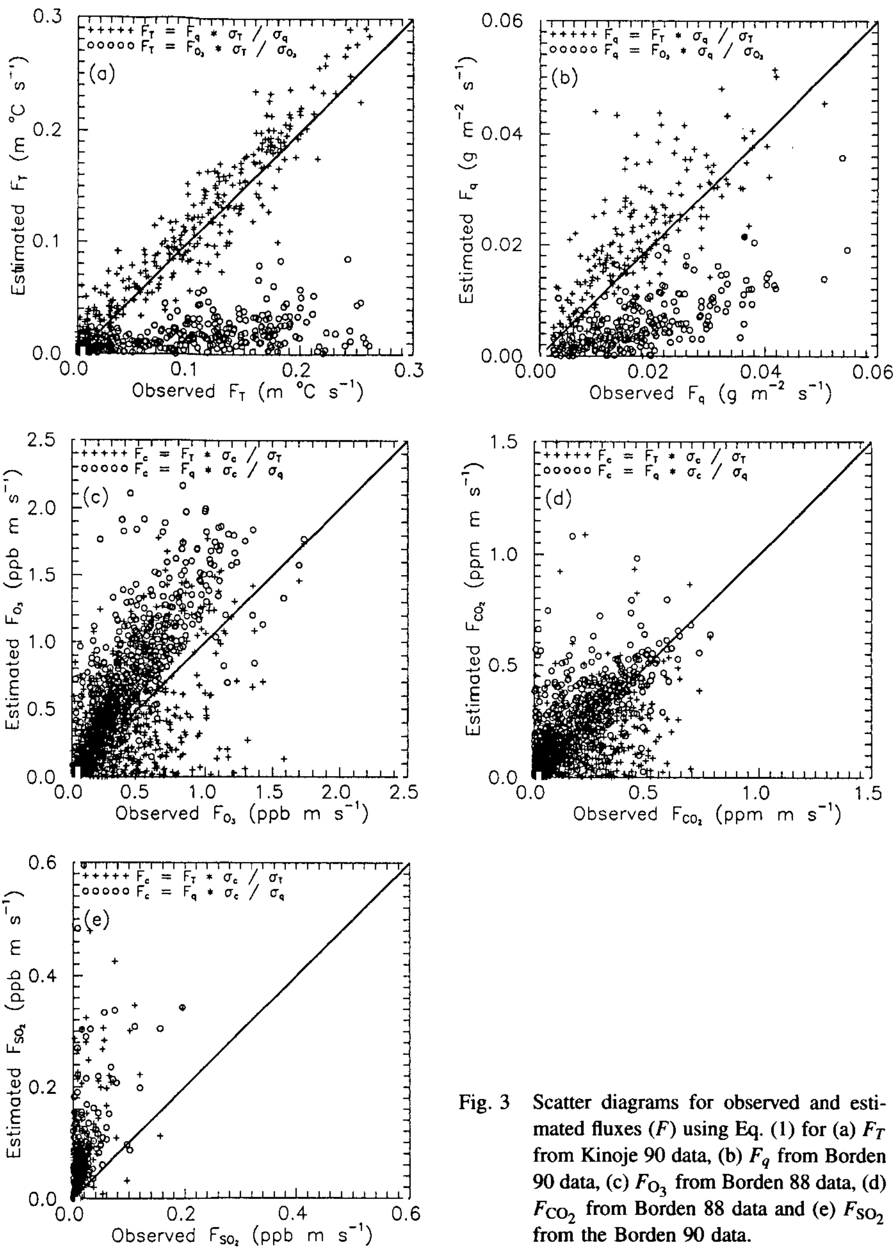

Fig. 3 Scatter diagrams for observed and estimated fluxes (F) using Eq. (1) for (a) F_T from Kinoje 90 data, (b) F_q from Borden 90 data, (c) F_{O_3} from Borden 88 data, (d) F_{CO_2} from Borden 88 data and (e) F_{SO_2} from the Borden 90 data.

applies better to the pair (F_T, F_q) than to either (F_T, F_{O_3}) or (F_q, F_{O_3}). A comparison of Figs 3a and b also indicates a better similarity for the pair (F_q, F_{O_3}) than for (F_T, F_{O_3}). In support of this possibility, Fig. 3c shows, using Borden 88 data, a better similarity for the pair (F_{O_3}, F_q) than for (F_{O_3}, F_T) when the observed F_T and F_q are used, respectively, as input in (1) to estimate ozone fluxes. Figure 3d gives

TABLE 1. Correlation coefficients and average errors for testing Eq. (1)

Dataset	Flux Computed with Eq. (1)	Observed Flux Used in Eq. (1)	Correlation Coefficient	Average Error (%)	Number of Data Points
Borden 88	F_T	F_q	0.88	37	908
Borden 90	F_T	F_q	0.87	29	247
Kinoje 90	F_T	F_q	0.97	17	397
Borden 88	F_q	F_T	0.67	67	908
Borden 90	F_q	F_T	0.86	35	247
Kinoje 90	F_q	F_T	0.92	22	397
Borden 88	F_{O_3}	F_T	0.52	58	908
Borden 90	F_{O_3}	F_T	0.52	75	247
Kinoje 90	F_{O_3}	F_T	0.62	81	397
Borden 88	F_{CO_2}	F_T	0.57	68	908
Borden 90	F_{SO_2}	F_T	0.98	86	247
Kinoje 90	F_{CO_2}	F_T	0.19	69	397
Borden 88	F_{O_3}	F_q	0.82	44	908
Borden 90	F_{O_3}	F_q	0.55	72	247
Kinoje 90	F_{O_3}	F_q	0.63	82	397
Borden 88	F_{CO_2}	F_q	0.61	61	908
Borden 90	F_{SO_2}	F_q	0.98	85	247
Kinoje 90	F_{CO_2}	F_q	0.17	70	397
Borden 88	F_T	F_{O_3}	0.77	65	908
Borden 90	F_T	F_{O_3}	0.62	315	247
Kinoje 90	F_T	F_{O_3}	0.59	479	397
Borden 88	F_q	F_{O_3}	0.81	54	908
Borden 90	F_q	F_{O_3}	0.79	129	247
Kinoje 90	F_q	F_{O_3}	0.53	339	397

a similar indication when CO_2 fluxes are estimated from the observed (Borden 88) F_T and F_q, respectively, showing a better similarity for the pair (F_{CO_2}, F_q) than for (F_{CO_2}, F_T), although both show a large scatter. The same characteristic may be deduced from Fig. 3e for the SO_2 fluxes even though both the F_T and F_q observations yield larger overestimates of F_{SO_2}. Again, we note in Figs 3a–e that the estimated fluxes bear a reasonable linear relationship to the observed fluxes, sometimes yielding high correlations that may be associated with large systematic errors. Table 1 lists the correlation coefficients and the associated errors (in per cent) between the estimated and the observed fluxes for all the variables and the datasets that were employed in the calculation of fluxes from (1).

Our second investigation tests the form of $\phi(Z/L)$ in (3) using the non-dimensional normalized σ_s in (2) for our datasets for each scalar variable s. The literature provides ample examples for the form of $\phi(Z/L)$ for T and q, but very little information about its form for gaseous species. Figures 4a–e show the form of $\phi(Z/L)$ for T, q, O_3, SO_2 and CO_2, using the datasets of Borden 88, Borden 90 and Kinoje 90. Figures 4a and b for $s = T$ and $s = q$, respectively, agree with similar forms of $\phi(Z/L)$ published by other investigators, who have used different sets of data (Wesely et al., 1970; Weaver, 1990). Significant characteristics of this agreement are the lack of scatter in the data in the unstable region and the bend close to neutral stability. The large cluster near the neutral region indicates that most of the data had near-neutral stability. The scatter near $Z/L = 0$ is large for

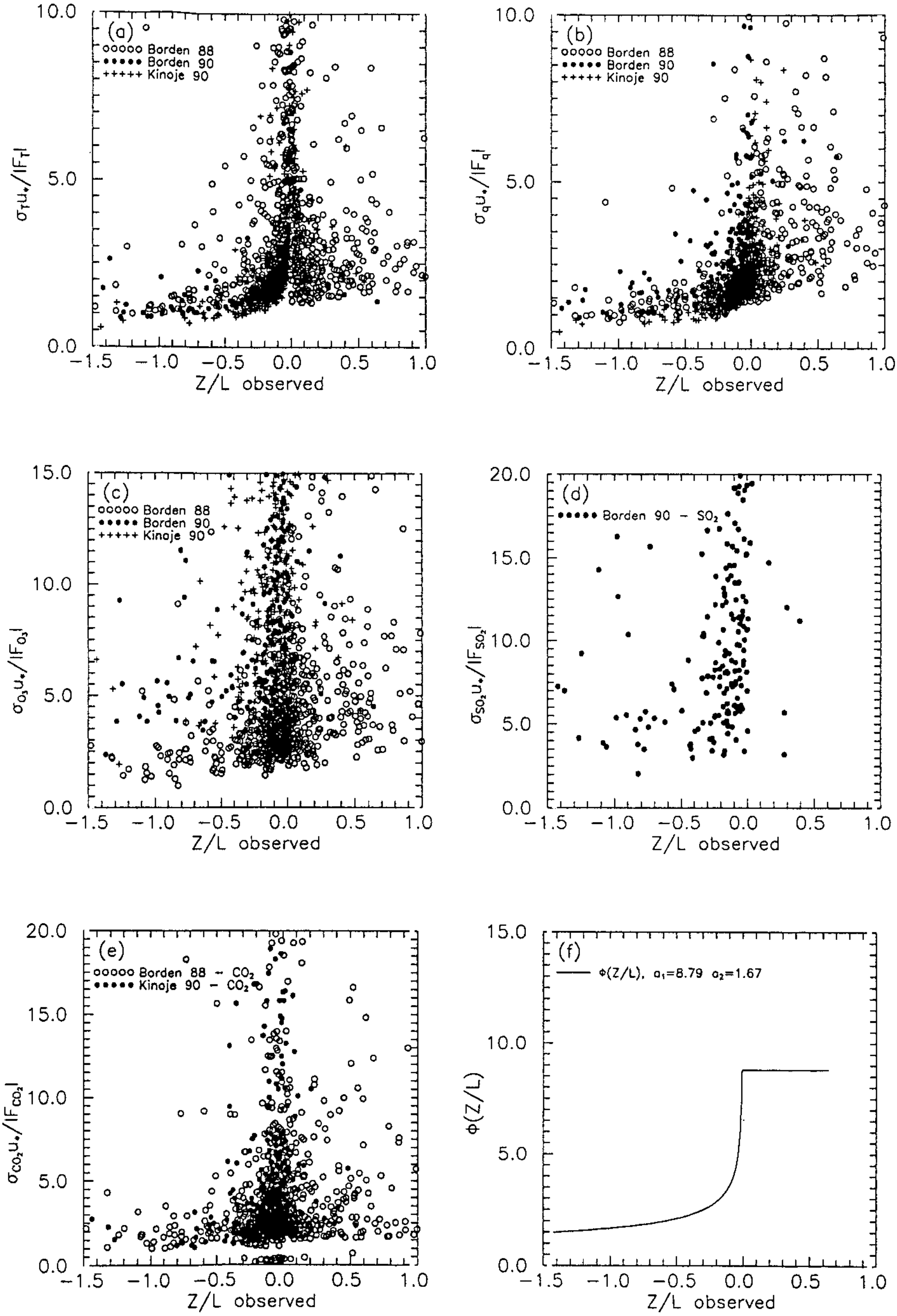

Fig. 4 Variations of the non-dimensional normalized standard deviation (σ) with stability Z/L for (a) temperature (T), (b) water vapour (q), (c) O_3, (d) SO_2, (e) CO_2 and (f) $\phi(Z/L)$ in Eq. (3) for $a_1 = 8.79$ and $a_2 = 1.67$ from the Borden 90 data.

TABLE 2. Coefficients a_1 and a_2 calculated using Weaver's (1990) method for Eq. (3)

Dataset	Flux	a_1	a_2
Borden 90	Temperature	5.44	0.84
	Humidity	5.95	1.13
	Ozone	8.79	1.67
	Sulphur Dioxide	12.37	4.23
Borden 88	Temperature	3.33	0.98
	Humidity	1.90	0.68
	Ozone	2.54	1.02
	Carbon Dioxide	4.61	2.90
Kinoje 90	Temperature	5.03	0.65
	Humidity	3.77	0.81
	Ozone	4.16	1.95
	Carbon Dioxide	4.83	1.30

both T and q, and in the stable region the scatter in q appears larger than in T. The commonly used assumption of a constant a_1 for T and q in the stable region does not appear to be justified because of the large scatter in this stability region. The scatter is larger for O_3, SO_2 and CO_2 in Figs 4c, d and e, respectively. Figure 4f shows the form of $\phi(Z/L)$ when the values of $a_1 = 8.79$ and $a_2 = 1.67$ are employed in (3). This agrees reasonably well with the form of $\phi(Z/L)$ for T in Fig. 4a and with that of q in Fig. 4b in the unstable region. However, Fig. 4c for the $\phi(Z/L)$ of O_3 shows a poor agreement with Fig. 4f when the Kinoje 90 and Borden 90 datasets are used. The characteristic bend in $\phi(Z/L)$ near neutral stability that appears in Fig. 4f seems to be absent in Fig. 4c for O_3 and in Fig. 4e for CO_2. The scatter in $\phi(Z/L)$ could limit the applicability of (2).

The third investigation tests the sensitivity of the estimated fluxes, computed from (2), to changes in the values of a_1 and a_2 that appear in the function $\phi(Z/L)$ of (3). Measured values were used for Z/L, u_* and σ_s for the purpose of this test. The a_1 and a_2 values were computed using our datasets and Weaver's (1990) method, as discussed in Section 3. These values are listed in Table 2 for all the scalar variables. Figures 5a–e show scatter diagrams of computed and observed fluxes of the scalar variables for the variety of a_1 and a_2 values listed in Table 2. For example, Fig. 5a shows little scatter in the fluxes when $a_1 = 5.44$ and $a_2 = 0.84$, calculated using the Borden 90 dataset for T. The corresponding correlation coefficient between the estimated and the observed F_T is shown to be 0.96 in Table 3, in contrast with the correlation coefficient of 0.86, obtained when Wesely's constants of $a_1 = 1.85$ and $a_2 = 1.25$ were employed. The small overestimation may be attributed, as in de Bruin et al. (1991), to non-uniform thermal conditions at the surface, which affect temperature advection. Figure 5b also shows a good agreement between estimated and observed F_q for $a_1 = 3.77$ and $a_2 = 0.81$, having a correlation coefficient of 0.89 compared with 0.66 obtained when Wesely's (1988) values were used, as recorded in Table 3. These two examples indicate the improvement that can be achieved by selecting the optimized values of a_1 and a_2, using Weaver's (1990)

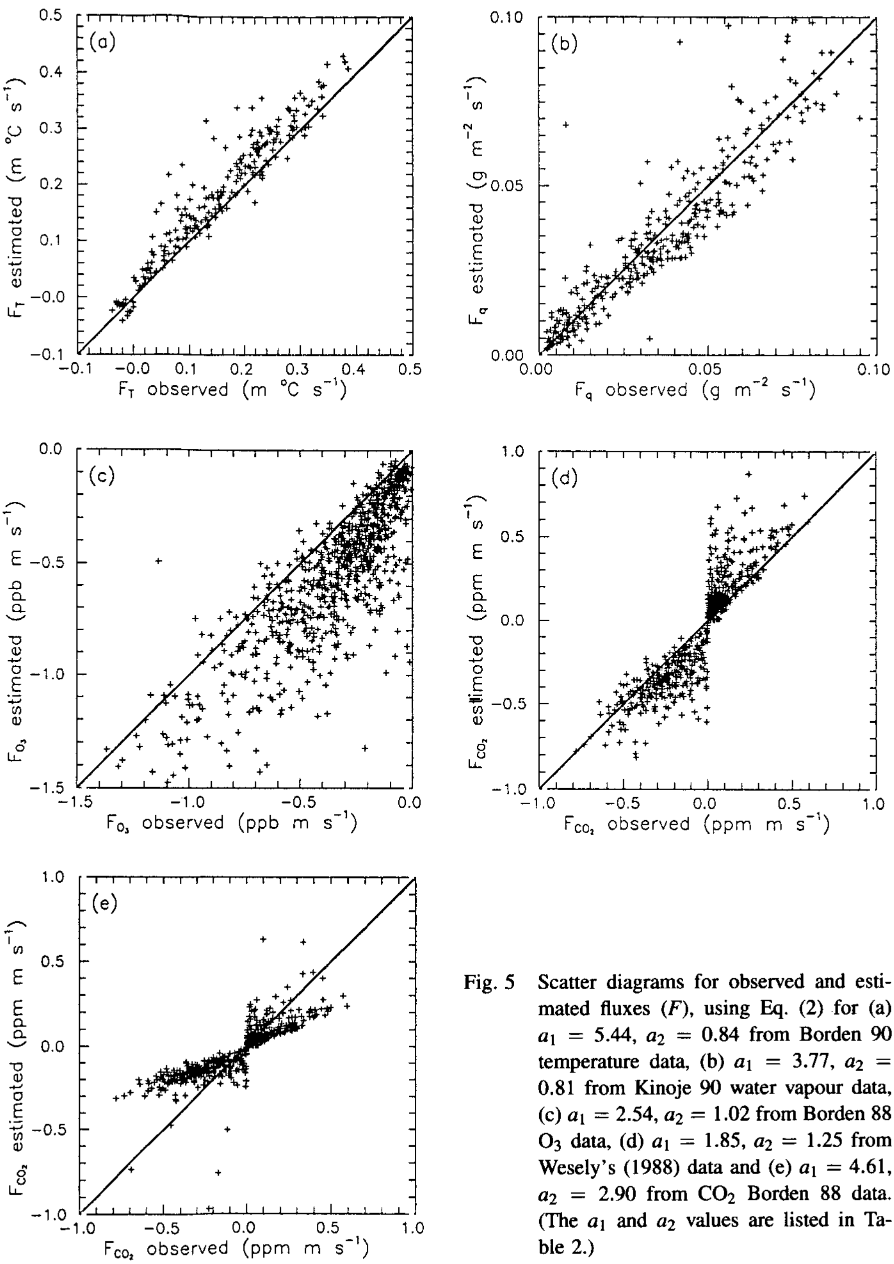

Fig. 5 Scatter diagrams for observed and estimated fluxes (F), using Eq. (2) for (a) $a_1 = 5.44$, $a_2 = 0.84$ from Borden 90 temperature data, (b) $a_1 = 3.77$, $a_2 = 0.81$ from Kinoje 90 water vapour data, (c) $a_1 = 2.54$, $a_2 = 1.02$ from Borden 88 O_3 data, (d) $a_1 = 1.85$, $a_2 = 1.25$ from Wesely's (1988) data and (e) $a_1 = 4.61$, $a_2 = 2.90$ from CO_2 Borden 88 data. (The a_1 and a_2 values are listed in Table 2.)

method. There were examples when Weaver's method yielded estimates of fluxes that were slightly inferior to those obtained when Wesely's constants were used, as can be seen for O_3 in Table 3 when $a_1 = 2.54$ and $a_2 = 1.02$, yielding a correlation coefficient of 0.77, insignificantly smaller than that of 0.8 obtained with Wesely's constants. The use of Wesely's constants, however, yielded a slightly larger average

TABLE 3. Correlation coefficients for fluxes computed via Eq. (2)

Dataset	a_1	q_2	Computed Flux	Correlation Coefficient	Average Error
Borden 88	3.33	0.98	F_T	0.71	52
Wesely	1.85	1.25		0.77	82
Borden 88	1.90	0.68	F_q	0.81	29
Wesely	1.85	1.25		0.83	22
Borden 88	2.54	1.02	F_{O_3}	0.77	39
Wesely	1.85	1.25		0.80	47
Borden 90	5.44	0.84	F_T	0.96	17
Wesely	1.85	1.25		0.86	24
Borden 90	5.95	1.13	F_q	0.83	37
Wesely	1.85	1.25		0.78	43
Borden 90	8.79	1.67	F_{O_3}	0.62	58
Wesely	1.85	1.25		0.58	78
Kinoje 90	5.03	0.65	F_T	0.98	23
Wesely	1.85	1.25		0.89	41
Kinoje 90	3.77	0.81	F_q	0.89	20
Wesely	1.85	1.25		0.66	37
Kinoje 90	4.16	1.95	F_{O_3}	0.67	61
Wesely	1.85	1.25		0.61	80
Borden 88	4.61	2.90	F_{CO_2}	0.59	380
Wesely	1.85	1.25		0.59	309
Borden 90	12.37	4.23	F_{SO_2}	0.98	244
Wesely	1.85	1.25		0.98	827
Kinoje 90	4.83	1.30	F_{CO_2}	0.24	104
Wesely	1.85	1.25		0.27	128

percentage error. Figure 5d shows the scatter diagram for computed and observed F_{CO_2} when Wesely's values for a_1 and a_2 were used. This can be contrasted with F_{CO_2} in Fig. 5e for $a_1 = 4.61$ and $a_2 = 2.90$, showing that both methods yield improved results only when the fluxes cluster near zero. The correlation coefficients that correspond to Figs 5d and e have equal values, recorded in Table 3 as 0.59 associated with large errors. Table 3 records the remaining correlation coefficients between the estimated and the observed fluxes that were obtained with Weaver's a_1 and a_2 values and with Wesely's. For CO_2, (2) yielded poor estimates of the fluxes when either Weaver's or Wesely's values for the constants were used. This may be due to the non-uniformity of the surface as a source of CO_2. These results seem to suggest that the a_1 and a_2 values may depend upon the site and perhaps the prevailing meteorological conditions. This suggestion appears in Wesely (1988), Weaver (1990) and others.

We now test (5) for computing u_* using our datasets. Hicks (1981) pointed out that the values of 1.3 and 2.0 in (5) may not be universal. Figure 6 shows little scatter between the computed u_* and observed u_*, obtained from the three datasets of the present study. The correlation coefficients are 0.97 for Borden 88, 0.97 for Borden 90, and 0.94 for Kinoje 90, and the corresponding average percentage

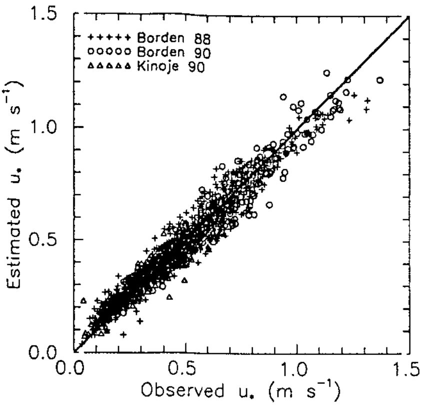

Fig. 6 Scatter diagram of estimated and observed u_*, using Eq. (5) for the three datasets.

errors are 9.0, 8.0 and 11.0. Thus, the values of 1.3 and 2.0 in (5) fit our datasets reasonably well.

We have discussed the accuracy that can be expected from each of (1), (2), (3) and (5) and applied the flux-adjusted values of a_1 and a_2 to each of our datasets. We now solve the combined set of equations (1)–(6) using (9) and (10); instead of discussing fluxes, we analyse the dry deposition velocities (fluxes divided by concentrations) for O_3, SO_2 and CO_2. The results are illustrated as time series in Figs 7 and 8 for specific days taken from each of the datasets. When possible, the same days were selected as in Padro et al. (1991, 1992a, 1992b) so that comparison could be made with dry deposition velocities (V_d) that were obtained from a resistance analogue model, known as ADOM (Acid Deposition and Oxidant Model) and its modified version. The figures also include dry deposition velocities obtained when Wesely's (1988) values for a_1 and a_2 were employed.

b *Estimated Dry Deposition Velocities*

Figure 7a shows V_d values of O_3 for days 3 and 4 of August 1988 (Julian days 216 and 217) over a fully leafed (summer) deciduous forest. Both Wesely's and Weaver's methods overestimate V_d but Weaver's method agrees better with the observed values of V_d. The overestimation may be an indication that the σ_{O_3} was large because of a small signal-to-noise ratio. Nevertheless, the patterns of the estimated V_d are similar to the observed pattern. A similar conclusion can be drawn about the O_3 V_d over the leafless deciduous forest (Fig. 7b) for 23 and 24 April 1990 (Julian days 113 and 114). In Fig. 7c Wesely's values for a_1 and a_2 yield large values of V_d for SO_2 whereas Weaver's method shows better agreement. Using Kinoje 90, the O_3 V_d obtained using Weaver's method shows (Fig. 8a), again, reasonable agreement with the observations. An example favouring Wesely's method is shown in Fig. 8b for the CO_2 V_d over the fully leafed deciduous forest where the agreement with observations is remarkable except for the midday values on the second day (hour 12 on day 217). However, for CO_2 over Kinoje, Weaver's method is again superior in estimating the V_d (Fig. 8c). It can be concluded that by adjusting the values of a_1 and a_2 with the site-specific data, the resulting dry deposition velocities are in better agreement with the observations. It can be seen that at times

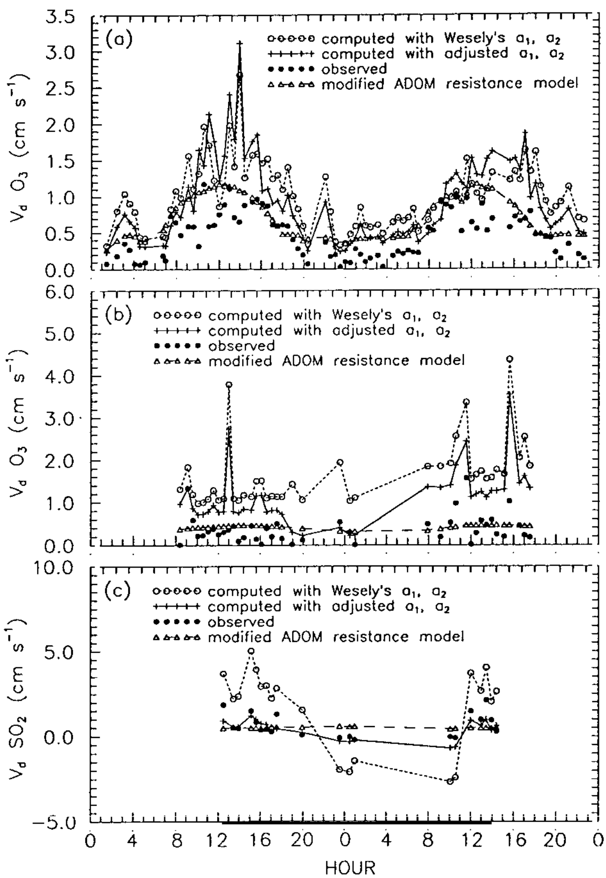

Fig. 7 Half-hourly variations of dry deposition velocity (V_d), estimated from Eqs (1)–(5) with adjusted a_1, a_2, and with Wesely's a_1, a_2, and the observed V_d for (a) O_3 from Borden 88 data (Julian days 216 and 217), (b) O_3 from Borden 90 data (Julian days 113 and 114) and (c) SO_2 from Borden 90 data (Julian days 113 and 114).

the adjusted (a_1, a_2) variance method yields V_d values that are comparable with those of the resistance analogue model.

c *Testing Weaver's Method with Independent Data*

The above discussions, alleging possibilities for improved flux estimates using constants obtained from Weaver's (1990) method, need to be tested with independent datasets. For example, a_1 and a_2 were computed using CO_2 data for 27 and 28 July 1988 and then applied to estimate CO_2 fluxes using data for 15 and 16 July 1988. The resulting values of a_1 and a_2 are 3.3 and 1.5, respectively. The results are shown in Fig. 9a in the form of scatter diagrams for the estimated fluxes using Weaver's (1990) and Wesely's (1988) methods. Wesely's method did not include any filtering of noise. The method of Weaver appears to yield improved estimates of the CO_2 fluxes. Similarly, when $a_1 = 3.3$ and $a_2 = 1.5$ were applied to estimate the CO_2 fluxes for 13 and 14 August 1988, Fig. 9b, again, shows improved fluxes.

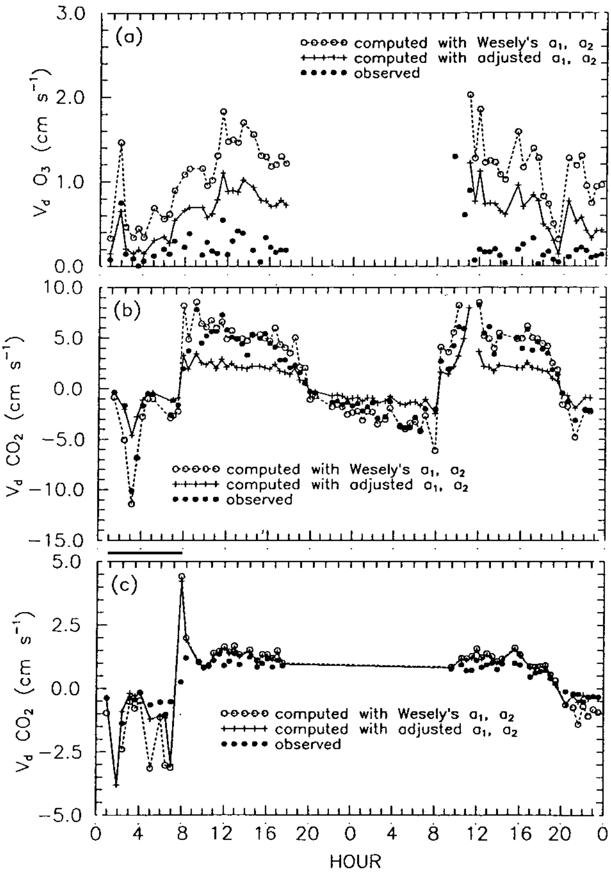

Fig. 8 Half-hourly variations of dry deposition velocity (V_d), estimated from Eqs (1)–(5) with adjusted a_1, a_2, and with Wesely's a_1, a_2, and the observed V_d for (a) O_3 from Kinoje 90 data (Julian days 201 and 203), (b) CO_2 from Borden 88 data (Julian days 216 and 217) and (c) CO_2 from Kinoje 90 data (Julian days 201 and 202).

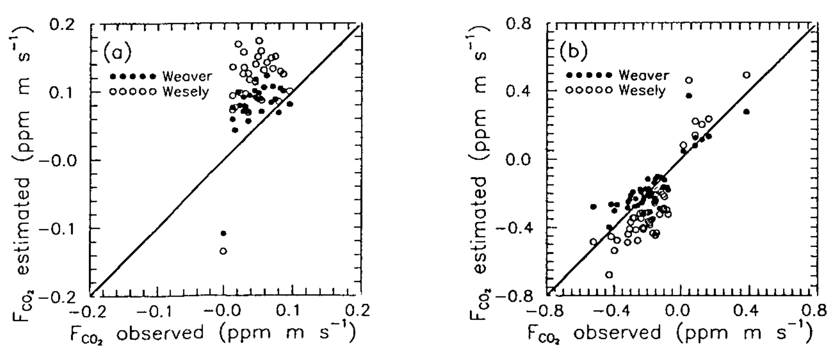

Fig. 9 Scatter diagrams for observed and estimated CO_2 fluxes (F_{CO_2}) for (a) 15 and 16 July 1988 and (b) 13 and 14 August 1988, using Wesely's constants $a_1 = 1.85$, $a_2 = 1.25$, and Weaver's constants $a_1 = 3.3$, $a_2 = 1.5$, obtained from Eq. (2) and data for 27 and 28 July 1988.

5 Summary and conclusions

We have investigated the possibilities of using the flux-variance relations for estimating the fluxes of temperature, water vapour, O_3, SO_2 and CO_2 and the dry deposition velocities of the latter three gases. To this end, we used large sets of data measured with fast-response instruments over three different land use categories. Each equation that forms part of the flux-variance relations was tested individually with the observed data. We shall summarize the cases when these equations yielded reasonable estimates and those when they did not.

When either the temperature flux (F_T) or the water vapour flux (F_q) was known from measurements, the other was derived from it quite accurately, given the standard deviations of temperature and water vapour. When either F_T or F_q was used to estimate the fluxes of O_3, SO_2 and CO_2 the estimates were not as successful but F_q appeared to bear a better similarity to these scalars than F_T did. Other studies (Wesely, 1988; Weaver, 1990) indicate that this result may depend upon the moisture conditions of the underlying surface.

The flux-variance relations assume the existence of a universal function ($\phi(Z/L)$) for the non-dimensional normalized standard deviations and another universal function for the inverse of the non-dimensional friction velocity (u_*). Although $\phi(Z/L)$ appeared to be non-universal and needed to be adjusted for each land use type and scalar variable, the function for u_* seemed to be universal. The site specific functions $\phi(Z/L)$ yielded estimates of dry deposition velocity that were in better agreement with the observations and with estimates from a resistance analogue model than the estimates that were obtained when the same $\phi(Z/L)$ was used for all sites and scalars, as in Wesely (1988).

For temperature and humidity, the flux-variance relations appear useful. This was not true for O_3, SO_2 and CO_2. However, for trace gases, in the cases where the estimated fluxes showed a linear relationship to the observed fluxes, in spite of large systematic errors, there is some optimism that the flux-variance relations may be useful. It may be possible that with accurate variance measurements, the flux-variance relations may be useful over surfaces that are smoother than those of the present study. More research needs to be carried out in devising methods for filtering the noise in the variances of trace gases.

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