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**Soil Phosphate Diffusion Coefficients: Their Dependence on Phosphorus Concentration
and Buffer Power**

P. B. S. Bhadoria, J. Kaselowsky, N. Claassen, and A. Jungk

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ABSTRACT

Measured values of effective P diffusion coefficients, D_e , were compared with D_e values calculated on the basis that P only diffuses in the liquid phase. Two soil blocks differing in P content were placed into contact for 2 wk, then quick frozen and cut into slices of 0.2 mm with a refrigerated microtome. The slices were extracted with 4 M HCl to obtain a P concentration-distance curve. An average diffusion coefficient, \bar{D}_e , was determined from the total amount of P that had diffused from the soil of high P concentration to that of low P concentration. Effective diffusion coefficients for a given soil P concentration, D_{eC} , were obtained by analyzing the concentration-distance curve. Phosphorus buffer curves were established by adsorption in the lower concentration range and by desorption in the higher concentration range, according to the process occurring during P diffusion in soil. The value of \bar{D}_e increased from 2.2 to $13.9 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ when the average soil P concentration increased from 380 to 580 mg P kg⁻¹ soil. The D_{eC} values increased with increasing P concentration, although the relationship was not unique. For example, the same D_{eC} value of $5 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ was obtained at 340 mg P kg⁻¹ when P was being adsorbed and at 600 mg P kg⁻¹ when P was being desorbed during the process of diffusion. This result was related to different P-buffer behavior of the soil during adsorption and desorption. The P diffusion coefficient in soil is, therefore, dependent not only on the buffer power, which is influenced by concentration, but also on whether P is being desorbed or adsorbed and on the time available for reaction. The value of D_e can be calculated if suitable values for the buffer power are used, and D_e for a wide P-concentration range can be calculated if a weighted average buffer power is used.

THE IMPORTANCE of the different transport mechanisms by which nutrients in soil are brought to the root surface depends on the nutrient ion in question. Barber (1962) established that mass flow may supply the root with much of the plant's need for Ca, Mg, and N but that this is probably not the main mechanism for K or P transport. Bouldin (1961), Olsen et al. (1962), and Lewis and Quirk (1967) have demonstrated that diffusion supplies almost all the plant's phosphate. Transport of P by diffusion in soil to plant roots depends, according to Fick's first law, on the concentration gradient between the soil and the root surface and the diffusion coefficient.

The concentration of an ion in soil affects the self-diffusion coefficient of that ion. Since diffusion in soil

occurs in the liquid phase only (Nye and Tinker, 1977), the self-diffusion coefficient of the ion increases as the ratio of the ions in the free soil solution to the total diffusible ions in the soil increases. This ratio usually increases with increasing nutrient content of the soil. The reciprocal of this ratio is called the buffer power.

Several researchers have shown that diffusion coefficients for cations in soil (Clark and Graham, 1968; Mott and Nye, 1968; Reddy et al., 1980) and in clay (Ellis et al., 1970; Phillips and Brown, 1964) depend on concentration. Measurements of the self diffusion of a number of ions in soil have been made by Graham-Bryce (1963b). In many practical applications of diffusion theory, however, such as the transfer of ions to plant roots (Barber, 1962), one does not deal with self diffusion, but with bulk diffusion, which is the diffusion of an ion that is accompanied by and exchanged for other ion species. Hendriks et al. (1981) found that plants reduced the isotopically exchangeable phosphate to only 50% of the initial concentration. From their results, it can be inferred that not all added ³²P was involved in diffusion to plant roots.

Phillips and Brown (1966) and Phillips et al. (1968) have measured concentration-dependent self-diffusion coefficients of ⁸⁶Rb and ³²P by analyzing the concentration vs. distance curves, using the procedure described by Matano (1933). Such techniques do not appear to have been applied to the measurement of concentration-dependent bulk-diffusion coefficients of P in soils.

The aim of this work was to measure effective P-diffusion coefficients by bulk diffusion with special emphasis on the dependence of the diffusion coefficient on P concentration and P buffer power. A further objective was to determine to what extent the measured concentration-dependent P-diffusion coefficients can be explained on the basis of the theory that P diffusion in soil occurs mainly through the liquid phase.

MATERIALS AND METHODS

Soil and Fertilization. The soil material used, from a Typic Hapludalf from Hottenrode near Goettingen, was taken from the layer 70 to 90 cm below surface. It had the following characteristics: 3% sand, 81% silt, 16% clay, pH 5.8, and 0.17% organic C. After sieving to 2 mm, P was added (80% KH₂PO₄, 20% K₂HPO₄ dissolved in water) in quantities of 0, 200, and 400 mg P kg⁻¹ soil.

Definition of Diffusion Coefficients used. The D_e is defined by Fick's first law, $F_D = -D_e(dC/dx)$, as the proportionality constant between the diffusive flux, F_D , and the concentration gradient, dC/dx , where C is the concentration of diffusible P, and x is distance. The D_e of soil P may vary with C . We separately regard here:

P.B.S. Bhadoria, Indian Inst. of Technology, Kharagpur-721302, W.B. India; J. Kaselowsky, N. Claassen, and A. Jungk, Inst. für Agrikulturchemie, Georg-August-Universität, von Siebold-Str. 6, D-3400 Goettingen, West Germany. Contribution from the Inst. für Agrikulturchemie, Georg-August-Universität, Goettingen. Received 24 Oct. 1989. *Corresponding author.

D_{ec} concentration-dependent effective diffusion coefficient, i.e., D_e at a specified P concentration
 — measured by bulk diffusion and using Eq. [2] or [3],
 — calculated by Eq. [6] using buffer power at the specified P concentration.

\bar{D}_e average effective diffusion coefficient for the total range of P concentrations included
 — measured by bulk diffusion and using Eq. [1],
 — calculated by Eq. [6] using the average weighted buffer power, \bar{b} .

Measurement of Phosphorus Diffusion Coefficients in Soil. The double half-cell technique of Phillips and Brown (1964), modified by Kaselowsky et al. (1990), was employed. Each cell consisted of a piece of plastic pipe with an inner diameter of 4.2 cm and a length of 1.8 cm, thereby giving a soil layer of 1.8 cm. Diffusion units were composed of two cells filled with the same soil, at the same water content (240 g kg⁻¹) and bulk density (1.46 g cm⁻³) but of different P level. Combinations were 200–0, 400–0, and 400–200 mg P kg⁻¹ soil with four replicates each. At the end of the 14-d diffusion period, cells were separated, soil cores were quick frozen in liquid N₂, and cut into slices of 0.2-mm thickness with a refrigerated microtome. Phosphorus was extracted from each soil slice with 4 M HCl. For details, see Kaselowsky et al. (1990). The procedure enables the measurement of P distribution throughout the diffusion medium and provides the possibility of determining both an average and a concentration-dependent diffusion coefficient of soil P.

Average Diffusion Coefficient. The \bar{D}_e for the concentration range between the fertilized and the unfertilized soil was measured from the amount of P that moved from the soil of high to the soil of low P concentration using Eq. [1] from Jost (1952, p. 22):

$$\bar{D}_e = \frac{M_P \pi}{A^2 (C_2 - C_1)^2 t} \quad [1]$$

where M_P = total amount of P which diffused into the soil of low P concentration in time t (14 d), A = cross-sectional area of diffusion (13.8 cm²), $(C_2 - C_1)$ = concentration difference between fertilized and unfertilized soil blocks.

Concentration-Dependent Diffusion Coefficient. According to Matano (1933) and Crank (1975, p. 230–234), D_{ec} can be determined from the concentration–distance curve. The D_{ec} for $C = C_n$ at the left-hand side of Fig. 1, where P diffuses out of the soil, is given by

$$D_{ec} = \frac{1}{2t} \frac{dx}{dC} \int_{C_n}^{C_2} \times dC \quad [2]$$

and for $C = C_m$ at the right-hand side of Fig. 1, where P diffuses into the soil, is given by

$$D_{ec} = -\frac{1}{2t} \frac{dx}{dC} \int_{C_1}^{C_m} \times dC \quad [3]$$

After the diffusion period, the concentration–distance curve is plotted (Fig. 1) and from it dx/dC and the area under the curve (the integral of Eq. [2] and [3]) are determined graphically. The integrals can be obtained by using a planimeter or by counting squares, and dx/dC by drawing tangents at the concentration for which D_{ec} is to be determined.

Buffer Power. The buffer power is needed to calculate the effective diffusion coefficient. Buffer power is defined as the slope of the buffer curve,

$$b = \Delta C / \Delta C_i \quad [4]$$

where C_i is the concentration of solute in the liquid phase (mg cm⁻³ soil solution) and C is the concentration of diffusible solute in soil (mg cm⁻³ of soil). To obtain a single

buffer power for a larger soil concentration range, an average weighted buffer power, \bar{b} , was calculated from

$$\bar{b} = \Sigma(b_i \Delta C_i) / \Sigma \Delta C_i \quad [5]$$

where b_i is the buffer power over a small range (i) in soil P concentration, ΔC_i .

In the process of diffusion, P is desorbed from the soil of high concentration and transported to the unfertilized soil, where it is adsorbed. The process of adsorption was simulated by fertilizing the soil with graded amounts of P and keeping it moist for 20 d, which is about the time of the diffusion experiment. The P concentration was then measured in the soil solution, which was obtained by the displacement procedure of Adams (1974). The process of desorption was simulated by shaking the fertilized soil (400 mg P kg⁻¹) with a 0.01 M CaCl₂ solution for a period of 14 d. The P concentration was then measured in the filtrate. The ratios of soil to solution varied from 1:2 to 1:1000.

Plotting the P added against soil-solution P concentration gave the adsorption or buffer curve for the soil in the half cell of low concentration. Plotting the P desorbed against the equilibrium solution P concentration gave the desorption or buffer curve for the soil in the half cell of high concentration.

The Impedance Factor. The impedance factor, f , which is necessary for applying Eq. [6] from Nye (1968) to calculate D_{ec} , was estimated from Cl diffusion in the same soil by using the same technique as for P. The effective diffusion coefficient for Cl was determined by the simplified equation of Schofield and Graham-Bryce (1960). In the next step, f was obtained from D_f/D_i , where D_i is the diffusion coefficient of Cl in water.

RESULTS

Concentration–Distance Curve and Amount of Phosphorus Diffused. As an example, the distribution of P diffusing from 400 to 0 mg P kg⁻¹ is presented in Fig. 1. The concentration–distance curve obtained for either 200–0 or 400–200 was similar to that shown in Fig. 1. It can be seen that, starting from the left side, the concentration curve becomes steeper towards the border of the half cell ($x = 0$). At this point, the concentration curve shows a discontinuity indicated by the gentler slope on the unfertilized than on the fertilized side.

The value of M_P was determined graphically. The

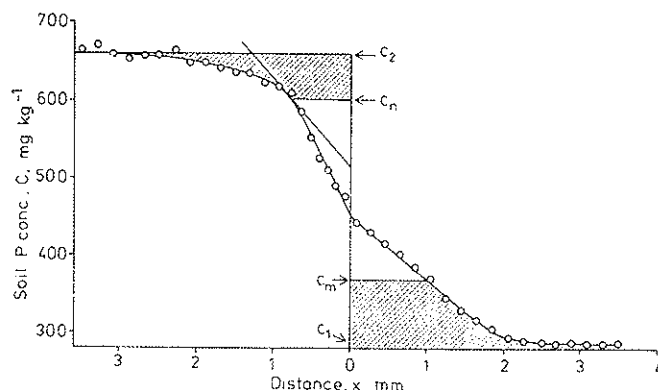


Fig. 1. Phosphate distribution in Hottenrode silt loam, after 2 wk of diffusion, used to determine effective P diffusion coefficients. Concentration-dependent diffusion coefficient (D_{ec}) is 5.6×10^{-13} m² s⁻¹ for concentration $C = C_n$, where P diffuses out of the soil, and 6.2×10^{-13} m² s⁻¹ for $C = C_m$ where P diffuses into the soil. (Average of four replicates, soil bulk density = 1.46 g cm⁻³. P addition to the soil: left side, 400 mg kg⁻¹; right side, no P).

effect of added soil P on M_i is presented in Table 1. The variation between the replicates was least at 400-0 and greatest at 400-200. Thus, measurement of M_i becomes less reliable when the concentration difference is small. The value of M_i increased from $7.65 \mu\text{g cm}^{-2}$ at 200-0 to a maximum of $26.38 \mu\text{g cm}^{-2}$ at 400-0. Hence, the size of M_i depends on the gradient, as seen by comparing 400-0 and 400-200. However, when the treatments 400-200 and 200-0 are compared, it is clear that P diffusion in soil is also dependent on the P concentration.

Average Diffusion Coefficient. With the M_i values from Table 1, \bar{D}_e were calculated by using Eq. [1]. The dashed lines in Fig. 2 represent the \bar{D}_e for the different P-concentration ranges. The value of \bar{D}_e increased from $2.2 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 200-0 to $6.0 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 400-0 and $13.9 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 400-200. In contrast to the amount diffused, the \bar{D}_e is dependent on the concentration in the soil but apparently not on the gradient.

Concentration-Dependent Diffusion Coefficient. Using P-distribution curves as shown, for example, in Fig. 1, D_{ec} were determined by means of Eq. [2] and [3], where dx/dC is the inverse of the slope of the tangent drawn to the distribution curve and the integral is equal to the shaded area at the point at which the tangent is drawn. In the right-hand side of Fig. 1, the concentration vs. distance curve at C_m is almost linear, taking all points into consideration. Using this procedure, the diffusion coefficient at any concentration can be calculated.

In this way, D_{ec} were determined from the four replicates of each treatment, averaged, and plotted against soil P concentration. As seen in Fig. 2, D_{ec} increased almost linearly with the increase in soil P concentration to a maximum value of $3.3 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 200-0, $9.1 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 400-0, and $19.9 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ at 400-200. However, there is always a discontinuity in the curve when going from the unfertilized to the fertilized soil, where D_{ec} is at first lower but increases again with increasing concentration. A comparison of these D_{ec} values with the \bar{D}_e values determined with Eq. [1] shows that the mean value of D_{ec} is similar to \bar{D}_e .

Calculation of Effective Phosphorus-Diffusion Coefficients. According to our current knowledge, P diffusion in soil occurs in the liquid phase. If so, the D_e in soil can be expressed by Eq. [6], which was suggested by Nye (1968):

$$D_e = D_i \theta f/b \quad [6]$$

where D_i = the diffusion coefficient of P in free solution ($0.89 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$) and θ = the volumetric water content.

For applying Eq. [6], the values for θ (0.35 g cm^{-3}) were obtained from water loss of the soil at 105°C , f (0.32) was estimated from Cl diffusion in the same soil, and D_i was taken from the literature.

The b as a function of soil P concentration is shown in Fig. 3. For the concentration range of 280 to 480 mg P kg^{-1} , b , which was obtained by adsorption, decreased from 600 to 38. For the range of 480 to 680 mg P kg^{-1} , b , as obtained by desorption, decreased

Table 1. Amount of P that moved by diffusion from the fertilized into the unfertilized soil block (M_i) in 2 wk. Soil bulk density = 1.46 g cm^{-3} and moisture content = 240 g kg^{-1} .

Treatment	M_i	Coefficient of variation
mg P kg^{-1}	$\mu\text{g P cm}^{-2}$	%
200 and 0	7.65	10
400 and 0	26.38	5
400 and 200	18.72	11

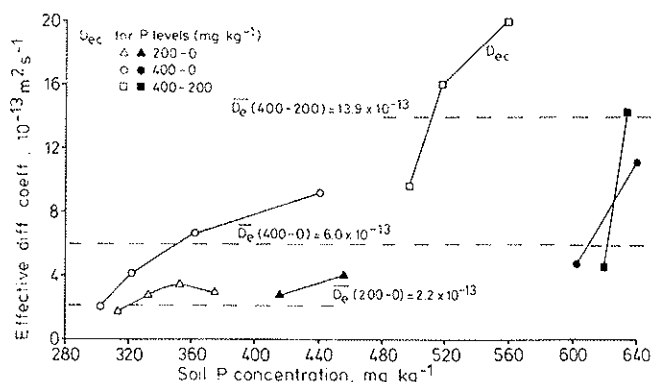


Fig. 2. Relation of concentration-dependent effective P-diffusion coefficients, D_{ec} , as measured by analyzing P-distribution curves, to soil P concentration and measured average diffusion coefficients, \bar{D}_e . Open and closed symbols refer to the half cells of low and of high P concentration, respectively.

from 409 to 4. Thus, the trend is in both cases equal: a decrease of b with the increase of P concentration. However, there is a large change when going from the initially fertilized to the unfertilized soil. At the interface, soil P concentration is about 480 mg P kg^{-1} in both cells (Fig. 1), but the buffer power is >500 on the initially fertilized side but <50 on the unfertilized side.

Equation [6] was used to obtain calculated D_{ec} and average \bar{D}_e . The values for θ , f , and D_i are given above. The values for b to calculate D_{ec} were taken from Fig. 3 and, for \bar{D}_e , b was obtained according to Eq. [5] from the data of Fig. 3. The calculated results are plotted together with measured values for the treatment of 400-0 in Fig. 4. It can be seen that, in the unfertilized soil in the low concentration range (300 to 400 mg P kg^{-1}), calculated and measured D_{ec} values agree fairly well and show a similar increase from 2 to $8 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ but, in the range of 400 to 480 mg P kg^{-1} , calculated D_{ec} was considerably higher than measured D_{ec} . Crossing the border from the unfertilized to the fertilized half cell, both measured and calculated diffusion coefficients decreased sharply to about 2 and $4 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$, respectively. Thereafter, higher soil P levels produced a sharp and similar increase in both calculated and measured D_{ec} . However, calculated D_{ec} is, in all cases, higher than measured D_{ec} .

The \bar{D}_e , as calculated with Eq. [6] and measured, using Eq. [1], are also shown in Fig. 4. These values agree fairly well. They represent the average of the range from 280 to 680 mg P kg^{-1} and include the effect of desorption and adsorption. The comparison of calculated D_e values with measured ones show that the current theory of P diffusion is quite realistic.

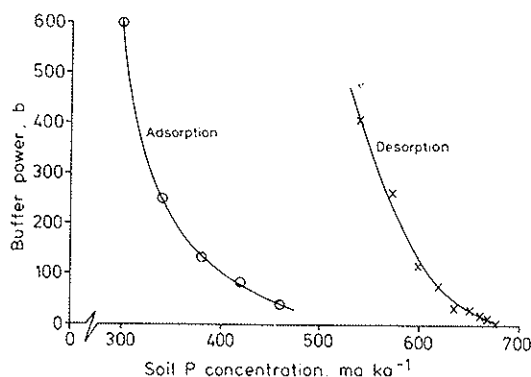


Fig. 3. Phosphorus buffer power in relation to P concentration obtained by adsorption for the lower range (diffusion into the soil) and by desorption for the higher range of P concentrations (diffusion out of the soil). (Bulk density 1.46 g cm^{-3}).

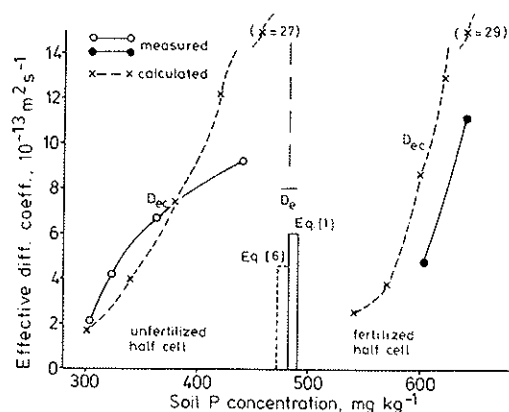


Fig. 4. Comparison of measured and calculated concentration-dependent, D_{ec} , and average \bar{D}_e , P-diffusion coefficients. Treatments: left side, no P; right side, 400 mg kg^{-1} .

DISCUSSION

The method described here to measure effective P-diffusion coefficients in soil is based on the diffusion of P from a soil of high to one of low P concentration. Other authors (e.g., Phillips and Brown, 1966; Lewis and Quirk, 1967) used the same technique, but with radioactive nuclides, in order to achieve a high accuracy of measurements if the amounts diffusing were small. Satisfactory precision of measurement was achieved without radioactive nuclides in this study, as shown by the coefficient of variation in Table 1 or by Kaselowsky et al. (1990). A relatively high concentration difference between the fertilized and the unfertilized soil blocks is necessary, however. This, in turn, is associated with large differences of the buffer power (Fig. 3) and of diffusion coefficients (Fig. 2 and 4). Phosphorus depletion at the soil-root interface of rape (*Brassica napus* L.) seedlings was found between 200 and 300 mg P kg^{-1} in a similar soil (Jungk and Claassen, 1989). Therefore, the conditions in this study appear to be very relevant for P nutrition of plants.

The concentration-distance curve (Fig. 1) shows an abrupt change of slope at the interface ($x = 0$) between fertilized and unfertilized soil. According to Fick's first law, $D_e(dC/dx)$, the flux by diffusion must be the same to the left and right of this point. Since the slope of the concentration-distance curve, dC/dx , is higher on

the fertilized than on the unfertilized side, D_e has to be smaller. That is, even though the concentration in soil is about the same on both sides of $x = 0$, the diffusion coefficient is smaller on the fertilized than on the unfertilized soil. This conclusion is confirmed by analyzing the concentration-distance curve with the procedure of Matano (1933), as shown in Fig. 2 and 4. A similar change of slope in the concentration-distance curve and the diffusion coefficient was found by Phillips and Brown (1966) for ^{89}Sr diffusing from a Sr-saturated to a H-saturated soil.

Considering the unfertilized and fertilized half cells separately, it can be seen that, with increasing soil P concentration, the D_{ec} increases (Fig. 2). This relationship is not unique but differs for the unfertilized and fertilized soil. For example, for the 400-0 treatment, a D_{ec} value of about $5 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ was obtained with 340 mg P kg^{-1} on the unfertilized but with 600 mg P kg^{-1} on the fertilized side. It must, therefore, be concluded that the P-diffusion coefficient in soil is, strictly speaking, not solely dependent on soil P concentration.

The approximate agreement between the measured and calculated diffusion coefficients, particularly \bar{D}_e (Fig. 4), and the same discontinuity of calculated D_{ec} values at $x = 0$ indicates that the equation of Nye (1968) (Eq. [6]) represents a realistic description of P diffusion in soil. However, it is now obvious that the increase of P-diffusion coefficients with P concentration (Fig. 2 and 4) is associated with the decrease in b (Fig. 3). The buffer behavior of soil P is different depending on whether P is being adsorbed or desorbed. At $x = 0$, soil P concentration (HCl soluble) is 480 mg P kg^{-1} , and b is <50 when P is being adsorbed but >500 when being desorbed. This abrupt change in P buffer power is the reason for the discontinuity of the concentration-distance curve (Fig. 1) and of the effective diffusion coefficients (Fig. 2 and 4) at $x = 0$. Therefore, it can be concluded that the effective diffusion coefficient is primarily dependent on soil buffer power and not on soil P concentration. This important relationship is expressed by Eq. [6].

The shape of the sorption or desorption curve determines the influence of soil P concentration on the D_e . With a linear sorption curve, i.e., b is a constant, concentration has no influence on D_e . Deviation from linearity, which is the case (Fig. 3), causes b to vary with concentration and thereby also causes the D_e to vary as seen from Fig. 2 and 4. In agreement with this, Vaidyanathan et al. (1968) explained changes in K-diffusion coefficients with the curvature of the sorption isotherm.

Because of the crucial influence of b on D_e , b has to be determined accurately. For example, when the buffer curve is established, the process of adsorption or desorption that occurs during diffusion must be observed. Time of reaction may also be important, as has been discussed by Nye and Tinker (1977, p. 54) in terms of hysteresis and relaxation. Rowell et al. (1967) and Vaidyanathan et al. (1968) obtained similar agreement between calculated and measured D_e values as that found in our study. Others (Warncke and Barber, 1973; Hira and Singh, 1977) found much larger discrepancies, which may be due to difficulties in measuring the buffer power.

The importance of realistic values of b can also be seen from the calculation of \bar{D}_c . As shown in Fig. 4, calculated \bar{D}_c was in fair agreement with measured \bar{D}_c . As a parameter of calculation, the \bar{b} was used. If, as is often done, a simple average buffer power, obtained by dividing total change in soil P ($400 \text{ mg kg}^{-1} \times 1.46$) by total change in solution P (6.3 mg L^{-1}), had been used, the calculated \bar{D}_c would have been higher by a factor of 2.4 and would thus markedly deviate from measured \bar{D}_c . The deviation of measured and calculated \bar{D}_c at high P concentration (Fig. 4) might also have been caused by problems in determining b . Provided that b is accurately measured, it thus appears that calculated \bar{D}_c values represent a realistic indicator of P mobility in soil. Calculation could, therefore, replace the tedious measurement of P diffusion and may elucidate reasons for variations of the diffusive flux.

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