

Cumulative and Residual Effects of Organic and Chemical Fertilizers on Chemical Properties and P Sorption-Desorption Reactions in a Calcareous Soil: II. Phosphorus Desorption Kinetics

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Abstract: The kinetics of phosphorus (P) desorption in soils is important from plant nutrition and environmental perspectives. An experiment was conducted on the field plots that received 100 t ha⁻¹ of solid dairy manure (DM), sewage sludge (SS), or urban solid waste compost (UC) and one rate of chemical fertilizer (CF) for 1, 3 or 5 consecutive years to investigate the residual and cumulative effects of organic and chemical fertilizers on kinetics of P release. The kinetic data was best described by parabolic diffusion equation as evidenced by the relatively higher values of determination coefficient (R^2). Application of both chemical and organic fertilizers increased the values of P diffusion coefficients of soils (R), the initial (Dr_{in}) and final (Dr_f) P release rate indices and the values of P release rate constants (a and b) obtained from the power function equation as compared to the control (C). The largest residual and cumulative effects of treatments on the above parameters were also observed in the plots received SS and DM, respectively. Application of both chemical and organic fertilizers decreased $1/\beta$ constant as compared to the control. Adding DM and SS to soil increased the quantity of soil desorbable P after 0.25 (Q_{in}) and 72 h (Q_f). With the exception of a , that did not show significant relationships with calcium carbonate equivalent (CCE), the CCE content was positively correlated with the other rate and quantity parameters. The organic carbon (OC) content showed close relationships with Q_{in} , Q_f , Dr_{in} , Dr_f and R parameters.

Key words: Cumulative and Residual Effects • Organic Fertilizers • Phosphorus Desorption Kinetics • Calcareous Soil

INTRODUCTION

The rate of P reaction with soil influence the fate of P fertilizers added to soils, release of P in the rhizosphere, and equilibrium between P on soil solid phases and in percolating water. Many soil chemical processes are time-dependent. In general the release of P from soils is initially fast and then proceeds slowly for a long time with no apparent end point [1]. The initial faster reaction corresponded to the rapid dissolution of poorly crystalline or amorphous P in the soils, which were metastable and ultimately would be converted to crystalline forms such as octacalcium phosphate and calcium hydroxyl apatite [2]. The slower second reaction may be ascribed to desorption of surface labile P and slow dissolution of crystalline phosphate compounds in the soils [3].

Overall, P release from different soils is controlled by dissolution of P minerals and desorption of sorbed P [4]. The dissolution of P minerals is dominate at long time, while desorption of sorbed P is limited at short time. The rate of P release depends on mineral surface and P sorption mechanism. The P release is controlled by Fe and Al content and percentage of clay in acidic soils and by CaCO₃ equivalent and percentage of clay in calcareous soils [5, 6].

Phosphate desorption data have been described by a variety of equations. Successful reaction-controlled kinetic models include first order [7] and second order [7, 8]. Diffusion-controlled kinetic models have also been used [9]. Other kinetic models include Elovich [6, 10] and power function equations [11, 12]. Several researchers have compared the ability of reported kinetics models to describe P desorption data of soils [5, 13, 14].

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These studies suggest that several equations describe soil P desorption data and that there is no best equation for all studies and soils. Nevertheless, little effort has been devoted in the literature to identifying the types of soil for which each kinetic equation works best.

Environmental concerns have caused many states to implement laws regarding land application of P to minimize the potential for P transport into agricultural runoff. Earlier studies of P desorption kinetics focused primarily on describing P availability for plants [15, 16]. In addition to plant availability, it is also important to understand the kinetics of P release from soils that have excess concentrations of P loading in order to better manage soil P release to runoff water [17]. Overall although there has been extensive researches regarding availability and cycling of P in soils, some questions remain unresolved. In view of this, the objectives of this study were to i) find the kinetic models describing P release rate on calcareous soils treated with different organic and chemical fertilizers, ii) determine the accumulative and residual effects of a long-term (5 years) application of 3 different organic fertilizers and one rate of chemical fertilizer on P release rate parameters, and iii) investigate the relationships between P release kinetic parameters and soil properties.

MATERIALS AND METHODS

Soils: The field location, application of organic and chemical fertilizers, experimental design and characterization of soil and the amendments are described in part I of this work as presented by the same authors ().

Phosphorus Release Experiments: The soil samples with 1, 3 and 5 years history of receiving different amendments were placed in plastic containers. Phosphorus was added to the soils at the rate of 100 µg P .g⁻¹ as KH₂PO₄. The whole sample was then thoroughly mixed, and incubated at 20°C and field capacity water content for 30 d. The moisture was maintained by daily weighing the containers and making up the loss of water. Soil samples were drawn after 30 days of incubation, dried, ground and used for studying the kinetics of P desorption. Five g sub-samples of each soil were accurately transferred to 100-ml polyethylene tubes along with 40 ml of 0.01 M CaCl₂ solution. These samples were shaken for 12 different periods; 0.17, 0.5, 1, 3, 5, 10, 12, 18, 27, 37, 48 and 72 h. After shaking, the samples were centrifuged at 3000 rpm for 15 min and filtered through a 0.2 µm pore-size filter membrane. The P concentration in the extracts was measured by the Murphy and Riley [18] procedure.

Table 1: Equations tested to describe P release kinetic data [11]

Kinetic equation	Parameters
First order: $\ln Q_t = \ln Q_e - k_1 t$	k_1 , first-order rate constant (min ⁻¹)
Second order: $1/Q_t = 1/Q_e + k_2 t$	k_2 , second-order rate constant (mg P .kg ⁻¹) ⁻¹
Simple Elovich: $Q_t = 1/\beta (\ln \alpha\beta) + (1/\beta) \ln t$	α , initial P desorption rate (mg P .kg ⁻¹ min ⁻¹) β , P desorption constant (mg P .kg ⁻¹) ⁻¹
Power function: $Q_t = at^b$	a , initial P desorption rate constant (mg P .kg ⁻¹ min ⁻¹) ^b b , desorption rate coefficient (mg P .kg ⁻¹) ⁻¹
Parabolic diffusion: $Q_t = Q_e + R t^{1/2}$	R , diffusion rate constant (mg P .kg ⁻¹) ^{-1/2}

Q_t (mg P .kg⁻¹) is the amount of P released after t hours; Q_e (mg P .kg⁻¹) is the amount of P release at equilibrium.

The kinetic release data for the soils were evaluated using first order [7], second order [7, 8], Elovich [6, 10], power function [11, 12], and parabolic diffusion [9] equations (Table 1). The goodness of fit of each kinetic equation was evaluated according to its coefficient of determination (R^2) and the standard error (S.E.) calculated from:

$$SE = [\sum(Q_t - Q'_t)^2 / (n-2)]^{1/2}$$

Where Q_t and Q'_t are the measured and predicted amounts of desorbed P (mg P .kg⁻¹) at time t (min), respectively and n is the number of measurements.

The instantaneous P desorption rates at 0.25 (15min) and 72 h (4320 min) after the start of the experiment have been suggested by Raven and Hossner [19] as the initial and final P release rates indices. The rate of P release (mg .kg⁻¹ soil min⁻¹) after 0.25 h of desorption (Dr_m) was used as an index of immediate desorption rate, while that after 72 h (Dr_j) reflects the long-term desorption rate. These rate parameters were calculated from the slope of the P release at 0.25 and 72 h. The amount of released P (mg .kg⁻¹) at 0.25 (Q_m) and 72 h (Q_j) were also determined as quantity indices of P desorption kinetics.

RESULTS AND DISCUSSION

The soil and amendments characteristics as well as the effect of the amendments on soil chemical properties are presented in part I of this work ().

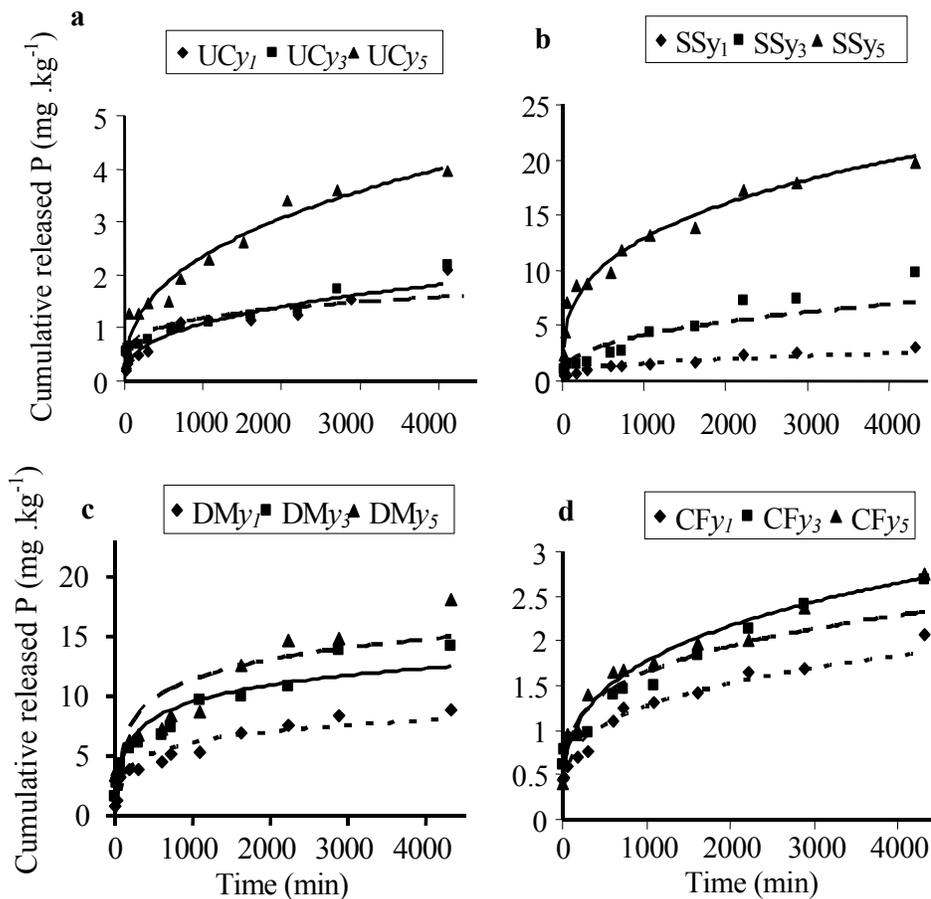


Fig. 1: Effect of number (years) of fertilizers' applications on the release of phosphorus on plots received UC (a), SS (b), DM (c) and CF (d) for 1 (y_1), 3 (y_3) and 5 (y_5) years.

Effects of Treatments on P Release Kinetics:

The phosphorus release patterns of the soils are presented in Fig 1 and 2. Release of P from the soils was rapid at first few hours and continued more slowly until an apparent equilibrium was approached. Similar characteristic curves of P desorption were also observed by Jeremy and Straw [3] and Abdu [5]. Jeremy and Straw [3] observed that for manure-amended soil, there was a sharp initial increase in P desorbed in the first hours followed by a continued slow release up to 504 h. Lookman *et al.* [20] studied P desorption from 36 Belgian and 8 German top soils. Their results showed that the rate of P desorption did not reach a plateau until after 1600 h, suggesting that true P desorption equilibrium might take more than 1600 h for some soils. McDowell and Sharpley [12] believed that P desorption and diffusion from inside of soil particles probably was the rate limiting steps in short- and long-term P release reactions, respectively.

The release of P increased with the number (years) of applications in all of the treatments (Fig 1). The magnitude of the increase was largest for SS and smallest for CF (Fig 1b,d). The total amount of released P ($\text{mg} \cdot \text{kg}^{-1}$ soil) after 72 h was greater in the plots received DM and SS compared to those received UC and CF (Fig 1 and 2). The larger effect of DM and SS on kinetics of P desorption from soil was probably due to the higher organic matter content of DM and SS as compared to UC as well as to the differences in the nature of organic matter resulting from differences in their production procedures. Organic matter may compete with P for sorption on the soil particle surfaces through blocking the adsorption sites and therefore may enhance the rate of P desorption. Additionally dairy manure and sewage sludge had the largest residual and cumulative effects on kinetics of P desorption from soil (Fig 2a,c) probably because of the greater residual and cumulative effects of these treatments on organic matter content of soil as compared

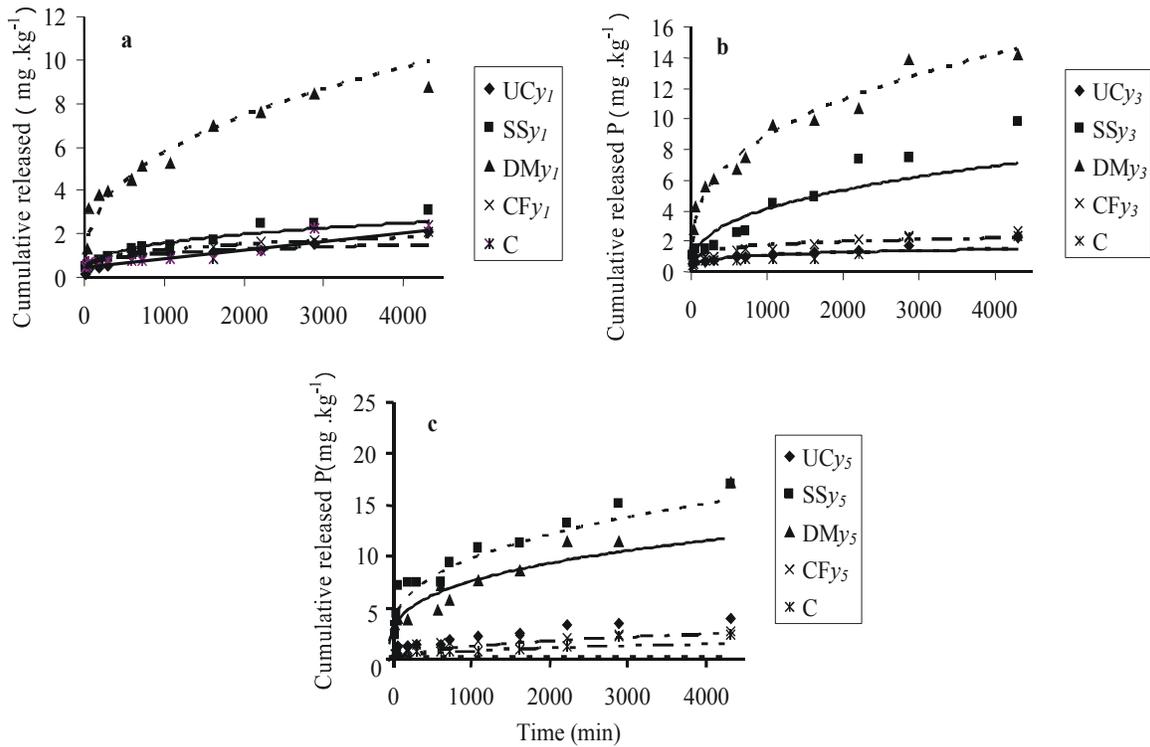


Fig. 2: Effect of the kind of fertilizer on the release of phosphorus on plots received UC, SS, DM and CF for 1 (a), 3 (b) and 5 (c) years.

to UC treatment. The kinetics of P desorption is affected by the solubility of P minerals in the soil. Application of organic material to soil may increase phosphorus solubility by soluble carbon compounds competing with P for sorption sites. Organic acids found in soil solution have been shown to inhibit the precipitation of HA and to stabilize the less insoluble Ca-P minerals OCP and DCPD [4, 21]. Speciation studies by Hansen [22] showed that the P being released from the soil is primarily orthophosphate. A possible mechanism of inhibition and stabilization of high energy Ca-P minerals is the adsorption of organic acids onto crystal surfaces, blocking sites acting as nuclei for new crystal growth [4, 23]. In soils, inhibition may be enhanced by complexation of Ca with the organic matter which has a high affinity for cations thus making it unavailable for forming well crystallized minerals. Toor and Bahl [13] also reported a higher desorption in poultry manure treated samples as compared to the untreated samples. Laboski and Lamb [24] compared P availability for swine manure- and phosphate fertilizer-amended soil after several months of incubation and found a significant increase in P availability (soil-test P) in the manure-amended soils compared with the phosphate fertilizer-amended soils.

Laboski and Lamb [24] postulated that the organic acids in the manure prevented P sorption on the soils and thus enhanced P availability.

Comparison of Kinetic Equations to Describe P Release

Data: The coefficients of determination (R^2) and the standard errors of estimate (S.E.) for kinetic equations tested to describe the P release data are presented in Table 2. Preliminary graphical tests (not shown) indicated that first and second order models did not adequately describe the P desorption data. Similar results were obtained by Raven and Hossner [19], who observed that a plot of the P release versus predicted by the first and second order equations showed a poor fit of these models. The lack of fit of first order equation to P-release data has been ascribed to more than one first order reaction taking place simultaneously: a fast reaction is taking place early, followed by an intermediate rate reaction and, finally, a slower reaction takes place, which continues for the duration of the experiment [11].

The P desorption was best described by the parabolic diffusion ($R^2=0.82-0.99$), closely followed by the power function ($R^2=0.84-0.97$). Toor and Bahl [13] reported that the Elovich and the parabolic diffusion equations were

Table 2: Coefficients of determination (R^2) and standard errors of estimates (S.E.) for kinetic equations used to describe the P release data

Treatments	Elovich		Power function		Parabolic		Second order		First order	
	S.E.	R^2	S.E.	R^2	S.E.	R^2	S.E.	R^2	S.E.	R^2
UCy ₁	0.69	0.85***	0.66	0.97***	0.65	0.95***	1.45	0.38*	0.62	0.65**
UCy ₃	0.73	0.85***	0.70	0.91***	0.60	0.99***	1.47	0.83***	0.56	0.93***
UCy ₅	1.77	0.94***	1.55	0.91***	1.60	0.91***	2.95	n.s.	1.95	0.72***
SSy ₁	1.34	0.80***	1.24	0.93***	1.07	0.97***	3.08	0.65**	0.39	0.90***
SSy ₃	2.01	0.70***	41.43	0.87***	1.62	0.95***	5.08	0.67***	2.77	0.86***
SSy ₅	5.00	0.93***	4.15	0.95***	3.82	0.95***	4.46	n.s.	3.69	0.69***
DMy ₁	1.93	0.93***	2.29	0.97***	2.14	0.94***	3.99	n.s.	3.19	0.51**
DMy ₃	3.02	0.91***	2.87	0.97***	2.78	0.96***	15.70	n.s.	4.27	0.60**
DMy ₅	3.15	0.82***	2.42	0.93***	2.08	0.97***	28.98	0.61**	3.49	0.82***
CFy ₁	0.48	0.88***	0.42	0.96***	0.41	0.98***	0.77	0.57**	0.4	0.73***
CFy ₃	0.66	0.82***	0.55	0.88***	0.55	0.94***	0.82	0.69***	0.65	0.87***
CFy ₅	1.25	0.95***	1.14	0.95***	1.08	0.89***	5.05	n.s.	0.66	0.74***
C	0.70	0.76***	0.69	0.84***	0.68	0.82***	0.68	0.78***	0.63	0.89***
Means	1.75	0.86	4.62	0.93	1.47	0.94	5.73	0.53	1.79	0.76

* P = 0.05, **P = 0.01, ***P = 0.001, n.s. is non-significant.

Table 3: P release rate parameters derived from the kinetic equations fitted the P release data and the quantity of P released (mg. kg⁻¹ soil) after 0.25 or 72h

Treatments	Parabolic	Elovich		Rate and quantity parameters of P desorption				Power function	
	R	$1/\beta$	α	Dr_m	Dr_f	Q_m	Q_f	a	b
UCy ₁	0.03	0.02	0.27	0.03	0.16×10^{-4}	0.26	2.08	1.45	0.37
UCy ₃	0.02	0.30	0.14	0.02	0.11×10^{-4}	0.59	2.88	1.18	0.16
UCy ₅	0.08	0.09	0.34	0.08	0.47×10^{-4}	0.44	5.7	1.51	0.41
SSy ₁	0.04	0.04	0.35	0.04	0.24×10^{-4}	0.51	6.08	1.36	0.31
SSy ₃	0.14	0.06	1.27	0.14	8.33×10^{-3}	1.17	9.80	1.45	0.37
SSy ₅	0.28	0.48	2.54	0.28	1.64×10^{-2}	3.45	29.92	1.37	0.31
DMy ₁	0.12	0.14	1.31	0.12	0.73×10^{-4}	1.03	8.78	1.49	0.40
DMy ₃	0.19	0.23	2.00	0.19	11.55×10^{-3}	2.15	14.14	1.40	0.34
DMy ₅	0.25	0.37	2.01	0.25	1.51×10^{-2}	3.62	20.28	1.34	0.29
CFy ₁	0.03	0.05	0.26	0.03	1.53×10^{-3}	0.45	2.08	1.31	0.27
CFy ₃	0.04	0.18	0.23	0.04	0.23×10^{-4}	0.7	3.26	1.28	0.25
CFy ₅	0.03	0.09	0.31	0.03	1.91×10^{-3}	0.53	5.44	1.32	0.28
C	0.01	1.00	0.10	0.01	0.72×10^{-5}	0.50	2.40	1.14	0.13

the best fitted equations. They showed that, the linear relationship between the cumulative P desorbed and $t^{1/2}$ was observed after only 8 h of equilibration which indicated that diffusion-controlled process became dominant after 8 h extraction as P concentration declines at later stage (because of the higher initial P extraction). With decline in the P concentration and corresponding increase in the concentration gradient, the P desorption becomes a diffusion-controlled process [9, 25]. Raven and Hossner [19] reported that the use of power function improved the fit of the data with respect to the parabolic diffusion equation in all tested soils. Close fit with power function has also been reported for P desorption from soils and minerals of variable chemistry and determined by different methodologies [14, 19].

It is generally believed that there is no single equation that describes equally well the P desorption kinetics of all soils. The fit for each treatment used in this study was about equal or better with the parabolic and power function equations than any other tested model (Table 2).

Effect of Chemical Fertilizers on P Desorption Kinetic Constants:

The P release rate parameters derived from the kinetic equations fitted the P release data are shown in Table 3. Since the parabolic diffusion was selected as the best equation describing the kinetic data, the initial (Dr_m) and final (Dr_f) P release rate indices were calculated from the slope of this equation $t=15$ min (0.25 h) and $t=4320$ min (72 h), ranged from 0.01 to 0.28 and

Table 4: Linear correlation coefficients between the P release rate and quantity parameters

	$1/\beta$	a	b	R	Dr_m	Dr_f	Q_m	Q_f
$1/\beta$	-	n.s.	n.s.	0.98***	0.98***	0.98***	0.92***	0.94***
a		-	0.99***	n.s.	n.s.	n.s.	n.s.	n.s.
b			-	n.s.	n.s.	n.s.	n.s.	n.s.
R				-	1***	1***	0.95***	0.95***
Dr_m					-	1***	0.95***	0.95***
Dr_f						-	0.95***	0.95***
Q_m							-	0.95***
Q_f								-

***P = 0.001, ^{ns} is non-significant.

Table 5: Linear correlation coefficients between the P release rate and quantity parameters and selected properties of the tested calcareous soils

Parameters	OC (%)	CaCO ₃ eq. (%)	CEC (Cmol+. kg ⁻¹)
$1/\beta$	0.75**	-0.62*	0.79**
a	n.s.	n.s.	n.s.
b	n.s.	n.s.	n.s.
R	0.84***	-0.69**	0.87***
Dr_m	0.84***	-0.69**	0.87***
Dr_f	0.84***	-0.69**	0.87***
Q_m	0.81**	-0.67**	0.79**
Q_f	0.84***	-0.82**	0.88***

OC, organic carbon; CaCO₃ eq., calcium carbonate equivalent; CEC, cation exchange capacity. * P = 0.05, **P = 0.01, ***P = 0.001, ^{ns} is non-significant.

0.00072 to 0.0164 mg P .g⁻¹ soil min⁻¹, respectively (Table 3). In fact, the mean rate of P release from soils at t=0.25 h was about 14 times as much as the P release rate at t=72 h. Application of both chemical and organic fertilizers increased the values of P diffusion coefficients of soils (R) and the initial (Dr_m) and final (Dr_f) P release rate indices as compared to the control. The magnitude of the increase was proportional to the type of fertilizer and the number of years fertilizers were applied. The increase was larger for organic fertilizers treated samples than CF treated samples due to the higher organic matter content of these fertilizers as compared to CF. The rate of P release from soils is affected by the solubility of P minerals in the soil. Application of organic matter to soil may increase phosphorus solubility by soluble carbon compounds competing with P for sorption sites and therefore may enhance the rate of P desorption. The high positive correlation between the OC content of the soil and R , Dr_m and Dr_f (Table 5) may demonstrate this idea. The largest residual and cumulative effects of treatments on the above parameters were also observed in the plots received DM and SS, respectively. The UC treatment indicated an intermediate effect in this respect.

Application of both chemical and organic fertilizers decreased $1/\beta$ constant as compared to the control dependent on the number of years (times) the fertilizers were applied and treatment type. Urban waste compost had the largest and dairy manure the smallest residual effect on $1/\beta$ parameter. Additionally application of UC and CF had the largest and SS the smallest cumulative effect on this parameter.

Adding DM and SS to soil increased the quantity of soil desorbable P after 0.25 (Q_m) and 72 h (Q_f) as compared to the control. Long-term application of UC and CF also increased the amount of released P at 0.25 (Q_m) and 72 h (Q_f) as compared to the control. The increase was proportional to the type of fertilizer and the number of years fertilizers were applied and was much larger for SS and DM as compared to UC and CF treatments. The residual effect of SS application on these parameters was smaller and its cumulative effect was larger than DM.

Application of both chemical and organic fertilizers increased the amounts of P release rate constants a and b obtained from the power function equation as compared to the control (Table 3). Addition of DM to soil had a greater residual effect on these constants in comparison with the other treatments. The largest increase in a and b constants was observed in the plots received UC for 5 consecutive years.

With the exception of b obtained from the power function equation, all of the P release rate and quantity parameters were affected by the OC content of soil (Table 5). Hence, soils containing more OC release more P in a given period of time and therefore show higher rates of P release as compared to soils containing lower amounts of these constituents. Overall, the largest P release rate and quantity parameters were observed in the plots received SS and DM. This may be due to the higher organic matter content of DM and SS as compared to UC. Toor and Bahl [13] observed higher R , b and β constants in soils treated with poultry manure (PM) rather than

the soils without PM. They found that the PM helped in maintaining the crystalline products of phosphate and calcium phosphate in metastable or poorly crystalline forms. In the present investigation, the DM and SS appeared to have played similar roles in soils. While in the acidic soils it might have complexed Fe and Al ions.

Relations among P Release Kinetic Parameters and Soil Properties: Linear correlation coefficients between the P release rate and quantity parameters are given in Table 4. Positive and significant correlations were observed between the overall diffusion constant (R) and the initial (Dr_{in}) and final (Dr_f) P release rate indices. These correlations support the hypothesis that P release is a diffusion-controlled process in the tested soil. Similar results were obtained by Garcia-Rodeja and Gil-sotres [26], McDowell and Sharpley [12] and Abdu [5]. They stated that the diffusion process became dominant at later stage of equilibrium because P concentration declines and P desorption becomes a diffusion-controlled process.

As shown in the Table 5, the OC content and CEC of soil showed close relationships with some P release rate and quantity parameters such as Q_{in} , Q_f , Dr_{in} , Dr_f and R (parabolic diffusion). Toor and Bahl [13] found that the values of R , b and β were generally higher in the soils treated with poultry manure. A higher P desorption in biogas-slurry treatments rather than the treatment without biogas-slurry, was also reported earlier by Steffens [27].

With the exception of a (power function) that did not show significant relationships with calcium carbonate equivalent (CCE), the CCE content was positively correlated with the other rate and quantity parameters (Table 5), indicating that calcium carbonate equivalent may control the P release rate in the soil we investigated. Although it is unlikely that the mineral phases in the soil are pure and well crystallized, results similar to those obtained in this study, were reported by Cooperband and Good [4] and Jeremy and Straw [3]. Sharpley [6] also reported a direct and significant relationship between CCE and Elovich $1/\beta$ parameter for some calcareous soils. Raven and Hossner [19] showed that, the amounts of desorbed soil P and desorption rates in the Udic Pellusert were larger than the calcareous soils. Jeremy and Straw [3] suggested that, Ca is a predominate factor in the speciation of P in neutral to alkaline soils and that the less insoluble calcium phosphate minerals, such as β -TCP or OCP control the concentration of P in solution. The stability of these P minerals is enhanced by the presence

of OM whereas organic acids can inhibit the formation of HA and stabilize OCP, and the OCP can have varying degrees of crystallinity, depending on the precipitation condition [3, 4].

CONCLUSIONS

Phosphorus desorption from organic fertilizer-amended soils began with a first initial reaction, followed by a slow secondary reaction, continuing up to 72 h. The P desorption was best described by the parabolic diffusion closely followed by the power function equations. Application of both chemical and organic fertilizers increased the values of P diffusion coefficients of soils (R), the initial (Dr_{in}) and final (Dr_f) P release rate indices and the values of P release rate constants (a and b) obtained from the power function equation as compared to the control. The largest residual effect of treatments on the above parameters were also observed in the plots received DM. Application of both chemical and organic fertilizers decreased $1/\beta$ constant as compared to the control. Adding DM and SS to soil increased the quantity of soil desorbable P after 0.25 (Q_{in}) and 72 h (Q_f) as compared to the control. With the exception of a (power function) that did not show significant relationships with calcium carbonate equivalent (CCE), the CCE content was positively correlated with the rate and quantity parameters. The OC content and CEC of soil showed close correlations with some P release rate and quantity parameters such as Q_{in} , Q_f , Dr_{in} , Dr_f and R (parabolic diffusion).

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