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RESEARCH ARTICLE

PHOSPHORUS FORMS AND SORPTION CHARACTERISTICS IN SOILS OF A TRANSECT OF KOHORA WATERSHED OF ASSAM, INDIA

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ABSTRACT

Phosphate sorption relating to soil properties in soils of Kohora watershed of Assam, India was studied. The soils were acidic in reaction with varying texture. Clay and CEC increased concomitantly down the slope. In all soils, Fe-P dominated over other P fractions contributing 29.9%. Phosphate adsorption data fitted better to Freundlich equation. Medium land soils had greatest affinity for P yielding highest sorption maxima (b = 2044.03 μg P g-1) and Freundlich K. Langmuir 'b' was significantly correlated with Fe_d, Al_d, Fe_2O_3+Al_2O_3, Al-P and total-P while negatively with pH, OC and available P fractions. Bonding energy was greatly affected by Fed (r = 0.683*) only while maximum phosphate buffering capacity (MPBC) was typically affected by Al and Fe oxides. Phosphorous supply parameters of the soils increased unevenly with equilibrium P concentration. Negative ΔG values indicated spontaneous adsorption processes. Results suggest that these soils require higher external P for maintaining optimal solution concentration.

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INTRODUCTION

Serving as the principal reservoir of P in terrestrial systems, soils contain P ranging from 35 to 5,300 mg kg^{-1}, with a median concentration of 800 mg kg^{-1} (Bowen 1979; Sparks 2003). However, a large portion of P in soils is not available to growing plants. Moreover, the use of such available P is very limited because of its fixation by soil minerals, such as Fe and Al oxides, hydroxides and oxyhydroxides (Wang *et al.,* 2013) and 1 : 1 layer lattice clays particularly in acid soils. Because of the polyprotic character of this ligand, phosphate adsorption takes place over a wide pH range. It has a maximum at low pH and gradually decreases with increasing pH. In addition, the presence of organic acids such as humic acid, fulvic acid, and citric acid on the surface of (hydr) oxides could inhibit phosphate adsorption through site competition, electrostatic effects, and steric hindrance (Borggaard *et al.,* 2005; Weng *et al.,* 2008). Phosphate sorption was correlated not only with the content of amorphous aluminum (Al) and iron (Fe), but with the clay percentage of soils (Quintero *et al.,* 1999). Other soil characteristics such as organic matter content, exchangeable cations, ionic strength, redox potential and temperature may also influence P-sorption (Curtin *et al.,* 1992; Agbenin and Tiessen 1994; Quang *et al.,* 1996).The inter-relationships among the various P fractions in soil are complex. However, information about these fractions can increase our understanding of the P sinks and sources of P in soil and are essential for an efficient P management programme. To better understand P availability in soil, many workers have conducted P fractionation studies (Sui *et al.,* 1999; Maquire *et al.,* 2000; Scherer and Sharma 2002). Phosphate sorption capacity was accurately predicted from the concentrations of oxalate-extractable Al and Fe, as is generally the case with acid soils (Horta and Torrent 2007). Hossain *et al.,* (2012) observed that soils possessing higher amounts of free iron oxide and clay sorbed more phosphate from applied phosphorus. Hence phosphorous deficiency is very common in countries with limited access to fertilizers whilst it is particularly acute in soils in the humid tropics because these soils are often rich in metal oxides and hydroxides that form strong chemical bonds with phosphate anions. The concentration of orthophosphate-P in the soil solution is basically controlled by adsorption/desorption and precipitation/dissolution processes and also by the immobilization and mineralization of organic P forms ((Frossard *et al.,* 1995; Sanyal and de Datta 1991; Condron *et al.,* 2005; Pierzynski *et al.,* 2005). The sorption of phosphate by soil is conventionally described by so-called quantity/intensity (Q/I), P-sorption curves, or isotherms, which are usually described with the Langmuir or Freundlich equations. Both Langmuir and Freundlich isotherms gave a good fit with the P adsorption data (Sathiyamurthi 2013). However, the same author reported that affinity parameter of P adsorption had negative significant correlation with soil clay particle (r = -0.895) and negative correlation with soil $CaCO_3$ (r= -0.640). But it had positive non-significant correlation with

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extractable soil Al (r=0.462). Conversely, isotherms of different Fe and Al minerals showed better conformity to the Freundlich model than the Langmuir model because of their surface heterogeneity (Wang *et al.,* 2013). By applying various models, Khan *et al.,* (2010) observed better fit of P adsorption data with Freundlich equation (R^2 = 0.99) over the Langmuir equation (R^2 =0. 97) in different soil series. The dynamics of P in soil is complex and it is difficult to establish general rules for the behaviour of P and so it is important to collect basic local data to determine adequate fertilizer practice. Indeed, routine tests for assessing P availability in soil should always refer to norms that depend on soil type. In this respect, P sorption-desorption data related to soil properties are especially helpful (Singh and Gilkes 1991). Such studies have been carried out in many regions and on different soil types of the humid tropics (Sanyal *et al.,* 1993; Tomar 2000) but no information exits on the P-sorption characteristics of soils of Assam particularly in the context of a watershed transect. The objectives of this research are to investigate the sorption properties of phosphate in acid soils under different topographic locations of Kohora watershed of Assam, India. In addition, we characterize the soils in the watershed with respect to physicochemical properties and P forms. This research could provide significant insights for controlling P bioavailability and explaining the pedo-chemical processes of P sorption in soil.

MATERIALS AND METHODS

Soil characterization

The Kohora watershed of Assam lies between $93^0 20`$E to $94^0 25`$E longitude and $26^0 30`$N to $25^0 45`$N latitude covering a part of Karbi Anglong and Golaghat districts of Assam and includes a part of Kaziranga National Park. The study area covers a part (transect) of Kohora watershed under Karbi Anglong district with a geographical area of 395 hectare having humid sub-tropical climate with mean annual rainfall of 2246 mm. The watershed along with crop and water resource management constraints consists of various soils related problems. Out of these, P-fixation emerged as a major problem owing to higher Fe and Al-oxides and clay content of the soils. Based on contour information available in the toposheet and FAO guidelines (FAO, 2006) and local modifications, eight land situations viz. hillock top (crest 0-5% slope), upper slope (30-35% slope), middle slope (15-30% slope), lower slope (10-15% slope), flatter land (moderately sloping, 5-10% slope), upland (gently sloping, 2-5% slope), medium land (very gently sloping,1-3% slope) and low land (level to nearly level, 0-1% slope) were identified in the transect in relation to vertical height in the toposheet to the distance on the ground. Major area is under nearly level to moderately sloping land (80.9% of total geographical area, TGA) supporting agriculture and wasteland. Other land forms are under degraded forest with severe erosion and occupy 20.1% of TGA. Eight representative surface soil samples varying in physico-chemical properties were collected from the transect. Processed soil samples (<2mm) were analyzed for different physico-chemical properties following standard procedures. Soil pH was determined on a 1:2.5 soil-to-water suspension after 30 min equilibrium with a combined glass electrode. Mechanical composition of soil was determined by the International Pipette method (Piper 1966) and cation exchange capacity (CEC) and exchangeable bases were determined by 1 mol L^{-1} ammonium acetate (pH 7.0) (Jackson 1973). Soil organic matter was determined by the H_2SO_4–potassium dichromate ($K_2Cr_2O_7$) oxidation method (Walkley and Black 1934). Free Fe_2O_3 and Al_2O_3 were extracted by sodium dithionite and citrate buffer (Coffin 1963); Fe and Al in the extract were determined by AAS (AA-203D, Chemito Ltd). Phosphorus fractionation in triplicate samples was done by the modified procedure of Chang and Jackson (1957) as modified by Peterson and Corey (1966) and reported by Page *et al.,* (1982). Total and organic P was determined by the NaOH digestion method (Mehra *et al.,* 1954). Available P was determined by extraction with Bray and Kurtz No. 1 and Bray and Kurtz No. 2 methods (Bray and Kurtz, 1945), Olsen's $NaHCO_3$ (Olsen *et al.,* 1954). Phosphorus in the extract was determined colorimetrically by the molybdenum blue method (Jackson 1973).

Batch Adsorption Experiments

For the batch adsorption studies, 5 g triplicate soil samples from each land situations were equilibrated with 50mL of 0.01M $CaCl_2$ solution as background electrolyte containing 5, 10, 20, 40, 60, 80, 100 and 200 µg P mL^{-1} as potassium dihydrogen phosphate (KH_2PO_4) in polyethylene tubes .The tubes were incubated at 25°C for 72 hours with 30 min shaking every day. Two drops of toluene was added to each tube to minimize the microbial growth. At the end of incubation, soil samples were centrifuged for 20 min at 8000 rpm and filtered through Whatman No 40 filter paper. The P in solution was determined colorimetrically by the molybdate blue colour method (Jackson 1973). The adsorption of added phosphate was calculated as the difference between the amount of phosphate added and the phosphate remaining in solution. The distribution coefficient (K_d) which indicates the distribution of phosphate between solid and solution phases at equilibrium was calculated as K_d = meq of P adsorbed per g soil/meq of P per mL of solution at equilibrium. Phosphate sorption data were fitted to the Langmuir as well as the Freundlich linear equations as follows:

Langmuir adsorption isotherm

$$C/(x/m) = 1/kb + C/b \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ (1)$$

Where C is the equilibrium solution P concentration (µg mL^{-1}), x/m is the weight of P adsorbed per unit weight of soil (µg g^{-1}), k is a constant related to bonding energy of P to the soil (mLg^1). A plot of C/(x/m) versus C gives a straight line. The constant b and k are obtained from the slope (1/b) and the intercept (1/kb), respectively.

Freundlich adsorption isotherm

$$\log x/m = 1/n \log C + \log K \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ (2)$$

Where x/m is the weight of P adsorbed per unit weight of soil (µg g^{-1}), C is the equilibrium solution P concentration (µg mL^{-1}), n and K are constants. A plot of log(x/m) versus log C gives straight line. The constant n and K are obtained from intercept (log K) and slope (1/n), respectively. The extent or the driving force of the adsorption reactions was mathematically described by determining the Gibbs free energy of the system through the following thermodynamic relationship (Silveira *et al.,* 1999; Dias *et al.,* 2003; Soares *et al.,* 2005):

$$\Delta G = RT (\log C_{eq} - \log C_0) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ (3)$$

Where ΔG is the variation in the free energy (J mol^{-1}), R is the universal constant of the gases (8.314 J $mol^{-1}K^{-1}$), and T is the absolute temperature (K). The amount of P adsorbed at 0.2 mg P L^{-1} was calculated from the adsorption isotherm x/m vs c. The maximum P buffering capacity (MPBC) was computed by multiplying adsorption maxima (b) with bonding energy (k) (Kuo *et al.,* 1988). The P supply parameter (SP) of soil was calculated by using the formula of Khasawneh and Copeland (1973) as:

$$SP = (q.c)^{1/2} (K_1.K_2)^{1/4} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

Where, q is the amount of P adsorbed (µg g^{-1}soil), c = equilibrium P concentration (µg mL^{-1}), K_1 and K_2 are constants, which are determined by plotting 1/q vs. 1/c, K_1 = reciprocal of intercept of 1/q vs. 1/c and K_2 = slope of plot multiplied by K_1.

Statistical Analysis

All reported values are expressed on an oven-dry soil weight basis. Statistical analysis of experimental data was done by CoStat Professional, Version 6.311, Copyright © 1998-2005 CoHort Software. CoHort Software, 798 Lighthouse Ave, PMB 320, Monterey, CA 93940, USA.

RESULTS AND DISCUSSION

Soil characteristics

Texturally the soils of Kohora watershed was skeletal sandy loam to sandy clay with clay content varying from 19.35 to 40.23 per cent (Table1). Clay fraction of the soils exhibited its accumulation toward lower topographic locations in the transect, the lowland possessing the highest amount; the sand fraction showed exactly opposite behaviour. This was because of washing off and transportation of finer particles by rain water from the hillock top (crest) to lowland and subsequent deposition in the lower position of the transect. Contrarily, distribution of silt fraction was erratic. Strong acidity with pH 4.7 to 5.3 was the common feature of the soils. Nevertheless, pH did not show any specific distribution pattern along the watershed transect. A significant negative correlation (Table2) of pH with dithionite extractable Al (Al_d) and free Fe_2O_3+Al_2O_3 revealed that soil reaction was basically governed by these oxides. Seats and Peterson (1964) reported that in acidic environment, Fe and Al hydrous oxides undergo hydrolysis with release of H^+ ions which results in drop of soil pH. Conversely, pH was fairly proportionate with CEC of the soils. Organic carbon (OC) content of all the soils was high (0.85 to 1.35%). The distribution pattern (Table1) of organic carbon in the transect showed an accumulation on the crest soil due to rich vegetation cover and gentle slope (0-5%). However, increase in slope (35%) downward enhanced swab of OC which was successively accumulated in the soils of lower slope to upland. The lesser content of OC in medium and lowland soils may be attributed to paddy cultivation under which alternate oxidation-reduction conditions promoted oxidation of organic matter and their subsequent decomposition (Ponnamperuma 1972). The total free Fe_2O_3+Al_2O_3 of these soils was quite high (2.41–4.28%) and this is due to Fe and Al rich parent material of the soil. Distribution of Fe_d and Al_d showed an increasing trend down the slope. The pH of the soils was significantly and negatively correlated with exchangeable Al, dithionite extractable Al and Fe and free Fe_2O_3+Al_2O_3 (r = -0.730, P =5%). This was due to increasing solubility of Fe and Al under condition of low pH (Bolt 1976). The exchangeable Al exhibited significant negative correlation with OC indicating that organic matter either forms insoluble complexes with Al^{3+} or blocks the exchange sites. The negative correlation of OC with Fe_d, Al_d and Fe_2O_3+Al_2O_3, (Table2) also indicated that organic matter inhibited crystallization of amorphous oxides of Fe and Al in these soils. These observations are consistent with those of Schwartmann (1966). The CEC of the soils varied from 5.6 to 11.2 (C mol (p+) Kg^{-1}) and increased from the crest to lowland soil (Table1) which is attributed to increase in the clay content of the soil. This observation was evidenced by a highly significant positive correlation between CEC and clay content of the soil (r = 0.906, P=0.01).

Phosphorus fractions

Results showed that the total P in the soils ranged from 478.4 to 542.8 mg Kg^{-1} with a mean of 523.7 mg Kg^{-1} (Table 4). The soils of the hillock top contained the highest amount of total P which decreased to the lowest value in the lower slope. It tended to increase towards the lower position of the transect. Amongst the different inorganic fractions of phosphate, Fe-P was the dominant fraction of phosphate, followed by Al-P and Ca-P was the lowest (Table 4).These three fractions ranged from 150.5 to 175 mg Kg^{-1}, 125.4 to 148.5 mg Kg^{-1} and 14.3 to 24.5 mg Kg^{-1}, respectively. The soils under lowland condition contained the lowest amount of Fe-P while under medium land situation its quantity was the highest. Fe-P constituted the highest percentage (29.1%) of total P while Al-P contributed about 26.5%. The inorganic P ranged from 289.4 to 413.9 mg Kg^{-1} with a mean of 388.4 mg Kg^{-1} and constituted 74.2% of total P, whereas the organic P varied from 109.9 to 127.8 mg Kg^{-1}(mean of 119.9 mg Kg^{-1}) and contributed only 22.9% to total P pool. The Fe-P, Al-P and occluded P fractions (Residual-P) contributed mostly to the total P. These results are in accordance with the findings of Singh and Datta (1987). Distribution of various inorganic P fractions as a constituent of total P in these soils followed the order: Fe-P (152.5%) > Al-P (138.7%) > Organic-P (119.9%) > Res-P (61.4%) > Ca-P (18.7%).

Organic-P of the soils ranged from 109.9 to 127.8 mg Kg^{-1} and did not show any definite trend of distribution within the transect. As expected, the organic-P was positively and significantly correlated with organic carbon (Table 5). Available P in the soil was extracted by three extractants viz. Bray's I (Bray's I-P), Bray's II (Bray's II-P) and Olsen's (Olsen's-P) reagents. The Bray's I-P, Bray's II-P and Olsen's-P in the soil ranged from 12.39 to 21.35 mg Kg^{-1}, 13.29 to 23.35 and 15.38 to 24.21 mg Kg^{-1}, respectively (Table 4). The amount of available P in soil extracted by these three extractants followed the order: Bray's I-P < Bray's II-P < Olsen's-P. The amount of available P extracted by Bray'-I method (recommended for acid soils) in these soils was found to be within medium range. The latter two reagents extracted relatively greater amount of available P which was due to strong acids involved in Bray's-II reagent to solubilize Ca-P fraction and their influence on dissolution of Fe-P and Al-P (Thomas and Peaslee 1973). On the other hand, high pH in the Olsen's reagent extracted large amount of P by dissolution of inorganic P. Available phosphorus extracted by all the reagents distributed almost similarly in the watershed transects.

Table 1. Physico-chemical properties of the soil

Soils	Mechanical separates (%)			Texture	pH	OC (%)	Exch-Al [C mol (p+) Kg^{-1}]	CEC [C mol (p+) Kg^{-1}]	CBD* extractable (%)		Fe_2O_3 $+Al_2O_3$ (%)
	Sand	Silt	clay						Al_d	Fe_d	
Hillock Top (Crest)	61.23	18.29	20.48	Skeletal scl	4.7	1.35	0.85	6.5	1.01	1.05	3.41
Upper slope	63.59	17.06	19.35	Skeletal sl	4.8	0.98	1.42	5.6	0.95	0.99	3.32
Middle slope	55.01	22.37	22.62	Skeletal scl	5.2	0.95	1.69	8.4	0.63	0.85	2.41
Lower slope	51.95	23.51	24.54	Skeletal scl	5.0	1.20	0.75	9.5	0.79	0.82	2.67
Flatter land (moderately sloping)	49.11	21.34	29.55	scl	4.8	1.10	1.53	9.4	1.11	1.15	3.74
Upland (gently sloping)	47.92	16.63	34.45	sc	5.1	1.20	0.83	10.2	0.75	0.94	2.76
Medium land very gently sloping)	43.15	19.64	37.21	sc	4.9	0.83	1.55	10.9	1.28	1.30	4.28
Low land (level to nearly level)	40.43	19.34	40.23	sc	5.3	0.85	0.85	11.2	0.73	0.75	2.45

*Citrate-Bicarbonate-Dithionite

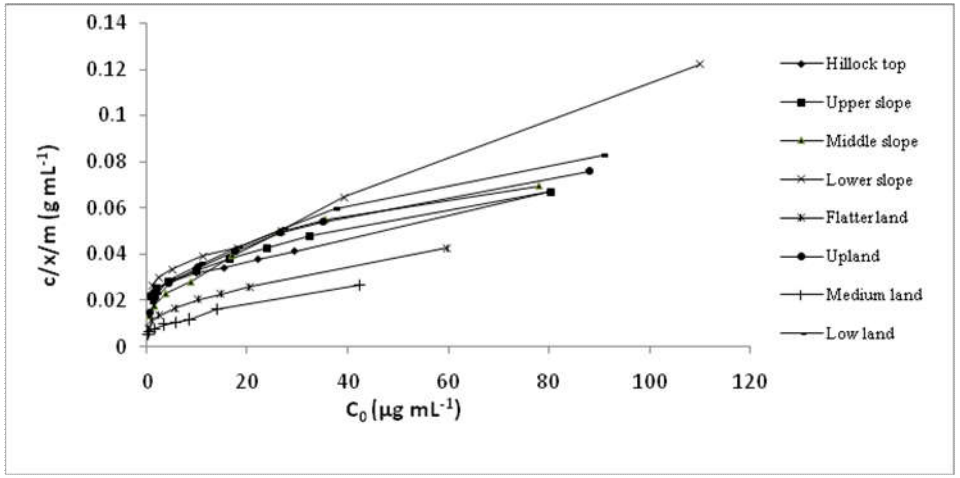

Fig. 1. Langmuir adsorption isotherms

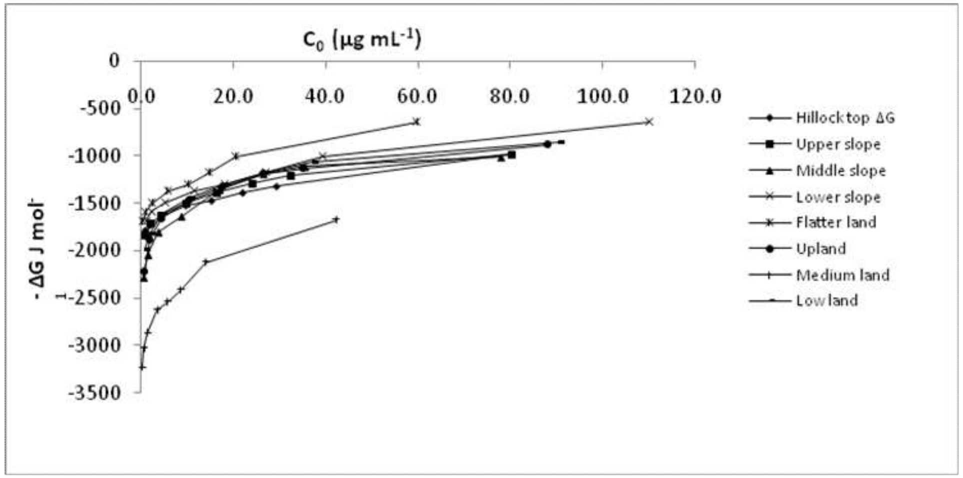

Fig. 2. Gibb's Free Energy (ΔG) vs Equilibrium P (C0)

It was observed that available P in the soil as extracted by Bray's I, Bray's II and Olsen's reagents was highly and significantly correlated with pH (r=0.916**, 0.9218**and 0.894**) indicating that available-P increased with increase in soil pH. On the other hand, they had significant but negative relationships with Fe_d, Al_d and $Fe_2O_3+Al_2O_3$ (Table 5). This suggests that available phosphate fractions in soil were largely influenced by different forms of Fe and Al and pH. The content and reactivity of oxides, hydroxides and oxyhydroxides of Fe and Al in many acid soils are the components that predominantly influence P adsorption because these oxides serve as the most effective P sorbents (Torrent 1987; Tek Chand and Tomar 1993). The oxides of Fe and Al are the residual products formed during silicate weathering and their amounts increase as weathering proceeds. Strongly weathered acid soils are, therefore, rich in Fe and Al oxides which are expected to cause decreased availability of P in acid soils. Nevertheless, it has been suggested that Fe-P and Al-P pool may maintain the levels of plant-available P through P desorption (Beck and Sanchez 1994; Zhang and Mackenzie 1997a). Similar observations were also made by Das (1999) in 14 acid soils of Assam.

Phosphate sorption

The absolute amounts of phosphate adsorbed (x/m) progressively increased while the percentages of added P adsorbed by different soils (X_{ad}) decreased with increasing P concentration in solution phase (Table 6). These observations are consistent with those of Tomar and Gautom (1996), Das (1999), Ahmed *et al.,* (2008) and Tek Chand and Tomar (2009). The soils under medium land situation adsorbed the largest percentage of added P (78.85 – 95%) while the soils under lower slope adsorbed the lowest (45 – 79%). At a given level of added P or equilibrium concentration, phosphate sorption (x/m) by soil under medium land situation in the transect was the highest vis-à-vis lower slope.

This is in conformity with the observed soil properties in medium land soil where Fe_d, Al_d and Fe_2O_3+Al_2O_3 were highest. The extractant citrate-bicarbonate-dithionite is known to estimate the total content of both amorphous and crystalline Fe and Al oxides in soils. Several workers reported that content of oxides, hydroxides and oxy-hydroxides of Fe and Al in many acid soils are the components that predominantly influence phosphate sorption (Singh and Glikes 1991; Das 1999; Tomer 2000, Wang *et al.,* 2013). The oxides of Fe and Al are the residual product of silicate weathering and their amounts increase as weathering proceeds. Therefore, strongly weathered acid soil, as evidenced in the present study, are rich in Fe and Al oxides which are expected to cause decreased availability of P in these soils. The correlation of X_{ad} with Fe_d, Al_d and free Fe_2O_3+Al_2O_3, were significant and positive at all levels of added-P except at 5 ppm level. This suggests that oxides and hydroxides of Fe and Al are the predominant sorbent of phosphate in these soils. These findings were in accordance with several earlier observations (Torrent 1987; Singh and Glikes 1991; Dey and Bhattacharyya 1996; Agbenin 2003) while Chanco and Dezzo (2004) evidenced organically bound forms of Al as the main component of P sorption in Venezuelan soils. It is interesting to note that clay exhibited non-significant positive correlations with distribution coefficient at lower added-P (r= 0.476) and the correlation decreased with increase in P level and became lowest at the highest level (r= 0.344). This suggests that at lower P concentration, phosphate had higher affinity for clay but with the increase P level, the oxides of Fe and Al (Fe_d and Al_d) became more strong adsorbent giving highly significant 'r' values between these oxides and x/m. This perception has further been supported by greater significant positive correlation between ΔG and Fe_2O_3+Al_2O_3 at maximum x/m (r= 0.846**) as against ΔG and clay (r= 0.305 NS). Phosphate sorption data were more satisfactorily described by Freundlich (R^2=0.99) than Langmuir (R^2=0.95) adsorption equations (Table 6).

Table 2. Exchangeable cations of the soils

Soils	Exchangeable cations of the soils [C mol (p+)Kg^{-1}]			
	Ca^{2+}	Mg^{2+}	K^+	Na^+
Hillock Top (Crest)	0.6	2.0	0.80	0.07
Upper slope	0.4	1.2	0.48	0.04
Middle slope	0.5	0.8	1.22	0.05
Lower slope	0.6	1.8	0.88	0.06
Flatter land (moderately sloping)	0.5	1.1	1.10	0.05
Upland (gently sloping)	0.8	1.0	1.22	0.08
Medium land (very gently sloping)	0.5	1.6	1.35	0.09
Low land (level to nearly level)	1.0	1.4	1.38	0.10

Table 3. Correlation co-efficient (r) amongst soil properties

Properties	pH	OC	CEC	Al_d	Fe_d	Fe_2O_3 +Al_2O_3
% clay	0.514	-0.491	0.909**	0.107	0.076	0.068
pH	1	-0.463	0.605	-0.740*	-0.693*	-0.075
OC		1	-0.395	-0.077	-0.054	-0.079
CEC			1	-0.027	-0.034	-0.069
Al_d				1	0.936**	0.990**
Fe_d					1	0.972**

*P<0.05, **P<0.01

Langmuir adsorption plot yielded curvilinear isotherms for the entire range of equilibrium P concentration in all the soils (Fig.1). The adsorption maxima (b) and bonding energy (K) values were the highest in medium land soil and they did not show any definite trend along the watershed transect. Since the K values reflect not only the binding strength of observable adsorbed phosphate but also that of native adsorbed phosphate, we suggest that native adsorbed phosphate in soils of upper part of the transect is less strongly retained and more likely to wash off with rain water. Lower K values in the lowland soil were indicative of releasing more easily the retained P into soil solution (Jiao *et al.,* 2006). The oxides and hydroxides of Fe and Al were proved to be the dominant factors that governed the adsorption maxima in these soils which was clearly indicated by significant positive correlation of adsorption maxima with Fe_d (r = 0.875 P= 0.01) and Al_d (r = 0.853 P= 0.01). Adsorption maxima decreased with increase in pH (r = -0.683 P= 0.05) as well as organic carbon (r = -0.113). Although, statistically non-significant correlation existed between organic carbon and adsorption maxima, the negative relationship still indicates that organic matter decreased P sorption (Tek Chand and Tomar 1993; Guppy *et al.,* 2005; Hoseini and Taleshmikaiel 2013) in these soils. Studied soils are highly weathered and the presence of organic matter reduces P- sorption capacity (Gillman *et al.,* 1989) due to direct result of competition for sorption sites between phosphate and organic ligands. It is also possible that organic matter reduces positive charge on variable charge surfaces by lowering pH, and this decreases the attraction of P to the soil surface, indicating that anthropogenic activities do alter soil properties. Similar observation was also evidenced by Chanco and Dezzo, (2004) and Tsado *et al.,* (2012). Phosphate exhibited greater affinity to CBD extractable Fe (K vs Fe_d r = 0.712 P= 0.05) than Al_d or Fe_2O_3+Al_2O_3. Langmuir adsorption maxima also had highly significant positive correlation with total-P and Al-P while negative with Bray's I-P, Bray's II-P and Olsen's-P (Table 9).

The maximum phosphate buffering capacity (MPBC) varied widely amongst the soils of the watershed (Table 7). The highest value of MPBC was observed in medium land while the lowest was in soils under lower slope, indicating that medium land soils will require higher P application for maintaining a desired P concentration in solution than the soils of lower slope. It is notable to mention that the CBD extractable Fe and Al, i.e. their free oxides and adsorption maxima (b) values were the highest in the former soil. The MPBC had highly significant positive correlation with these properties (Table 9) and non-significant negative correlation with OC (r= -0.465 NS) and positive with pH (r= 0.250 NS). Ahmed *et al.,* (2008) also observed highly significant positive correlation between P sorption maxima (b) and phosphate buffering capacity. The total, organic, Fe- and Al-P exhibited fair correlations with the MPBC in these soils. As mentioned earlier, the linear form of the Freundlich adsorption equation fitted better with the adsorption data for the soils than Langmuir adsorption equation (Table 7). The Freundlich constant 'K' and '1/n' did not show any definite trend along the watershed transect. Freundlich 'K' and '1/n' did not have any significant correlations with adsorption maxima (b) and bonding energy constant (k), while, the former had significant negative correlation with MPBC (r= -0.685 P= 0.05). Such type of association, however, suggests inter-dependence amongst the parameters. The Freundlich 'K' was significantly and negatively correlated with Fe_d and Al_d (Table 9) while positively with Al-P (r= 0.268, NS). On the other hand, '1/n' was positively correlated with Al_d and Fe_d while negatively with clay, pH, CEC, Fe_2O_3+Al_2O_3, Ca-P, residual-P and Bray's-I, Bray's-II P and Olsen's P.

Table 4. P-fractions in the soil (mg Kg^{-1})

Soils	Total-P	Org-P	Fe-P	Al-P	Ca-P	Residual P	Bray's- I P	Bray's- II P	Olsen's P
Hillock Top (Crest)	542.8	125.5 (23.1)	174.5 (32.1)	148.5 (27.3)	14.3 (2.6)	57.6 (10.6)	12.4 (2.3	13.4 (2.4)	15.4 (2.8)
Upper slope	529.4	115.4 (21.8)	172 (32.5)	144.0 (27.2)	15.5 (2.9)	58.9 (11.1)	14.6 (2.7)	14.9 (2.8)	17.8 (3.4)
Middle slope	507	109.9 (21.7)	154.0 (30.4)	132.2 (26.1)	18.5 (3.6)	61.8 (12.2)	19.6 (3.9)	21.8 (4.3)	24.2 (4.8)
Lower slope	478.4	127.8 (26.7)	156.7 (32.7)	135.4 (28.3)	19.5 (4.1)	61.0 (12.7)	18.9 (4.0)	19.6 (4.1)	21.6 (4.5)
Flatter land (moderately sloping)	535.3	119.3 (22.3)	174.3 (32.6)	142.15 (26.6)	18.1 (3.4)	60.8 (11.4)	13.6 (2.5)	15.6 (2.9)	18.3 (3.4)
Upland (gently sloping)	539	125.4 (23.3)	163 (30.2)	136.8 (25.4)	20.4 (3.8)	63.1 (11.7)	21.3 (3.9)	23.3 (4.3)	24.2 (4.5)
Medium land (very gently sloping)	536.2	113.7 (21.2)	175.0 (32.6)	145.5 (27.1)	19.2 (3.6)	59.2 (11.0)	15.0 (2.8)	16.6 (3.1)	18.1 (3.4)
Low land (level to nearly level)	521.4	122.5 (23.5)	50.5 (9.7)	125.4 (24.0)	24.5 (4.7)	68.7 (13.2)	20.3 (3.9)	21.6 (4.1)	22.3 (4.3)

* Figures in parentheses indicate per cent contribution

Table 5. Correlation (r) between Physico-chemical properties and P-fractions of the soils

Properties	Total-P	Org-P	Fe-P	Al-P	Ca-P	Residual P	Bray's- I P	Bray's- II P	Olsen's- P
%clay	0.213	0.09	-0.55	-0.418	0.830**	0.669*	0.413	0.459	0.327
pH	-0.404	-0.074	-0.742*	-0.947**	0.848**	0.866**	0.916**	0.921**	0.894**
OC	0.028	0.697*	0.437	0.379	-0.506	-0.406	-0.205	-0.225	-0.221
CEC	-0.125	0.127	-0.473	-0.528	0.866**	0.646	0.544	0.598	0.501
Al_d	0.516	-0.157	0.485	0.748**	-0.388	-0.579	-0.797*	-0.759*	-0.798**
Fe_d	0.606	-0.307	0.648	0.813**	-0.415	-0.645	-0.716*	-0.634	-0.654
$Fe_2O_3+Al_2O_3$	0.56	-0.233	0.566	0.820**	-0.441	-0.629	-0.790*	-0.739*	-0.767*

*P<0.05, **P<0.01

Table 6. Initial (C_o) and equilibrium concentration (C) of solutions (µg mL^{-1}), sorption of P (x/m) (mg g^{-1}) and distribution coefficient (K_d) of the soils

Soils	Co	5	10	20	40	60	80	100	200
Hillock Top (Crest)	C	0.95	2.00	4.40	9.70	15.25	22.00	29.30	80.20
	x/m	40.5	80	156	303	447.5	580	707	1198
	K_d	42.6	40	35.4	31.2	29.3	26.3	24.1	17.3
Upper slope	C	0.9	2.03	4.4	9.91	16.5	24	32.5	80.25
	x/m	41	79.7	156	300.9	435	560	675	1197.5
	K_d	45.6	39.3	35.4	30.4	26.4	23.3	20.8	14.9
Mid slope	C	0.6	1.5	3.75	8.75	17	26.5	35.5	78
	x/m	44	85	162.5	312.5	430	535	645	1120
	K_d	73.3	56.7	43.3	35.7	25.3	20.2	18.2	14.4
Lower slope	C	1.05	2.3	5	11.25	18	26.88	39.25	110
	x/m	39.5	77	150	287.5	420	532.5	607.5	900
	K_d	37.6	33.5	30	25.6	23.3	19.8	15.5	8.2
Flatter land (moderately sloping)	C	0.36	1	2.38	5.7	10.25	14.75	20.5	59.75
	x/m	46.4	90	176.25	343	497.5	652.5	795	1402.5
	K_d	128.9	90	74	60.2	48.5	44.2	38.8	23.5
Upland (gently sloping)	C	0.64	1.75	4.3	10.25	17.5	26.5	35	88
	x/m	43.6	82.5	157	297.5	425	535	650	1160
	K_d	68.1	47.1	36.5	29	24.3	20.2	18.6	13.2
Medium land (very gently sloping)	C	0.25	0.6	1.4	3.5	5.7	8.5	14	42.25
	x/m	47.5	94	186	365	542.5	715	860	1577
	K_d	190	156.7	132.9	104.3	94.3	84.1	61.4	37.3
Low land (level to nearly level)	C	0.79	1.85	4.5	10.5	18	27	37.25	90.5
	x/m	42.1	81.5	155	295	420	530	627.5	1095
	K_d	53.3	44	34.4	28.1	23.3	19.6	16.8	12.1

Table 7. Langmuir and Freundlich adsorption coefficients and maximum phosphate buffering capacity of the soils

Soils	Langmuir adsorption coefficients		R^2		Freundlich adsorption coefficients		R^2
	b (µg g^{-1})	K (mL µg^{-1})		MPBC (mL g^{-1})	K (mL g^{-1})	n (g mL^{-1})	
Hillock Top (Crest)	1889.31	0.0209	0.99	39.48	1.675	0.785	0.99
Upper slope	1838.21	0.0205	0.95	37.78	1.682	0.761	0.99
Middle slope	1402.48	0.0328	0.88	46.04	1.818	0.655	0.99
Lower slope	1158.82	0.0307	0.99	35.61	1.66	0.701	0.99
Flatter land (moderately sloping)	1858.28	0.0435	0.99	80.91	1.983	0.688	0.99
Upland (gently sloping)	1547.74	0.0259	0.89	40.14	1.77	0.675	0.99
Medium land (very gently sloping)	2044.03	0.0687	0.96	140.48	2.148	0.698	0.99
Low land (level to nearly level)	1462.69	0.0258	0.92	37.78	1.728	0.69	0.99

Table 10. Gibb's free energy

Soils	Initial P concentration (mg Kg^{-1})								Mean	Amount of P adsorbed to maintain 0.2 mg Kg^{-1} in equilibrium solution
	5	10	20	40	60	80	100	200		
Hillock Top (Crest)	0.451	0.919	1.904	3.941	6.005	8.221	10.462	22.53	6.804	122.33
Upper slope	0.519	1.087	2.239	4.667	7.24	9.907	12.657	26.492	8.101	128.83
Middle slope	0.618	1.358	2.968	6.288	10.281	14.317	18.195	35.54	11.195	131.52
Lower slope	0.538	1.111	2.287	4.749	7.261	9.991	12.895	26.271	8.138	178.21
Flatter land (moderately sloping)	0.54	1.254	2.707	5.844	9.348	12.965	16.872	38.258	10.973	187.64
Upland (gently sloping)	0.751	1.709	3.697	7.857	12.27	16.941	21.46	45.458	13.768	134.45
Medium land (very gently sloping)	0.425	0.926	1.99	4.409	6.859	9.161	13.535	31.839	8.643	225.01
Low land (level to nearly level)	0.608	1.295	2.785	5.869	9.169	12.615	16.123	33.198	10.208	136.75

Table 9. Correlation coefficients (r) between P supply parameters and soil

Properties	%clay	pH	OC	CEC	Al_d	Fe_d	$Fe_2O_3+Al_2O_3$	Total-P	Org-P	Fe-P	Al-P	Ca-P	Residual P	Bray's- I P	Bray's- II P	Olsen's P
b	0.036	-0.683*	-0.113	-0.243	0.853**	0.875**	-0.35	0.838**	-0.333	0.426	0.776*	-0.485	-0.526	-0.784*	-0.726*	-0.765*
k	0.465	-0.098	-0.485	0.529	0.664	0.712*	-0.161	0.116	-0.435	0.249	0.246	0.141	-0.186	-0.209	-0.122	-0.149
MPBC	0.433	0.250	-0.459	0.406	0.794*	0.837**	-0.233	0.327	-0.451	0.307	0.407	0.014	-0.281	-0.370	-0.283	-0.323
x/m at 0.2ppm	0.413	-0.193	-0.321	0.533	0.676*	0.63	-0.24	-0.081	-0.157	0.254	0.244	0.156	-0.192	-0.229	-0.181	-0.202
K	0.488	-0.116	-0.49	0.481	0.667*	0.770*	0.026	0.351	-0.523	0.260	0.268	0.108	-0.149	-0.251	-0.130	-0.149
1/n	-0.520	-0.740*	-0.414	-0.728*	0.388	0.228	-0.719*	0.325	0.260	0.267	0.662	-0.688	-0.566	-0.708*	-0.797*	-0.823*
MSP	0.352	0.568	-0.017	0.392	-0.529	-0.318	0.469	0.130	-0.068	-0.158	-0.486	0.511	0.528	0.652	0.745*	0.752*

Note: MPBC-maximum phosphate buffering capacities, MSP- mean supply parameters; *P<0.05, **P<0.01

Table 8. Phosphate Supply Parameters of the soils

Soil	Min	Max	Mean	STDEV
Hillock Top (Crest)	-1787	-983	-1480	257
Upper slope	-1845	-983	-1446	284
Middle slope	-2281	-1013	-1554	462
Lower slope	-1679	-643	-1279	337
Flatter land (moderately sloping)	-2831	-1300	-2053	480
Upland (gently sloping)	-2212	-883	-1467	432
Medium land (very gently sloping)	-3223	-1673	-2558	501
Low land (level to nearly level)	-1985	-853	-1403	385

(ΔG) of P adsorption in different soils of the transect

P- Supply Parameters

The supply parameters (SP) combines the effect of quantity, intensity and buffering capacity of phosphate which describes the ability of soil to supply P to growing crops. The SP values increased with increasing concentrations of P added indicating an increase in the capacity of the soil to supply P with increasing level of phosphorus application (Table 8). The lowest values for SP at all levels of added P were recorded in soils of crest followed by medium land soils. Both these soils had almost similar physico-chemical properties except organic carbon (Table 1) which was 1.35% in the former soil. The correlation co-efficient value of mean supply parameters with organic carbon is negative and very meager (r= -0.017) while the values with Fe_d, Al_d and Fe_2O_3+Al_2O_3 were fairly high (Table 9). This indicates that the role of organic carbon on SP is negligible, rather negative in these soils. This contradicts the findings of Das (1999) and Sahu and Achryya (2002) who observed positive effect of organic matter on SP of sulphate in some Alfisols of Orissa. The oxides of Fe and AL in these soils were the predominant factors affecting phosphate supply parameters. On the other hand, decrease in SP values also corresponded to increase in magnitudes of P adsorption maxima (b) suggesting that the soils with higher adsorption maxima supplied the lowest amount of available P to the growing crops (Majumdar *et al.,* 2005).The amount of phosphate adsorbed to maintain 0.2 µg P mL^{-1} in soil solution which is considered to be sufficient for plant growth, was highest in medium land soils having the largest adsorption maxima. However, this amount varied according to oxides of Al and Fe (Al_d and Fe_d) and clay content which was evidenced from positive correlation between them (Table 9).

Gibbs Free Energy Variation of P Adsorption Reaction

Gibbs free energy variation, or Gibbs energy, was used as a quantitative parameter of the extent of the driving force of P adsorption reaction on acid soils. The Gibbs free energy values were negative for all the soils and confirmed the feasibility and spontaneity (Mei *et al.,* 2012) of P adsorption (Figure 2 and Table 10). Similar results were reported for Cd (Dias *et al.,* 2003, Anirudhan and Radhakrishnan 2008), Cu (Silveira *et al.,* 1999), Zn (Casagrande *et al.,* 2008), and B (Soares *et al.,* 2005) adsorption on highly weathered soils.

Conclusion

The study revealed that the acid soils of Kohora Watershed transect of Assam had very high P adsorption capacity and more than 94% of added P was adsorbed at lower concentration. Phosphorus supply power of these soils increased with increasing P doses and hence phosphatic fertilizers need to be applied at higher doses for increasing crop productivity. The oxides of Al and Fe were the dominant factors of P adsorption in soils under various topographic situations in the watershed transect.

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