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Phosphorous Sorption in Some Great Soil Groups of Semi-Arid Region of Turkey

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Abstract: Removal of fertilizer phosphorous by adsorption and precipitation processes is an important factor for yield loss in low input soils of arid and semi-arid regions. The phosphate sorption characteristics of surface and subsurface horizons of four semi-arid region soils and their relationships with soil chemical and physical properties were investigated. Two sorption sites were identified. The adsorption capacity was greater for region 1 while bonding energy was higher for region 2 sites. The adsorption maxima (b_1) of the low concentration range (region-I) was 625-1250 $\mu\text{g P/g}$ and adsorption energy coefficient k_1 ranged between 0.159-0.800 $\text{mL } \mu\text{g}^{-1}$. The calculated b and k values of high concentration range (region-II) were not treated as real adsorption parameters, because as the concentration increased, the data weakly confirmed Langmuir isotherm. This revealed the fact that Langmuir isotherm can be applied to phosphorous adsorption in soils and soil materials to a limited extent. Adsorption maximum of soils for P were found to be greater for soils and horizons high in CaCO_3 , clay and CEC. Statistically significant positive relationships were found between adsorption maximum and CaCO_3 , clay and CEC whereas important negative relationships were found between adsorption capacity and organic matter, sand, Fe_2O_3 and Al_2O_3 content of soils.

Key words: Phosphorous, adsorption, precipitation, Langmuir isotherm, calcareous soils

INTRODUCTION

The reaction chemistry of phosphorous is very complex and it is very difficult to manage the behavior of P in arid and semi arid soils. Soil components such as clay, various pools of iron oxides (Castro and Torrent, 1998; Ryan *et al.*, 1985; Li *et al.*, 2007; Saavedra and Delgado, 2005), soil carbonates (Carreira *et al.*, 2006; Hamad *et al.*, 1992; Tunesi *et al.*, 1999) and organic matter (Li *et al.*, 2007) play very important role in the adsorption chemistry of P in calcareous soils. The mechanism of the adsorption of phosphate to CaCO_3 has been previously studied (de Kanel and Morse, 1978; Millero *et al.*, 2001; Stumm and Leckie, 1970). Earlier studies suggested that the initial uptake of phosphate on calcite occurs by chemisorption, followed by a slow transformation of amorphous calcium phosphate to crystalline apatite. Other studies aimed at defining the role of surface area and distinguishing between total and active carbonates have redefined the role of this mineral phase and have indicated that the low density of calcium atoms at the calcite surface reduces the importance of this mineral in P precipitation processes (Borrero *et al.*, 1988).

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Tunesi *et al.* (1999) reported that below an initial concentration of 0.5 mM the adsorption processes were predominant while above this level precipitation became predominant in calcareous soils. They also concluded that the surfaces of carbonate minerals were not necessary for induction of P precipitation. Ca-P precipitation can be governed by exchangeable-Ca ions at a convenient pH with increasing reaction time. On the other hand, surfaces of calcite are highly dynamic, even when it is exposed to air and able to incorporate adsorbed material into near-surface bulk (Stipp *et al.*, 1996). This nature of calcite surfaces can continuously create new sorption sites that may have important implications in the behavior of adsorbed P in arid climate soils with high carbonate content.

The sorption of P in Vertisols and Inceptisols were highly correlated with CDB-extractable Fe and clay (Solis and Torrent, 1989). Similarly, Li *et al.* (2007) found that single point sorption index of purple soils significantly correlated with clay content. The P sorption maximum of pure CaCO₃ was reported to be 9.2-25 mg P kg⁻¹ (Hamad *et al.*, 1992; Griffin and Jurinak, 1974) whereas limestones showed much higher P sorption maximum between 105-124 mg kg⁻¹ due to presence of impurities, particularly Fe (Holford and Mattingly, 1975). Ming *et al.* (2002) pointed out that the P adsorption capacity of the organo-mineral colloidal complexes differed with the soil types, being higher for the acid and calcareous purple soils than for the neutral purple soils and partial removal of the organic matter increased the adsorption capacity of the colloidal complexes.

The aims of this study were to investigate sorption characteristics of surface and subsurface of four great groups developed on the alluvial and calcareous parent material over a very large concentration range of phosphate (0-300 µg mL⁻¹), that enables both adsorption and precipitation and to define the relationships between Langmuir parameters and soil properties.

MATERIALS AND METHODS

Soils

The top two horizons of a Fluventic haploxeroll (A), a Typic xeropsamment (B), a Typic calcixerept (C) and a Calcic haploxerept (D) from Ankara (Turkey) were used in this study. The soils are derived from calcareous and alluvial materials and occur at elevation about 800 m (Dengiz, 2002). Climatic conditions in the area are semi-arid with 450 mm rainfall and 10-13°C average temperature. Mineralogical analysis showed that chlorite was the dominant phyllosilicate in the Typic xeropsamment and Fluventic haploxerepts whereas; calcite, quartz, feldspar and 2:1 type smectite minerals were dominant in clay fraction of the Typic calcixerepts and Calcic haploxerepts.

The samples were air-dried, gently crushed and passed through a 2 mm sieve. Particle size distribution was determined by hydrometer method (Gee and Bauder, 1986). Soil chemical properties were determined following standard procedures (Sparks, 1996), viz, pH in 1:2.5 soil: water suspension, electrical conductivity in saturation paste, organic matter by K₂Cr₂O₇ oxidation, CaCO₃ equivalent by a manometric method, available P by extraction with 0.5 M NaHCO₃ at pH 8.5 (1 g: 20 mL), CEC by saturating the exchange sites with 1 M NaCH₃COO at pH 8.2 then replacing the Na with M NH₄CH₃COO at pH 7.0. Total free Fe and Al oxides were extracted with citrate-dithionite buffer (Mehra and Jackson, 1960) and determined by AAS. The data are shown in Table 1.

Phosphate Sorption

P-sorption isotherms of soils were determined by equilibrating 1.000 g soil with 40 mL of 0, 10, 25, 50, 75, 100, 150, 200, 250 and 300 µg P (as KH₂PO₄) mL⁻¹ solution prepared in 0.01 M CaCl₂ for 24 h at 25°C. An equilibration time of 24 h was chosen for experimental convenience. Any period ranging 6-48 h would ensure completion of the rapid reaction at the surface sorption sites, without a significant contribution from the subsequent slow reactions in which diffusion into the solid phase is likely to occur. At the end of shaking process, the supernatant solutions were separated by

Table 1: Some physical and chemical properties of soils

Soil	Sand (g kg ⁻¹)	Silt (g kg ⁻¹)	Clay (g kg ⁻¹)	pH (1:2.5)	Salt (%)	CaCO ₃ (g kg ⁻¹)	OM (g kg ⁻¹)	CEC (cmol kg ⁻¹)	Fe ₂ O ₃ (g kg ⁻¹)	Al ₂ O ₃ (g kg ⁻¹)	Olsen-P (kg ha ⁻¹)
1	461	272	266.0	7.61	0.015	1.0	29.8	21.5	62	119	26.4
2	584	243	173.0	7.52	0.009	3.5	21.3	16.2	54	95	23.5
3	386	338	276.0	7.67	0.017	15.8	23.5	16.4	50	118	44.0
4	608	167	224.0	7.64	0.020	16.5	20.1	18.4	50	120	29.0
5	247	400	347.0	7.67	0.023	23.8	19.4	28.9	40	85	19.3
6	176	452	372.0	7.88	0.018	35.5	4.0	25.1	39	75	10.3
7	386	317	29.7	7.50	0.012	27.6	16.8	29.9	24	45	35.0
8	270	385	34.5	7.09	0.017	17.0	11.1	42.7	28	52	27.0

1: Typic xeropsamment (0-18 cm), 2: Typic xeropsamment (18-47 cm), 3: Fluventic haploxerepts (0-25 cm), 4: Fluventic haploxerepts (25-65 cm), 5: Typic calcixerepts (0-13 cm), 6: Typic calcixerepts (25-60 cm), 7: Calcic haploxerepts (0-16 cm) and 8: Calcic haploxerepts (16-44 cm)

centrifugation and filtration through Whatman No. 42 filter paper. Then, the equilibrium P concentration was determined by spectrophotometry (Murphy and Riley, 1962). The amount of sorbed-P (x/m) was calculated from the difference between the amount of P added and that which remained in the equilibrium solution (C).

The data were fitted to the linear form of the Langmuir adsorption isotherm:

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

Where:

b = The monolayer adsorption maximum ($\mu\text{g g}^{-1}$)

k = The adsorption energy coefficient ($\text{mL } \mu\text{g}^{-1}$)

Since very large initial concentration range ($0\text{-}300 \mu\text{g mL}^{-1}$) was used, the sorption isotherms were split into two different regions by step wise regression analysis (Steel and Torrie, 1960). Adsorption was the dominant processes at the lower part whereas; precipitation was likely to occur at upper site of the isotherms as indicated by apparent decrease in the C/x ratio.

Correlation analysis between the Langmuir parameters and soil characteristics was performed by using SPSS.

RESULTS AND DISCUSSION

The soil samples from the four great soil groups with two horizon almost completely adsorbed all of the applied phosphorous from the solutions in the 0 to $150 \mu\text{g mL}^{-1}$ concentration range. As the phosphorus concentration was increased, however, each great soil group developed a different equilibrium concentration. The plots of C versus C/(x/m) for great soil groups are shown in Fig. 1. There are two main parts in the sorption isotherms where adsorption and precipitation driven sorption mechanisms are displayed. Langmuir parameters of the soils for different soil depth and isotherm regions with the regression coefficient are shown in Table 2. Sorption maxima (b1) for region 1 ranged between 625 (Typic xeropsamment) to $1250 \mu\text{g g}^{-1}$ (Calcic haploxerepts) and for region 2 between 2702 (Fluventic haploxerepts) to $25000 \mu\text{g g}^{-1}$ (Typic calcixerepts). Amrani *et al.* (1999) reported a maximum adsorption range of 146 to $808 \mu\text{g P g}^{-1}$ for Moroccan calcareous soils. The data better conformed the theoretical Langmuir isotherm in the low concentration range (region 1), that was in agreement with findings of other researchers (Derici and Kaya, 1991; Derici and Agca, 1991). On the other hand, P adsorption isotherm can be better obtained at an initial concentration below 15mg L^{-1} (Bertrand *et al.*, 2003). But in present case due to common agricultural practices which is rain-fed agriculture with minimal input management, natural P content of the soils can be minimal (Table 1).

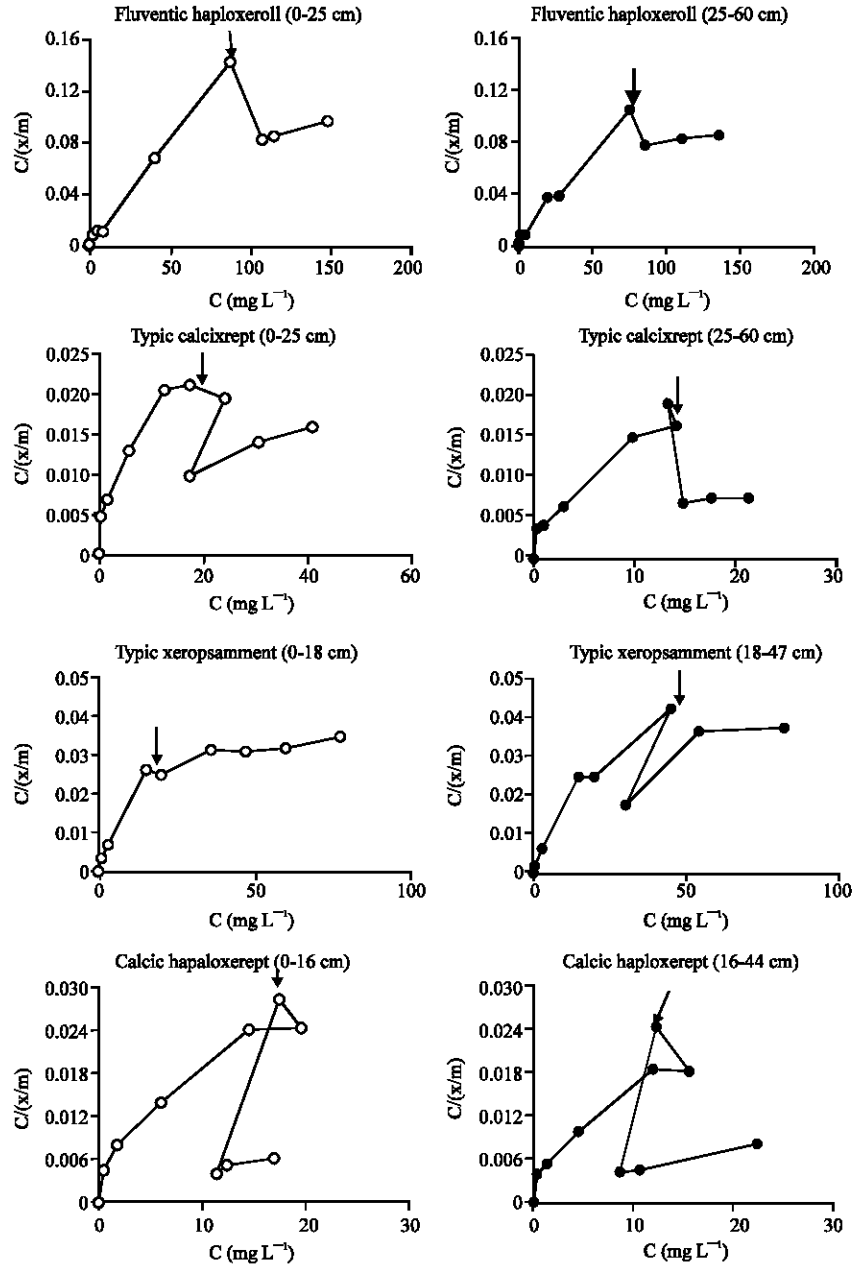


Fig. 1: Langmuir sorption isotherms of surface and subsurface horizon of different great soil groups. Arrow shows separation between adsorption and precipitation-driven reaction of phosphate

In fact, this consequence leads soils to adsorb more P than high input soils. High input soils usually demonstrate monolayer adsorption maxima; therefore, small additions of P make the soil attain maximum adsorption. At high P concentration range ($>150 \text{ mg P L}^{-1}$) the precipitation process was the dominant mechanism and the data in this range somehow conformed to the Langmuir isotherm with smaller regression coefficient than the first part. It is reported that precipitation driven reactions can

Table 2: Parameters of Langmuir isotherms

Soil No.	Langmuir equation	b ($\mu\text{g g}^{-1}$)	k ($\text{mL } \mu\text{g}^{-1}$)	r^2	F	P
1	Y1 = 0.00200+0.00160X	625	0.800	0.998	2605.20	0.01
	Y2 = 0.04300+0.00036X	2777	0.008	0.999	1073.00	0.05
2	Y1 = 0.00394+0.00129X	775	0.320	0.991	548.00	0.01
	Y2 = 0.06069+0.00016X	6250	0.002	0.991	110.70	0.05
3	Y1 = 0.00319+0.00093X	1075	0.291	0.878	36.08	0.01
	Y2 = 0.02426+0.00013X	7692	0.005	0.940	15.67	0.01
4	Y1 = 0.00283+0.00097X	1030	0.342	0.942	81.08	0.01
	Y2 = 0.01006+0.00037X	2702	0.037	0.735	2.77	0.10
5	Y1 = 0.00443+0.00094X	1063	0.212	0.791	15.10	0.05
	Y2 = 0.00515+0.00004X	25000	0.007	0.974	37.68	0.01
6	Y1 = 0.00314+0.00096X	1041	0.305	0.952	99.38	0.01
	Y2 = 0.00188+0.00028X	3571	0.148	0.753	3.05	0.10
7	Y1 = 0.00503+0.00080X	1250	0.159	0.931	67.62	0.01
	Y2 = 0.00514+0.00027X	3703	0.052	0.793	3.83	0.05
8	Y1 = 0.00278+0.00086X	1162	0.309	0.865	32.10	0.01
	Y2 = 0.00862+0.00005X	20000	0.005	0.994	177.80	0.05

Table 3: The correlation coefficients between Langmuir parameters (b, k) and some soil properties

	b_1	k_1	b_2	k_2
b_1	-			
k_1	0.551	-		
b_2	0.317	-0.339	-	
k_2	0.251	-0.192	-0.393	-
Clay	0.649*	-0.259	0.323	0.397
Silt	0.580	-0.340	0.363	0.248
Sand	-0.625*	0.313	-0.355	-0.319
Org. mat.	-0.621*	0.529	-0.518	-0.328
CEC	0.351	-0.245	0.114	0.820*
CaCO ₃	0.863**	-0.646*	0.684*	0.050
Fe ₂ O ₃	-0.648*	0.664*	-0.574	-0.478
Al ₂ O ₃	-0.639*	0.551	-0.331	-0.285

*: Significant at $p < 0.05$ and **: at $p < 0.01$

be successfully described by the Langmuir isotherm (Veith and Sposito, 1977). There was a very strong deviation in the border line of the two process with an apparent increase before precipitation of any solid phase which was indication of the supersaturation of solution P against relevant solids or decrease after precipitation of a discrete solid phase. In general the mineralogy of the soils was likely an important factor in sorption maximum. Soils containing chlorite mineral (Typic xeropsamment and Fluventic haploxerepts) sorbed more P than montmorillonitic soils (Typic calcixerepts and Calcic haploxerepts). Mollisols and vertisols dominated by 2:1 clay minerals (e.g., montmorillonite) were reported to have relatively lower sorption capacities (Sims and Baker, 2003).

Correlation between soil properties such as clay content, oxides of Al and Fe, CaCO₃ equivalent, organic matter and P sorption parameters of different theoretical models have been widely used to determine the main soil components in P behavior (Brennan *et al.*, 1994; Bertrand *et al.*, 2003). The relation between Langmuir parameters and some physical, chemical and mineralogical properties of experimental soils were investigated through correlation analyses and the results are given in Table 3. Correlation analysis revealed that adsorption maxima in region-1 were positively affected by CaCO₃ ($r^2 = 0.863^{**}$) and clay ($r^2 = 0.649^*$) content whereas Fe₂O₃ ($r^2 = -0.648^*$), Al₂O₃ ($r^2 = -0.639^*$) and sand ($r^2 = -0.625^*$) content had a negative significant effect.

The contribution of exchangeable Ca-ions to Ca-phosphate precipitates was reported to be higher than that of CaCO₃ (Akinremi and Cho, 1991a, b; Tunesi *et al.*, 1999) but the role of CaCO₃ in replenishing Ca in soil solution and exchange sites and calcification process during soil formation should not be discarded. On the other hand, above 150 mg L⁻¹ initial concentrations (region-II) adsorption maxima was correlated with CaCO₃ ($p < 0.01$). This suggests that phosphorous was first precipitated

with CaCO₃ as thermodynamically less stable discrete solid phase (i.e., di/tri calcium phosphate) with subsequent formation of more stable phases such as apatite (Lindsay, 1979; Carreira *et al.*, 2006). In addition, it was reported that majority of applied P can precipitate as poorly soluble Ca-phosphates in low P containing calcareous soils (Delgado and Torrent, 2000) such as the experimental soils.

Oxides of Fe and Al are important cementing agent in soils that reduce the total surface area thus sorption sites despite their very high adsorption ability in pure systems. The negative correlation between oxide content and sorption parameter may be attributed to alkaline pH that reduce the solubility of oxides while increasing the stability of surface coating on other soil particles and aggregating effect and oxides of Al and Fe approach to the point of zero charge, therefore the affinity of oxide surfaces decreases for P adsorption (McBride, 1994). On the contrary, a significant correlation between P retention capacity and a soil property does not necessarily imply a significant direct effect of the soil property on P retention (Ige *et al.*, 2007). For example sand content, which have indirect effect, usually reduces the exchangeable Ca content of soils (Ige *et al.*, 2007). Negative correlation between organic matter content and adsorption maxima could be related to competition between low molecular weight organic acids and phosphate for sorption sites that usually occurs in favor of organic acids and delays P adsorption (Geelhoed *et al.*, 1999; Staunton and Leprince, 1996; Violante and Gianfreda, 1993). The coating of high affinity sorption sites by organic matter and organo-mineral complex formation could be more significant as organic matter content of soil increased and in especially fine textured clay soils. When different horizons are taken into consideration, horizons high in clay and CaCO₃ and low in sand, adsorption maximum was significantly high. These findings are in agreement with the reason given above.

CONCLUSIONS

Phosphorus added to calcareous soils was removed from soil solution by the combination of adsorption and precipitation processes. At lower end of the sorption isotherm, the adsorption was the dominant process whereas higher concentration addition promoted precipitation. Initial P concentration above 150 µg mL⁻¹ precipitation-driven reaction was dominant. In general, P sorption characteristics of arid region soils are likely related to the amount of clay and reactive oxides or carbonates present in the soil and are influenced by the physical and chemical characteristics of the solid phase (e.g., mineralogy and crystallinity). Soils containing chlorite clay mineral sorbed more P than montmorillonitic soils. Oxides minerals and organic matter are negatively correlated with the sorption parameters. It can be concluded that P sorption characteristics along with the soil properties should be considered for economically and environmentally friendly fertilization.

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