

Characterization of an Alfisol Collected from Dry Zone of Sri Lanka to Elucidate the Retention Mechanisms of Pollutants

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ABSTRACT. *Characterization of physicochemical properties of a soil enables predicting the behavior of nutrients and pollutants added to the soil. A study was conducted to characterize environmentally important physicochemical properties and processes of an Alfisol (Reddish Brown Earth) collected from the Dry zone of Sri Lanka. Organic carbon content (OC), pH, Electrical Conductivity of soil suspension (EC) and saturated paste (ECe), Cation Exchange Capacity (CEC), and available and total metals (Cu, Zn and Cd) were analyzed. Potentiometric titration and adsorption study were conducted to determine the Point of Zero Charge (PZC) and phosphorus adsorption capacity of the soil. Clay mineralogy of the soil was determined using X-ray diffraction technique. Among the basic soil chemical properties analyzed, CEC ($3.2 \text{ cmol}_+ \text{ kg}^{-1}$) showed a low cation retention attributed to low organic carbon percentage (mean 0.37 %) and the predominating of low-activity clay minerals of kaolinite and illite. The EC values ($EC=0.03 \text{ dS m}^{-1}$, $ECe=0.3 \text{ dS m}^{-1}$) indicated a low salinity level. Point of zero charge of the soil was 4.0 indicating net negative charges at the existing soil pH of 6.37. At the near neutral pH of the soil, the cation retention enhanced, subsequently affecting the mobility of metals. Phosphorus adsorption data fitted to Langmuir isotherm gave an adsorption maximum of 4285 mg kg^{-1} indicating a high P fixing ability. The soil properties studied can be used to elucidate the retention mechanisms of pollutants in this soil.*

Keywords: *Physicochemical properties, clay mineralogy, phosphorus adsorption, point of zero charge.*

INTRODUCTION

The Alfisol Catena is the most extensive soil association in Sri Lanka covering 2.5 million ha and has the greatest potential for agricultural crop production (Dimantha, 1994). Well drain Reddish Brown Earth (RBE) soils (Rhodustalfs) occur on the crests and upper slopes of the dry zone catena (Somasiri, 1984). Further, a larger proportion of Sri Lanka's agriculture is confined to the Dry zone. With the green revolution and introduction of newly improved crop varieties intensive agricultural practices have been adopted by farmers. Long term adaptation of such practices alter the basic chemical properties of soil such as soil pH (Simek *et al.*, 1999), organic matter content (Manna *et al.*, 2005), cation exchange capacity (CEC) (Zhao and Zhang, 1997) soil nitrogen (Simek *et al.*, 1999), phosphorus and potassium contents

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(Parham *et al.*, 2002). Further, many researchers have revealed that trace metals such as Cadmium, Zinc, Copper and Arsenic contents in soil can increase as they are found in most of the synthetic fertilizers and agrochemicals such as pesticides and herbicides as impurities or active ingredients (McLaughlin *et al.*, 2000, Ajayi *et al.*, 202). Metals transfer from soil to other ecosystem components such as groundwater and crops is one of the most important sources of trace metal contamination of humans. This can be a threat to human health due to their persistence and toxicity (Mico *et al.*, 2006). Research conducted in intensive agricultural areas evidenced presence of elevated trace metal levels. Premarathne *et al.*, (200) reported elevated amount of some trace elements in intensively vegetable grown wet zone fields in Sri Lanka.

Since clay mineral inter-phase plays a major role in metal transport and cycling through the geosphere, electrochemical properties of clay mineral surfaces are important in environmental and geochemical studies (Sinitsyn *et al.*, 999). Soil colloid surfaces can bare permanent and variable charges. These variable charges can be net negative, positive or zero charge depending on the soil pH. This amphoteric character is important in soil nutrient management and retention of soil contaminations (Naindu *et al.*, 997). The pH at which the net total particle charge is zero is called the point of zero charge (PZC), which is one of the most important parameters used to describe variable charge surfaces (Morais *et al.*, 976). The PZC governs the ion exchange, adsorption and other chemical processes occurring at the liquid colloid inter-phase. Such reactions control physicochemical properties of ions and affects soil-water-plant ecosystems, specially retention of metal on the soil surface (Karak *et al.*, 2005). Among the different methods for the determination of the PZC in different materials, soil researches generally rely on potentiometric titration. This method determines the point of zero salt effect (PZSE) which assesses changes in surface potential with changes in the activities of H^+ and the OH^- (Appel *et al.*, 2003). Point of zero salt effect value can be used to explain the mechanisms associated with specific adsorption of cations (Ca^{2+} , Cu^{2+} , Pb^{2+} , Zn^{2+}) and the anions (F^- , PO_4^{3-} , SO_4^{2-}) which can modify the proportion between the negative and positive surface charge of the soil (Yu *et al.*, 997 and Zhao & Zhang, 997). Further difference of PZSE and pH jointly affect the degree of flocculation of the soil particles and consequently, the water dynamics, gas transfer and susceptibility to erosion losses (Alves and Lavorenti, 2005).

Adsorption is the mechanism commonly responsible for the retention of ions and molecules, particularly trace element cations, anions and organic compounds by soil (Essington, 2003). Tan (994) revealed that the soil phosphorous fixation reactions may allow only a small fraction (0 to 5%) of the phosphorus in fertilizer to be taken up by plants. Meantime, farmers apply two to four times more quantities of phosphorus as it would be removed as crop harvest. Repeating such practices over many years will saturate the phosphorus fixation capacity and build up the level of available phosphorus in agricultural soils (Daniel *et al.*, 998). The accumulation and transfer of phosphorus in intensive farming systems has increased phosphorus export from agricultural watersheds and accelerated eutrophication of surface water (Sharpley and Tunney, 2000). Phosphorus retention capacity was found to be a measure of the ability of soil to retain P, and an important factor controlling the release of P from soil to water (Akinremi *et al.*, 2007). Hence, characterization of a soil with respect to environmentally important physicochemical properties is important to maximize the crop production while minimizing environmental pollution. Therefore, objective of this study was to characterize the selected Alfisol (RBE) with respect to environmentally important physicochemical soil properties and processes to understand the retention behavior of inorganic pollutants at colloidal inter-phase.

MATERIALS AND METHODS

Soil sampling and analysis of basic chemical properties

Soil sample was collected from the Field Crop Research and Development Institute, Mahalluppallama at 08° 06.73' N, 80° 28.353' E located within DL agro ecological region in the dry zone of Sri Lanka. According to the records, the selected location was not cultivated for about four years. Three sub samples were collected to the depth of 0-30 cm to obtain a composite sample representing well drain RBE soil. Soil sample was air dried and passed through 2 mm sieve prior to laboratory analysis. The physiochemical properties measured were organic carbon, pH, Electrical Conductivity (EC), Cation Exchange Capacity (CEC), and total and available concentrations of Cu, Zn and Cd. Organic carbon percentage was determined according to method described by Nelson and Sommer (1996). Soil pH was measured by using digital pH meter in both water and M KCl solution having :2.5 soil: solution ratio. Electrical conductivity meter was used to measure the EC in :5 soil water suspensions and E_c in saturated paste. Ammonium acetate (pH=7) extraction was used to determine the cation exchange capacity (CEC) as described by Summer and Miller (1996).

Extraction of metals

Total metal contents were determined according to the method proposed by Sposito *et al.*, (1982). In this method soils were digested with 4M HNO₃ at 80°C for four hours. Metal contents in filtered samples were measured using Atomic Absorption Spectrophotometer. Metals associated with all the pools except structural components are extracted by the nitric acid-digested procedure. Available metal contents were determined using DTPA at pH 7.3 and M HCl (Bandara, 2007).

Point of zero charge determination

Potentiometric titration method was used to determine the PZSE using 0.1M, 0.01M and 0.001M KCl as described by Appel *et al.* (2003). Soil sieved through 0.5 mm was used. Soil was washed with 0.001M HCl and air dried prior to the analysis. Ten milliliters of electrolyte solution was added to four grams of soil. Deionized water was used to make 25 mL of final volume. Soil pH was adjusted to a range between 2 and 8 with measured amount of 0.1M HCl and 0.1M KOH. After seven days of incubation period soil pH was measured in each suspension.

Clay mineralogy analysis

Pretreatments for the clay mineralogy analysis were done according to the method described by Kunze and Dixon (1986). Removal of soluble salts, organic matter and iron oxides were done prior to the clay fraction separation. Diffractograms were obtained between 2-30° 2θ from K-, Mg- saturated and Glycerol solvated clay samples. X-ray diffraction test was carried out by the Bruker FD800 Diffractometer (Germany) operated at the potential of 40 kV and the amperage of 30 mA, producing Cu Kα radiation at a wave length of .54 Å equipped with Ni filter and silicon monochromator. The diffractograms were obtained using a continuous scanning technique at the speed of 0.02° per second.

Phosphorus adsorption study

Phosphorus adsorption study was conducted using 0g L⁻¹ soil and 0.0M KCl as background electrolyte at pH 5.4. Six concentration levels (0, 25, 50, 00, 50 mg P L⁻¹) of phosphorus and soils sieved by 0.5 mm were used for the study. Ten mL of 0.0 M KCl solutions containing 0, 25, 50,00 and 50 mg P L⁻¹ were added to soil. Solution pH was adjusted to 5.4 using 0.M HCl and 0.M KOH as required and samples were shaken for 30 minutes and incubated for 24 hours at 29^oC. After centrifugation, suspension was filtered and phosphorus concentration was determined colorimetrically using the molybdenum blue method. The phosphorus adsorption maximum was obtained fitting to the Langmuir equation, after checking conformity.

RESULTS AND DISCUSSION

Organic carbon content is one of the most important soil properties affecting the physical, chemical and biological properties. Mean organic carbon content of the soil was 0.37% (Table) indicating very low organic carbon content in the soil. Low soil organic carbon contents are common for tropical soils. Kumaragamage and Kendaragama (200) also discussed the necessity of proper organic matter management practices for dry zone soils in order to improve the nutrient retention in soil. Poor organic carbon content also resulted in low CEC value (3.2 cmol_ckg⁻¹) indicating a moderate cationic nutrient retention in the soil. It suggests that soil fertility improvements could be done by addition of organic materials in this studied reddish brown earth soil in the dry zone of Sri Lanka. Soil sample showed a pH value of 6.37confirming the value of 6.4 reported by Kumaragamage and Kendaragama (200) for reddish brown earth soil in this area. Electrical conductivity gives an idea about the soluble salt content in soil. Presence of high amount of soluble salts in the soil is a major constraint limiting crop growth and yield in many of the soil series of the dry zone (Kumaragamage and Kendaragama, 200). However the measured EC and ECe values indicated low amount of soluble salts in soil.

Table 1. Some important basic soil properties measured in the studied soil

Soil property	Value*
Organic Carbon	0.37%
Soil pH (:2.5, distilled water)	6.37
Soil pH (:2.5, M KCl)	5.29
EC (suspension)	0.03 dS/m
EC (saturated paste)	0.3 dS/m
CEC	3.2 cmol _c /kg
Clay %	24.3
Textural class	Sandy clay loam

*values are average of three replicates

Sources of trace metals in soil are mainly due to industrial and agricultural activities and natural metal deposits. Dry zone of Sri Lanka is not currently engaged in industrial activities at commercial levels and natural metal deposits are not reported (Indraratne, 2009)

Therefore, agricultural practices could be the main source to the trace metal accumulation in dry zone soils. Measured total and available metal contents are presented in Table 2.

Table 2. Available and total concentrations of Cu, Zn and Cd measured in the studied soil

-----Cu (mg kg ⁻¹)-----			-----Zn(mg kg ⁻¹)-----			-----Cd(mg kg ⁻¹)-----		
DTPA	MHCl	total	DTPA	MHCl	total	DTPA	MHCl	total
0.64	3.06	8.84	1.08	3.58	20.85	0.03	0.09	0.16

*values are average of three replicates

Cadmium is one of the trace elements that accumulate in plants to high concentrations that cause toxicity in animals but have no effect on plant growth. Whereas Cu and Zn show phytotoxicity at concentrations much lower than those that affect humans negatively (Chaney, 1973). Therefore, Cd has a hidden toxicity. Cadmium is present in fertilized soils as a divalent cation (Cd²⁺) and associated with organic (Cd-dissolved organic carbon) and inorganic complexes (Stacey *et al.*, 200). Observed total metal concentrations were below the regulatory limits given by the European community for agricultural soils. European community set standards for Cu, Zn and Cd as 50-40 mg kg⁻¹, 50-300 mg kg⁻¹ and -3 mg kg⁻¹, respectively (McGrith and McCormack, 1999). Assessing of different fractions of soil metals other than total metal content is important in predicting the availability and mobility of trace metals in soils (Kabata –Pendias and Pendias, 200). Available (HCl and EDTA extractable) metals also showed low concentrations indicating that the soil is not contaminated. Further, DTPA extractable fraction is lower than the M HCl exchangeable fraction. Tu., *et al.* (2000) found that organic bound metal fraction is positively correlated with soil organic matter content. Therefore, low organic matter in the soil (Table) could be a reason for the lower concentration of available Cd, Cu and Zn.

Clay mineralogy analysis

X-ray diffractogram of the clay fraction of the Alfisol is shown in Fig. A peak at 0.0Å in K-saturated, Mg saturated and Mg-Glycerol solvated samples respectively evidenced the presence of illite. Presence of kaolinite signified the peaks at 7.23 Å in K-saturated and Mg saturated samples. The bulge at 8.2 Å with glycerol treatment indicates the presence of montmorillonite as monir clay mineral.

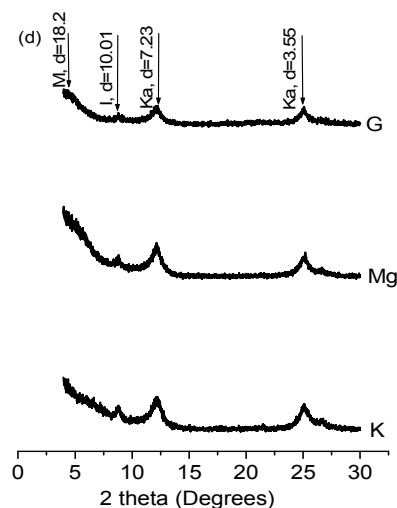


Fig. 1 X-ray diffractogram of the clay fraction of RBE soil (Ka= Kaolinite, I= Illite, M= montmorillonite, K= K-saturated, Mg= Mg saturated, G= Glycerol solvated).

This confirmed illite and kaolinite are the dominant clay mineral while montmorillonite is the minor clay mineral in soil clay fraction of this Alfisol (Indraratne, 200). Therefore, it is evident that the contribution of kaolinite for the surface charge development is significant for this soil. Further, dominance shown by kaolinite and illite could be the reason for low CEC in this studied soil.

Point of zero charge determination

The PZSE is the pH where the net adsorption of potential determining ions, H^+ and OH^- , on variable charge surfaces and is independent of electrolyte concentration (Appel *et al.*, 2003). Figure 2 shows the potentiometric titration curves at varying electrolyte concentrations (0., 0.0 and 0.00M KCl). Asymmetric charge behavior was observed (Fig. 2) indicating the heterogeneous nature of soil. Apparently two intersecting points were observed in this heterogeneous soil colloidal system in contrast to pure inorganic colloids. The titration curves for 0.0 and 0.00 M KCl showed an intersecting point at pH 6.9 which is above the line of charge symmetry. Whereas titration curves for 0. and 0.0 M KCl showed an intersecting point at pH 4.0 which is below the line of charge symmetry. The appearance of multi intersections of the titration curves is indicative of the activation of different soil surface sites for proton transfer.

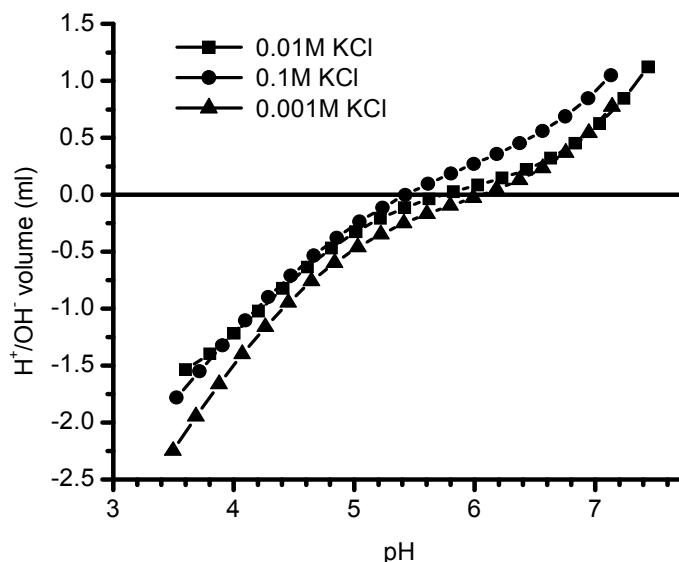


Fig. 2. Potentiometric titration curves for the studied soil at different electrolyte concentrations.

If the soil pH is above PZC, soil surfaces have a net negative charge and exhibit ability to exchange cations. If its pH is below its PZC, soil will mainly retain anions. The difference of pH measured between KCl and distilled water can provide a good estimation of the net surface charge of soils (Alves and Lavorenti, 2005). In this Alfisol where pH is 6.37, the difference of pH in KCl and pH in H₂O of the soil was -0.08 (table) which indicates negative net surface charge. Further, estimated PZC value according to the equation proposed by Keng and Uphara, (974) ($PZC = 2 \text{ pH}_{\text{KCl}} - \text{pH}_{\text{H}_2\text{O}}$) was 4.2. This also confirms that the PZC value of the soil could be around pH 4. The variable charges in soils develop on the edges of lattice clay minerals and on the surface of sesquioxide, amorphous materials such as allophone imorgolite and organic matter (Karak *et al.*, 2005). Negatively charge organic matter is contributing more to negative surface charges. Since soil is a mixture of different minerals, soil PZC value is determined by minerals which are having different PZC values and organic matter. Presence of minerals which are having lower PZC values; kaolinite (4.6), montmorillonite (2.5), feldspar (2.0-2.4), albite (2.0) SiO₂ (2.0), and MnO₂ (2.8) tends to reduce the point of zero charge in soils. Further Ogunwale and Isa, (2004) reported that PZC of soil tends towards lower value when organic matter content is high. In this soil, organic carbon content is low indicating high contribution of kaolinite for surface charge development. The lowering effect of kaolinite on PZSE is probably due to its edge single OH groups coordinated with Si⁴⁺, the silanol groups. Being more easily ionizable than the single OH groups coordinated with the Al³⁺ (AlOH) and to Fe³⁺ (FeOH), the silanol groups tend to only dissociate protons (Sparks, 995). For this reason protonation of silanol groups takes place only at high H⁺ concentration which contribute to lowering of PZSE in soil mainly with nonallophanic kaolinite. Further when soil become more weathered their PZSE values

increase due to kaolinite destabilization and tend toward the soil pH values dropping the soil net surface charge to values near zero (Alves and Lavorenti, 2005).

The titration curves of the soil sample did not follow the usual trend of higher electrolyte concentration resulting in higher pH below the PZSE because at lower pH values protons were primarily consumed in the mineral dissolution reactions present in soils instead of generating surface charge (Appel *et al.*, 2003, Karak *et al.*, 2005). In order to explain the behavior of 0.00 M KCl titration curve further analysis is needed. However, an intersection point at pH 6.9 indicates the presence of positive charges at colloidal surfaces, enabling soil to retain some anions.

Phosphorus adsorption study

Plot of the amount adsorbed against amount in solution (Fig. 3a) indicated that phosphorus adsorption increased with increasing phosphorus concentration in solution. In order to obtain maximum adsorption of phosphorus for the soil, data were transformed to fit the linear form of Langmuir adsorption isotherm (Fig. 3b) using the following equation.

$$\frac{1}{q} = \frac{1}{q_{\max}} + \frac{1}{kq_{\max}} \frac{1}{c}$$

where c is the final concentration after adsorption has occurred (mg L⁻¹), q is the amount of P adsorbed (mg kg⁻¹), q_{max} is the adsorption maximum (mg kg⁻¹) and k is a coefficient related to the bonding energy.

Conformity to the linear form of the Langmuir equation was observed for P adsorption with r²=0.99 (p<0.00). Therefore, using the linear adsorption equation adsorption maximum (q_{max}) calculated was 4285 mg kg⁻¹.

According to adsorption data, this soil is having high phosphorus adsorption capacity indicating high potential to adsorb phosphate ions. It indicates when runoff water detach soil colloids and transport P enriched soil particles to surface water bodies that could result environmental problems, such as eutrophication.

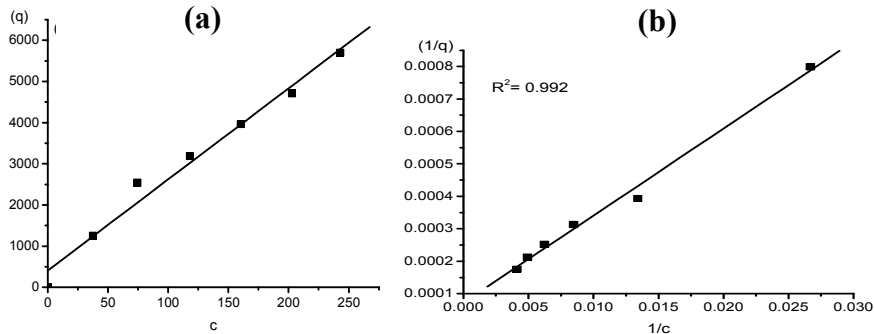


Fig. 3. Amount of P adsorbed against increasing solution P concentrations (a) and linear form of the Langmuir adsorption isotherm (b) (q= amount adsorbed, mg kg⁻¹; c= equilibrium concentration, mg L⁻¹).

CONCLUSIONS

Studied soil shows a moderate retention ability of cations attributed to poor organic carbon content and low-activity clay mineralogy. Therefore, adaptation of proper organic matter management practices can minimize the loss of cations from soil and thereby minimize nutrient depletion due to leaching. Unlike many of the soils in dry zone, salinity is not a major problem in this location. Available and total metals concentrations in soil are below the threshold levels to cause any toxicities. Point of zero charge of the soil suggests that with the soil pH of 6.39 cations could be exchanged and become important in metal transport from soil to other ecosystems. Adsorption maximum of P in the soil is high indicating P deficiency as a main agronomic limitation of this soil. However, it has a potential for P enrichment with long term application of phosphorus fertilizers. Thus, measures are needed to increase the CEC and minimize erosion to prevent eutrophication in surface water bodies. Since this soil has not been cultivated for about four years, it could be used as a reference or model soil to monitor impacts due to agricultural practices in other soils in the area.

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