

PALYGORSKITE FORMATION UNDER THE INFLUENCE OF GROUNDWATER IN CENTRAL IRANIAN SOILS

S. HOJATI^{1,*} H. KHADEMI¹ A.F. CANO²

¹Isfahan University of Technology, College of Agriculture, Department of Soil Sci. Isfahan, Iran

²Technical University of Cartagena, Departamento de Ciencia y Tecnología Agraria, Murcia, Spain

*e-mail: saeidhojati@ag.iut.ac.ir

Abstract: Palygorskite is a dominant clay mineral in most soils and sediments in central Iran. Few studies investigated the formation of palygorskite under the influence of groundwater in central Iran. This study aims to identify the effects of groundwater on the formation of palygorskite and associated clay minerals. Four soil profiles along a calcareous catena with different depths of groundwater were sampled and analyzed using X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM) and inductively coupled plasma mass spectroscopy (ICP-MS). Chemical composition of groundwater and that of soil samples were also determined. The XRD results showed the presence of palygorskite, smectite, mica, chlorite, quartz and feldspars in soils studied. The highest amount of palygorskite was found in horizons with permanent occurrence of groundwater. Palygorskite was not detectable using XRD in surface horizons. However, TEM studies confirmed presence of palygorskite in surface horizons. Smectite showed an inverse trend when compared with palygorskite. Stability diagrams for smectite-palygorskite system in studied profiles showed that geochemical conditions were conducive for the formation of palygorskite from smectite. Growth of palygorskite fibers on gypsum and calcite crystals, as revealed by SEM observations, clearly indicates the in situ formation of palygorskite. Chemical composition of groundwater in study area showed high concentrations of soluble Mg^{2+} and silica which are necessary for the formation of palygorskite from solution. In conclusion, it seems that the two likely sources of palygorskite in the study area include the transformation from smectite and precipitation from solution.

Key Words: Palygorskite, Smectite, Soil solution, Central Iran

1. INTRODUCTION

Palygorskite is a fibrous clay mineral which is commonly found in soils and sediments of arid regions (Singer, 1989). In Iran, palygorskite was first reported by Henderson and Robertson (1958) and Burnett et al. (1972), but the sources of this mineral were not discussed in their reports (Burnett et al., 1972; Henderson and Robertson, 1958). Two main origins for palygorskite in soils have been reported including the inheritance from parent materials and pedogenic formation. Inherited palygorskite is either passed over from the parent rocks to the soil (Khademi and Mermut, 1998; Viani et al., 1983) or added to the soil by palygorskite-rich aeolian dusts (Danin and Ganor, 1991) or alluvial materials (Khademi and Mermut, 1998). Pedogenic palygorskite can be the result of either in situ transformation of another mineral (Hillier and Pharande, 2008) or authigenic formation from solution (Birsoy, 2002). Singer (1984) believes that significant amounts of pedogenic palygorskite can be associated with the following three situations:

- a) paleosols that include distinct and sharp textural transitions,
- b) pedogenic features, not necessarily associated with soils, such as calcretes, crusts and caliches,
- c) modern soils that are currently or have in the past been affected by rising groundwater.

Groundwater and soil pore water in arid regions frequently have high concentration of Mg^{2+} and H_4SiO_4 . Depending on the activity of aluminum, either Mg-smectite, or palygorskite or sepiolite may form (van Breeman and Buurman, 2003). Hodge et al. (1984) reported the in situ formation of palygorskite and smectite in an environment subjected to prolonged

seasonal inundation by surface waters and groundwaters in South Australia (Hodge et al., 1984). Because of the changes in environmental conditions in different saline soils, various types of clay minerals such as smectite and palygorskite may form. Mahjoory (1979) proposed concurrent formation of smectites and palygorskite in the Natrargids of southern Iran due to high electrolyte content of soils studied (Mahjoory, 1979). In contrast, marked degradation of smectite in salt affected soils has been widely reported. Abtahi (1977) proposed the alteration of smectite into palygorskite under highly saline and alkaline conditions in soils of southern Iran (Abtahi, 1977). Reid (1992) attributed the decrease in smectite content of some Californian soils to the increase in salinity (Reid, 1992). While there is enough information about the distribution and genesis of palygorskite in Iranian soils as well as for other arid soils elsewhere in the world, very few studies have investigated the effects of rising groundwater on the formation and stability of palygorskite. This study aims to identify the influence of saline and alkaline groundwater on the formation and stability of palygorskite and associated clay minerals along a calcareous catena in Great Kavir Basin, central Iran.

2. MATERIAL AND METHODS

The study area is located in Great Kavir Basin, Central Iran (Figure 1). Thirty five percent of the basin is composed of intricately folded Miocene sediments which have been eroded to a peneplain surface. Interfingering within the peneplain surface are salt encrusted depressions (Krinsley, 1970). The climate in the study area is continental with the mean annual temperature of 20.8 °C. The soil temperature regime is estimated as thermic bordering hyperthermic

according to USDA Soil Taxonomy (Soil Survey Staff, 2006). The mean summer and winter temperatures (1986-2003) are 32.8 °C and 8.1 °C, respectively. The area receives less than 100 mm precipitation annually. Genetic horizons of four pedons having different depths of water table (5, 97, 142, and 182 cm) were sampled along a calcareous catena near the town of Jandaq, northeast of Isfahan (Figure 1). The soils were described according to Schoeneberger et al. (2002) and classified (Table 1) based on the criteria of USDA Soil Taxonomy (Soil Survey Staff, 2006). General physicochemical characteristics of soils including particle size distribution, electrical conductivity (EC), pH, soluble Si, Mg and Ca, organic carbon (OC), gypsum and carbonate contents were determined using standard procedures (USDA-NRCS, 1996). The chemical compositions of all the samples were plotted on a stability diagram as proposed by Weaver and Beck (1977) to understand whether the environment was conducive for palygorskite formation in the study area (Weaver and Beck, 1977). Prior to mineralogical analysis, soil samples and a sample from Miocene sediments as the soil parent material in the study area, were oven-dried at 105 °C. After removal of gypsum by repeated dissolution in distilled water, carbonates, organic matter and Fe oxides were also removed using 1N sodium acetate buffered at pH 5, 30% H₂O₂ and citrate-bicarbonate-

dithionate (CBD), respectively (Jackson, 1979). Clay fractions of treated samples were separated by centrifuge (Jackson, 1979) and oriented slides were prepared for Mg-saturated, Mg-Saturated and ethylene glycol-solvated, K-saturated, K-saturated and heated at 550 °C treatments. The X-ray diffraction (XRD) patterns of treated clay samples were obtained with a Bruker AXS D8 diffractometer, operated with Cu-K α radiation generated at 40 kV and 30 mA. Palygorskite was identified by 1.04-1.05 nm (001) and 0.636-0.644 nm reflections (002) (Singer, 1989). The semi-quantitative estimation of clay minerals was performed based on their relative XRD peak intensities (Jackson, 1979). Undisturbed sub-samples of all horizons in studied pedons were also examined by a Hitachi S-3500N scanning electron microscope (SEM) equipped with energy dispersive X-ray spectrometry (EDX). The cation exchange capacity (CEC) of the carbonate free clay fractions of all the samples was measured using the BaCl₂ extraction method as described by McKeague (1978).

3. RESULTS AND DISCUSSION

Soils physicochemical characteristics are shown in Table 1. These properties indicate large quantities of soluble salts and carbonates in soils studied. All the soils examined are calcareous, saline and sodic.

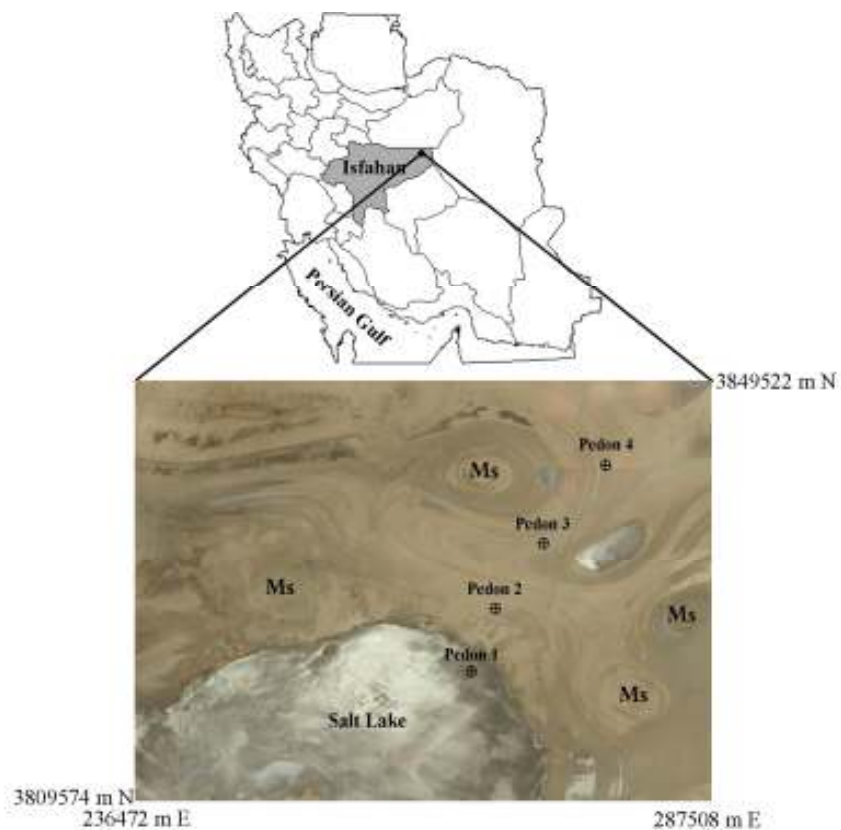


Figure 1. Location of study area and sampling sites in the Isfahan Province, central Iran

Table 1. Selected physical and chemical characteristics of soils and chemical properties of the groundwaters studied

Layer	Depth (cm)	pH	EC (dS m ⁻¹)	H ₄ SiO ₄ (meq lit ⁻¹)	Mg/Ca	OC	Gypsum	Carbonates (g kg ⁻¹)	Sand	Clay	Silt	CEC* (cmole(+) kg ⁻¹ clay)		SAR
Pedon 1 (Typic Aquisalid)														
Az	0-5	7.9	334.8	4.9	0.96	2.9	13	114	109	491	400	43	49.5	
Bzg	+5	8.0	147.9	5.7	1.00	0.5	15	176	120	500	380	45	37.1	
GW ^a	+5	7.9	148.6	1.9	1.05	-	-	-	-	-	-	-	13.8	
Pedon 2 (Typic Aquisalid)														
Az	0-20	8.1	223.6	5.6	0.95	6.5	8	266	200	380	420	56	25.5	
Bz1	20-55	7.7	184.4	4.4	0.51	3.8	10	305	180	393	427	47	18.4	
Bz2	55-75	8.4	119.5	6.5	0.87	4.7	10	340	146	419	435	43	18.3	
Bzm	75-97	7.8	152.8	4.9	1.35	1.1	13	271	154	409	437	40	14.5	
Bzg	+97	8.0	113.9	5.9	1.31	1.9	19	369	145	421	43.4	34	12.9	
GW	+97	7.9	144.6	2.3	1.38	-	-	-	-	-	-	-	14.1	
Pedon 3 (Typic Haplosalid)														
Ayz	0-20	8.0	167.7	5.2	0.45	1.8	85	385	350	360	290	48	21.4	
Byz1	20-65	8.1	181.5	6.4	0.67	4.2	121	464	180	370	450	45	27.1	
Byz2	65-95	7.8	210.8	4.8	1.71	3.6	75	525	145	401	454	46	25.6	
Bz	95-142	8.8	243.7	7.6	1.81	8.8	18	655	190	397	413	39	24.6	
Bzg	+142	8.1	135.9	6.6	2.00	5.9	13	468	90	430	480	36	12.0	
GW	+142	7.9	138.6	2.0	1.42	-	-	-	-	-	-	-	14.0	
Pedon 4 (Typic Haplosalid)														
Az	0-14	7.8	40.4	4.4	0.47	2.6	5	260	131	439	430	62	16.6	
Bz1	14-43	8.0	190.7	5.2	1.24	1.3	7	434	102	465	433	57	15.9	
Bz2	43-71	7.7	55.1	4.8	1.31	3.6	11	486	98	452	450	48	15.6	
Bz3	71-115	7.7	62.5	6.4	1.08	1.5	18	715	300	342	358	41	25.9	
Bz4	115-182	7.9	185.7	7.2	1.13	1.9	10	686	290	369	341	39	31.1	
Bzg	+182	8.2	148.8	7.0	1.26	2.4	27	342	155	485	360	33	13.0	
GW	+182	7.9	137.6	2.1	1.39	-	-	-	-	-	-	-	14.5	

^aGW = groundwater

Based on the EC values, a salic diagnostic horizon was recognized in all the pedons studied. The clay mineralogy of pedons along the catena is summarized in Table 2. The XRD results revealed the presence of quartz, feldspars, smectite, illite, chlorite and palygorskite with various quantities in different horizons and pedons. The quantity of palygorskite increases with depth as that of smectite and quartz decreases (Table 2). Palygorskite was not detectable using XRD in surface horizons of all pedons studied. The diffraction intensities of illite and chlorite clay minerals remained relatively constant in surface and subsurface horizons of all pedons. Two main origins for palygorskite have been stated: a) inheritance from parent materials (Khademi and Mermut, 1998; Viani et al., 1983), and b) pedogenic formation (Birsoy, 2002; Hillier and Pharande, 2008). The feasibility of inheritance origin and pedogenic formation of palygorskite in the study area is discussed.

3.1. Inheritance of palygorskite from parent materials

Clay mineralogy of neighboring Miocene deposits (Ms), as the major parent materials of the soils studied, showed the presence of palygorskite in these sediments (Table 2). This supports the idea that at least part of palygorskite in soils can be of inheritance origin. Khademi and Mermut (1998) reported a detrital origin for palygorskite in gypsiferous alluvial

soils of central Iran. Viani et al. (1983) proposed an inherited origin for palygorskite and other clay minerals in soils with shallow water table in central basin of Saudi Arabia.

3.2. Pedogenic formation of palygorskite

Pedogenic formation of palygorskite can be explained by two processes: (i) alteration of a pre-existing mineral such as illite, smectite and kaolinite, and (ii) precipitation from solution. No trace of kaolinite was found in soils studied, rejecting the likely formation of palygorskite from kaolinite in the study area. The diffraction intensity of illite was relatively constant with depth in all the pedons studied also suggesting no alteration of illite to palygorskite in the study area. In all the pedons studied, a discrete and relatively well-crystallized smectite, which effectively collapses with K saturation and heating (data not shown), dominates the Az horizons clay mineralogy (Table 2 and Figure 2). Little, if any, trace of palygorskite is evident from the Az diffractograms. In contrast, the 1.05 nm palygorskite reflection intensity increases steadily in the XRD patterns of the lower horizons, commensurating with a decrease in the 1.7 nm peak of smectite (Figure 2 and Table 2). The mineralogical trends found in the solum of pedons studied suggest an inverse correlation between the relative diffraction intensity of palygorskite and that of smectite (Figure 2).

Table 2. Relative abundance of different clay minerals in the carbonate-free clay fraction of the pedons studied and that of the parent sediments

Horizon	Pal	Sm	Kfs	Chl	Ill	Qtz ^{b)}
Pedon 1 (Typic Aquisalid)						
Az	“xx”	xxx	x	xx	xxx	xx
Bzg	xx	xxxx	x	xx	xxx	x
Pedon 2 (Typic Aquisalid)						
Az	-	xxxx	xx	x	xx	xxx
Bz1	-	xxx	x	xx	xx	xx
Bz2	x	xxx	x	xx	xx	xxx
Bzm	xx	xx	x	xx	xx	xxx
Bzg	xxx	x	x	xx	xxx	xx
Pedon 3 (Typic Haplosalid)						
Ayz	-	xxx	xx	xx	xxx	xxx
Byz1	-	xxx	x	xx	xxx	xxx
Byz2	x	xx	x	xx	xxx	xxx
Bz	xx	xx	x	xx	xxx	xx
Bzg	xxxx	x	x	xx	xxx	x
Pedon 4 (Typic Haplosalid)						
Az	-	xxxx	xx	xx	xx	xxx
Bz1	-	xxx	x	xx	xx	xxx
Bz2	-	xxx	x	xxx	xxx	xx
Bz3	x	xx	x	xxx	xxx	xx
Bz4	xx	xx	x	xx	xxx	xx
Bzg	xxxx	x	x	xx	xxx	xx
Parent sediments (Miocene, Ms)						
-	xx	xxx	-	xx	xx	xx

a) Relative quantities: xxxxxx = > 70%, xxxxx = 50-70%, xxxx = 30-50%, xxx = 20-30%, xx = 10-20%, x = 5-10%

b) Pal, palygorskite; Sm, smectite; Kfs, K-feldspar; Chl, chlorite, Ill, illite; Qtz, quartz

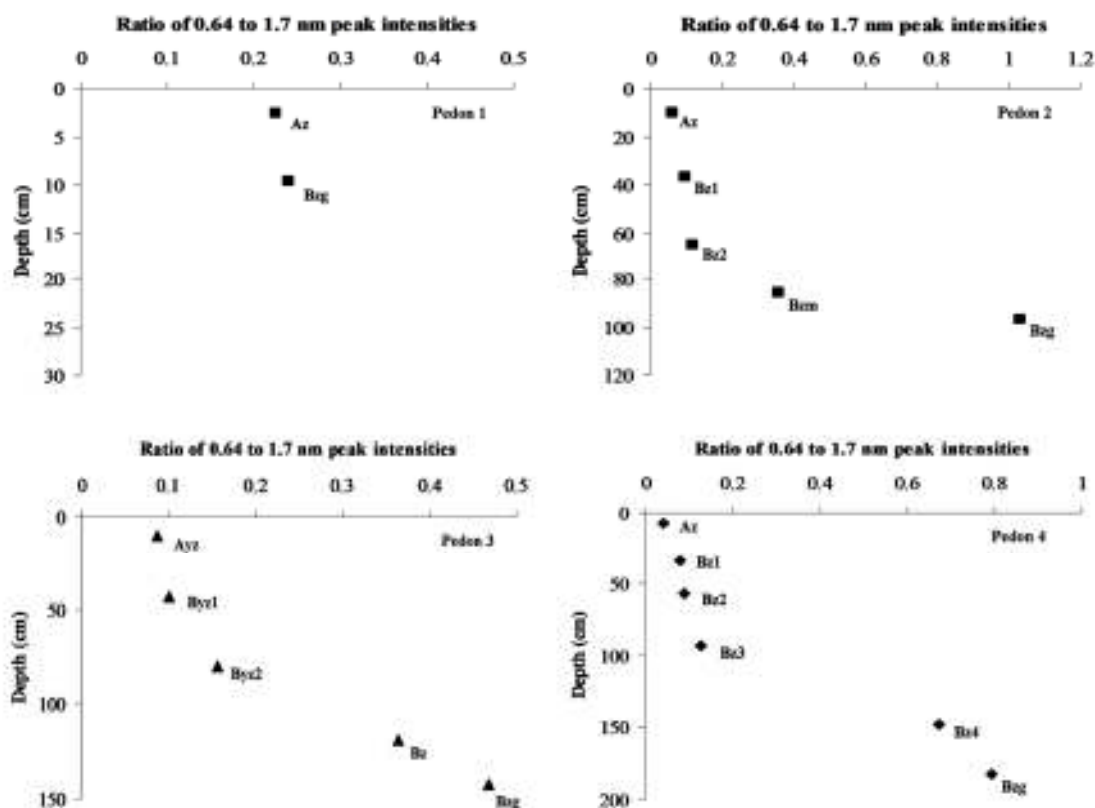
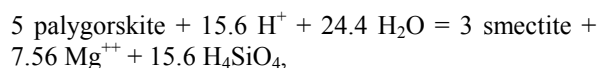
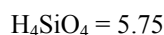


Figure 2. Changes in the ratio of 0.64 nm (palygorskite) to 1.7 nm (smectite) XRD peak intensity of the Mg-saturated ethylene glycol solvated clay samples with depth in pedons studied

The inverse palygorskite/smectite quantitative relationships in the pedons suggest the transformation of smectite to palygorskite as also reported for Texas High Plain soils (Bigham et al., 1980) and Eastern Saudi Arabian Aridisols (Lee et al., 1983). If smectite is forming from the weathering products of palygorskite, the expanding derivative is not the high Mg- mineral. The CEC values of clay fractions in subsurface horizons are consistently less than those of upper horizons (Table 1). The CECs of relatively pure palygorskite samples range from 5 to 30 cmole (+) kg⁻¹ (Singer, 1989). Relatively low CEC values of the clay fractions of the subsoils studied are further indicators of high amounts of palygorskite and the absence of smectite, as discussed earlier. To evaluate whether the environment of sedimentation was favorable for the formation of palygorskite from smectite in the study area, the chemical composition of soils studied was plotted on a mineral stability diagram (Figure 3) as proposed by Weaver and Beck (1977). The stability diagrams depict the tendency towards a more stable mineral phase (Bouza et al., 2007). The proposed equation is as bellow:



$$\text{Log } K_{\text{smectite-palygorskite}} = \text{log } \text{Mg}^{++} + 2 \text{ pH} + 2 \text{ log}$$



Results indicated that the lower horizons which are enriched in palygorskite fall within the stability field of palygorskite. The surface horizons of all the pedons studied fall in the smectite stability field (Figure 3a) and other horizons fall near the boundary line. This suggests that in horizons close to the water table, geochemical conditions are favorable for the stability of palygorskite and probably its formation from smectite. All the surface horizons with an abundance of smectite fall in the smectite stability field suggesting that in these horizons palygorskite is not stable and tends to alter to smectite. Interestingly, some subsurface horizons with predominant occurrence of smectite fall within the stability field of palygorskite. This may imply that a transformation/alteration state is acting toward the formation of palygorskite against that of smectite stability. However, more detailed studies are needed to verify this hypothesis. Singer et al. (1995) reported similar results suggesting that the clay minerals are in an alteration/transformation state within other mineral phases caused by climatic changes during the Quaternary. Using equilibrium diagrams, Bouza et al. (2007) reported that in Aridisols of Patagonia region in Argentina, palygorskite and sepiolite are the most stable phases in clacic and petrocalcic horizons, respectively. Hillier and Pharande (2008) showed that

saturation paste composition of saline-sodic and normal soils of Maharashtra region in India, plot in palygorskite and smectite stability fields, respectively. The concentration of soluble Mg, Si and pH values of soils as a function of depth (Figures. 3b and 3c) revealed that Bzg horizons have the highest values. In contrast, surface horizons in pedons 2, 3, and 4 exhibit the lowest values of these parameters. These data along with those obtained from the stability diagram indicate that palygorskite in soils studied is likely a product of interaction between Mg-rich groundwater and smectite minerals. It is assumed that under the alkaline pH of 8 to 9 and a high concentration of Si and Mg, the formation and stability of palygorskite are favored (Singer, 1989). The main evidences for palygorskite formation from solution in the subsurface horizons are explained. Scanning electron micrographs show the growth of palygorskite fibers in pore spaces (Figures 4a and 4d) implying its in situ formation from solution. Also the direct proximity of calcite crystals and palygorskite fibers (Figure 4c) suggests that palygorskite were formed after crystallization of calcite minerals in soils studied.

The relationship between calcite and palygorskite is the action of calcite particles in maintaining a high

Mg level. After CaCO_3 precipitation, the Mg concentration in soil solution increases, combining with high concentration of Si due to a high pH, the formation of palygorskite from solution proceeds. Bouza et al. (2007) showed that during precipitation of low-Mg calcites the activity of soluble Mg in soil solution increases which favors the transformation of smectite to palygorskite. Geochemistry of subsurface horizons rich in palygorskite showed that the concentration of soluble Mg and Si is significantly higher than that of surface horizons (Table 1, Figures 3b and 3c). The pH values of lower horizons are in the range of 8 to 8.2. Geochemical analyses of underground water in the study area also showed a pH of 7.9, and a relatively high concentration of Mg and Si (Table 1). This suggests that under arid climates such as that in the study area, palygorskite is stable and probably precipitates from solution. Mahjoory (1979) believes that the formation of palygorskite in soils of southern Iran can be attributed to a highly alkaline and saline condition in soils which promotes the solubilization of silica and alumina from feldspars. He suggested that these soluble constituents combine with soluble bases and form palygorskite.

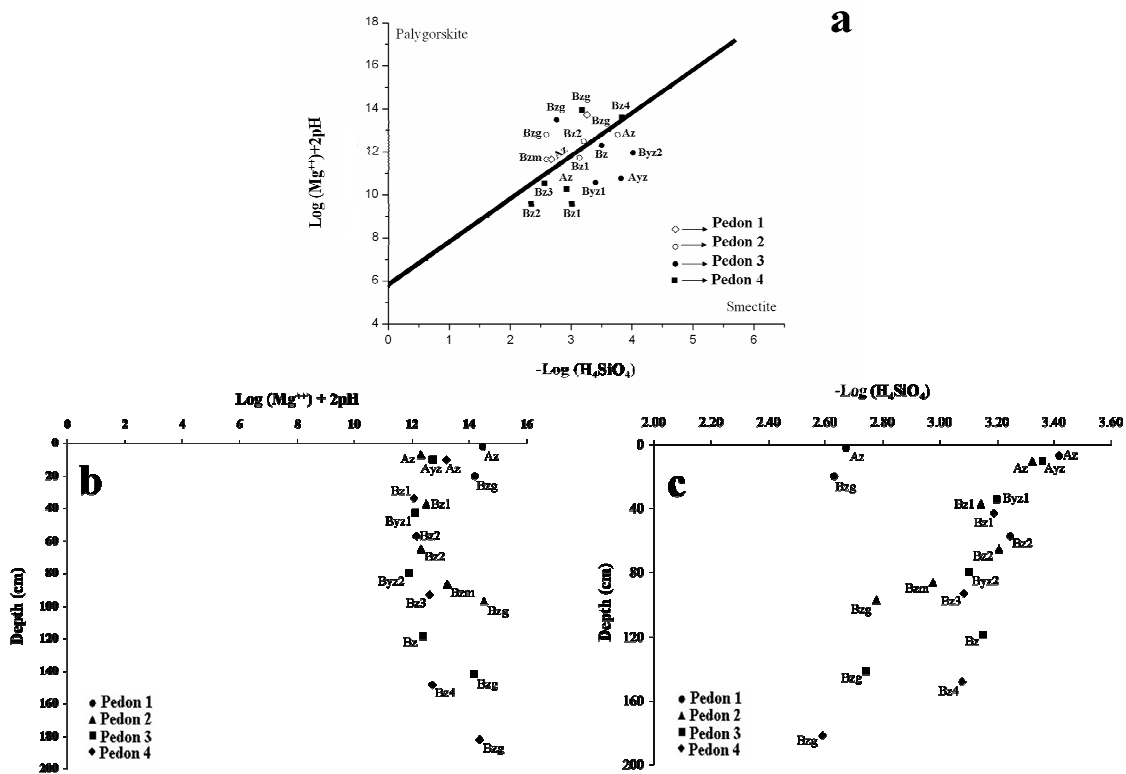


Figure 3. (a) Stability diagram for palygorskite-smectite system and plotted composition of samples taken from studied pedons; (b) and (c) changes in $\text{Log (Mg}^{++}) + 2\text{pH}$ and $-\text{Log (H}_4\text{SiO}_4)$ values with depth in the pedons studied

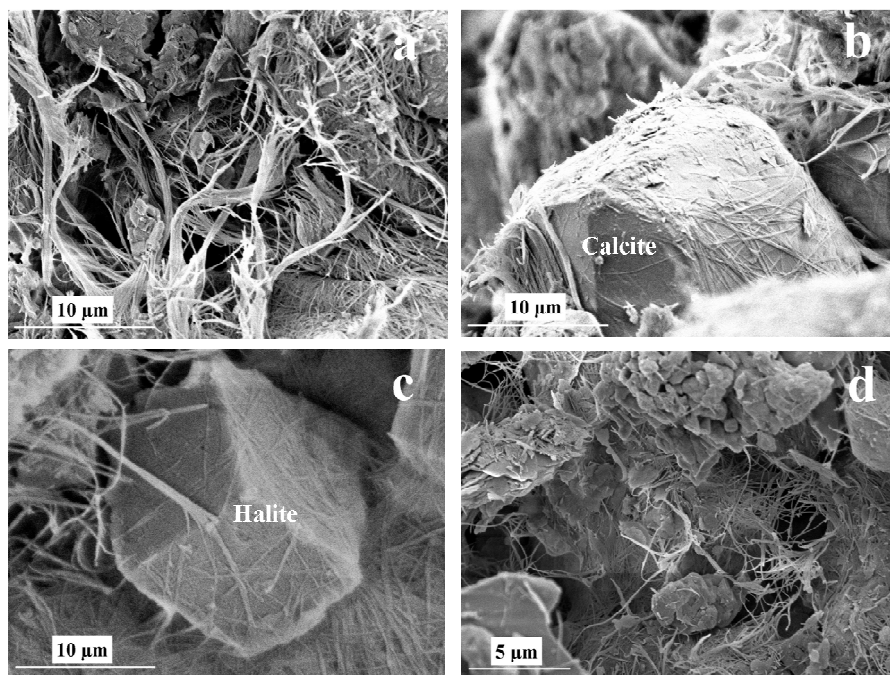


Figure 4. (a) Growth of palygorskite fibers showing the neoformation of this mineral in the Bzg horizon of pedon 2, (b) and (c) calcite and halite grains of the Bzg horizon of pedon 2 and 4, as determined by energy dispersive X-ray spectroscopy (data not shown), covered by palygorskite fibers, (d) formation of palygorskite fibers in pore spaces between aggregates of the Bzg horizon of pedon 4

4. CONCLUSIONS

Palygorskite has two main origins of inheritance from parent materials and pedogenic formation. In subjacent horizons of all pedons studied, very close to the water table, there was a higher amount of palygorskite when compared with that in surface horizons. Stability diagrams of palygorskite-smectite system showed that palygorskite was more stable in subsurface horizons and probably is a product of smectite alteration. In pedon 1 with very shallow water table, palygorskite coexists with smectite. It seems that under such condition, palygorskite forms directly from the solution. However, in other pedons, with deeper water table, palygorskite forms probably from degradation of smectite in a saline-sodic environment. This is further confirmed by lower amounts of palygorskite and higher amounts of smectite in surface horizons. The relative increase of smectite in the surface horizons of pedons 2, 3 and 4 may be due to concentration by removal of palygorskite via weathering. Growth of palygorskite fibers on halite and calcite crystals, as determined by SEM observations, clearly represents the in situ formation of palygorskite. Chemical composition of groundwater rich in soluble Mg^{2+} and silica may suggest that formation of palygorskite was promoted in soil horizons with permanent occurrence of groundwater. Other clay minerals appear to have derived mainly from parent materials.

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