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Reclamation of a Sandy Desert Through Floodwater Spreading: II. Characterization of Clay Minerals in the Watershed and the Freshly-Laid Sediment

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ABSTRACT

The artificial recharge of groundwater (ARG), where practicable, is an easy and economical method for desertification control in the arid and semi-arid zones where over-pumping has critically lowered the watertable. Although desiltation of floodwaters, which is a prerequisite for the ARG, leads to rehabilitation of the eroded sites that house the sedimentation basins (SBs), the gradual impermeability of these basins and infiltration ponds by migrating phyllosilicates shortens the economic life of such projects. Therefore, characterization of these minerals is necessary if treatments for their removal are to be found. As soil erosion in the watershed that supplies the ARG systems with floodwater deteriorates the land, and threatens the recharge facilities and other hydraulic structures downstream with siltation, identification of the erosion source facilitates prioritization of soil conservation activities. Clay mineralogical analysis has been developed in recent years as a powerful tool in sediment source attribution. However, applicability of such methods to cases in Iran requires regional studies. Sediment samples were collected at 0-10,10-20, and 20-30 cm increments on two transects in the 1st, 4th and 6th SBs in a Gareh Bygone Plain ARG system. Rock samples were collected on the southern flank of the Bisheh Zard Basin that supplies floodwater to the ARG systems. All samples were characterized by the XRD method. Transmission-electron micrograph (TEM) of one sample was also obtained. Chlorite, mica, smectite, and possibly kaolinite were detected in all samples. The presence of palygorskite was revealed only in a TEM. The non-clay minerals were calcite, dolomite, alkali and plagioclase feldspars, gypsum, halite and quartz. The common origin of these inherited clay minerals precludes their utility in sediment source detection.

Keywords: The Agha Jari Formation, Aquifer Management, Erosion, Gareh Bygone Plain, Sedimentation.

INTRODUCTION

The man-made drought due to overpumping of groundwater in arid areas, where most of the water needs are supplied through underground resources, invariably leads to desertification. It is therefore, obvious that the artificial recharge of groundwater (ARG) and its controlled withdrawal is the most rational method of desertification control in such environments. This will ensure sustainable yield of wells and qanats (underground water collection and transmission galleries) and prevent saltwater intrusion into the coastal and inland aquifers. Considering that flash floods provide a very important, and sometimes the sole source of water for the ARG in arid and semi-arid environments, and as these waters are often highly laden with sediment, construction of the ARG facilities entails provision of carefully designed sedimentation basins (SBs) which

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clarify the turbid waters and lengthen the economic life of infiltration ponds. Floodwater spreading on marginal land that overlie the potential aquifers is a very easy and economical method of the ARG while simultaneously achieving reclamation of the eroded soils, particularly the drifting sand (Kowsar, 1991).

Alluvial cones, colluvial deposits, coarse-grained alluvial fans, even sand dunes, usually among the least productive agricultural soils due to their very high infiltration and percolation rates, low available water holding capacity, and meager nutritive status form the most suitable recharge sites. The sediment collected in the SBs, which under other circumstances is considered a waste and pollutant, and therefore problematic (Brown *et al.,* 1974), is actually a valuable resource that may be used to rehabilitate the eroded soil by increasing its depth and overall productivity (Carter, 1976).

Phyllosilicates, clay-size particles and microorganisms are among the major problems associated with the ARG systems, as these particulate matter remain in suspension for a long time, migrate towards the watertable, eventually clog the soil, and virtually terminate the useful life of the SBs and infiltration ponds. It is, therefore, necessary to characterize the clay minerlas and other potentially objectionable matter to be able to develop techniques to trap them in the SBs. Moreover, as the recharge sites in Iran are usually put under irrigated cultivation after they have acquired the needed soil depth through sedimentation, have abundant ferruginous cherts, are infested with termites, and are planted to deep- rooted trees, it is essential to be able to predict the changes which are likely to take place in these soils. Alteration and/or neoformation of clay minerals is one of these probable changes.

Jackson (1969) stated that an environment abundant in exchangeable Ca^{2+} and Mg^{2+}, silica- rich, moist, and not excessively leached, favors the formation of montmorillonite. lvarson *et al.* (1978) reported the formation of jarosite from glauconite, illite and microcline during the oxidation of ferrous iron by *Thiobacillus ferrooxidans.* Boyer, as reported by Robert and Berthelin (1986), observed that termites can devide mica into illite, and transform mica into vermiculite, or even smectite. Spyridakis *et al.* (1967) reported the formation of kaolinite from biotite by the seedlings of white cedar, hemlock, white pine, white spruce, red oak and hard maple. These, along with obscure biological processes, might evolve minerals unknown at the ARG sites. Thus, identification of the present clay minerals is an important part of establishing a baseline against which future soil development may be measured, and characterization of a future bench mark soil (Miller and Nichols, 1979) on which long term research projects will be performed.

Although the present study is restricted to a 250 km^2 subwatershed of the 48500 km^2 Mand River Basin (MRB) it has relevance for management of numerous potential ARG sites with identical geological formations and similar environment in southwestern Iran. Furthermore, construction of at least two large dams on this basin has been planned. It is, therefore, essential to identify the sediment sources and to treat them according to the intended uses of the water and the suspended load. Clay mineralogical analysis is a rather direct, simple, and economical method for tracing the sources of erosion if there are sufficient differences in soils or geological formations within and between watersheds (Laird and Harward, 1982; Hsieh, 1984), if no preferential clay erosion occurs (Rhoton *et al.,* 1979), and if no transformation of clay species in the river environment takes place (Murad and Fischer, cited in Rhoton *et al.,* 1979). However, applicability of this method to the watersheds in Iran requires regional studies.

The objectives of this ongoing study are to (i) characterize the clay minerals in a subwatershed of the MRB for evaluation of the method on its applicability in Iran for sediment source identification, and (ii) charcterize the clay minerals of the freshly-laid sediment to be used later in the design of the SBs.

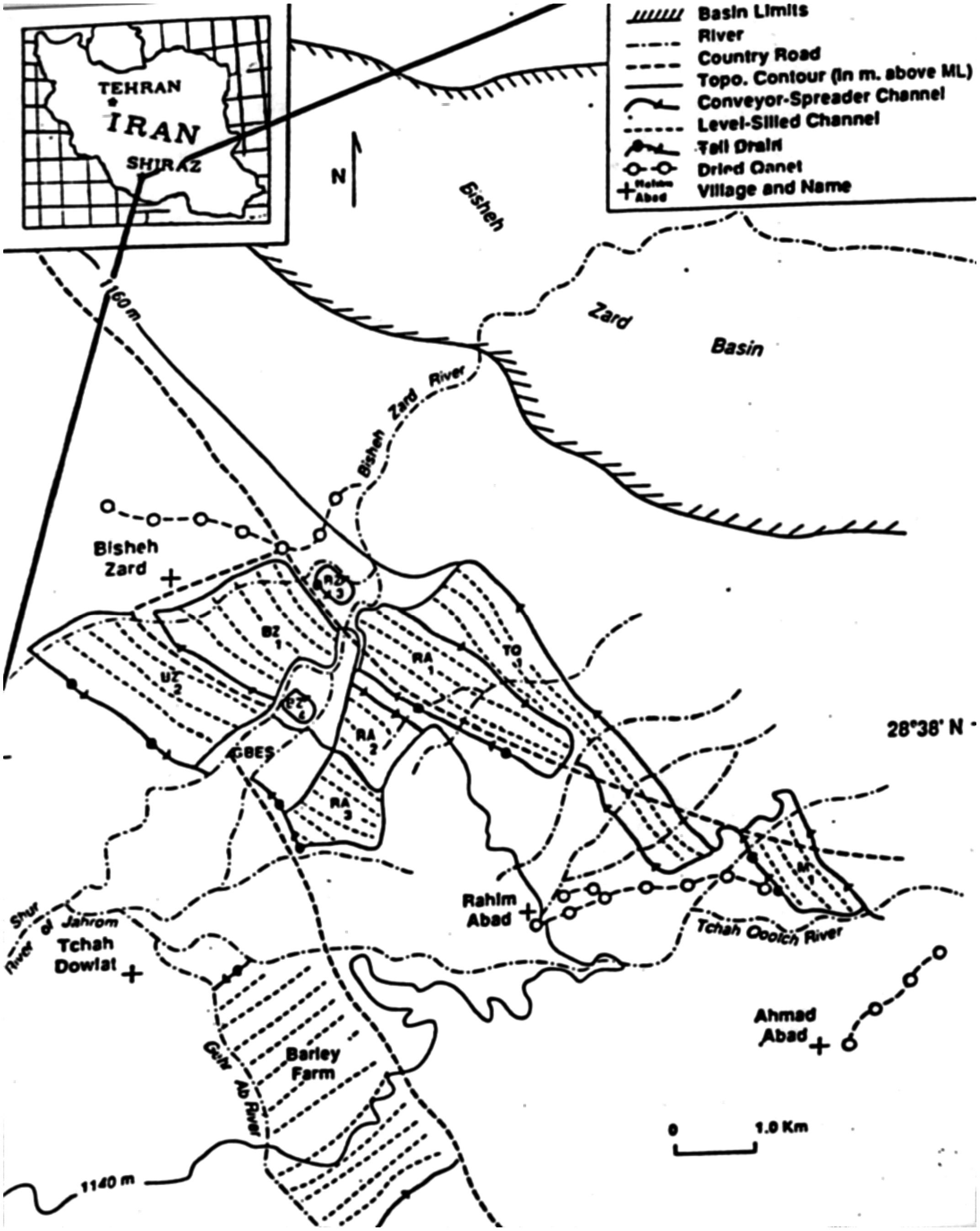

Figure1. Sketch map of the Gareh Bygone Plain floodwater spreading systems.

MATERIALS AND METHODS

Study Area

The study area is 200 km southeast of Shiraz, Iran (28 ^{0}38′ N, 53^{0} 55′ E) in the 192 km^2 Bisheh Zard Basin (BZB) and on the alluvial fan formed by Bisheh Zard River which

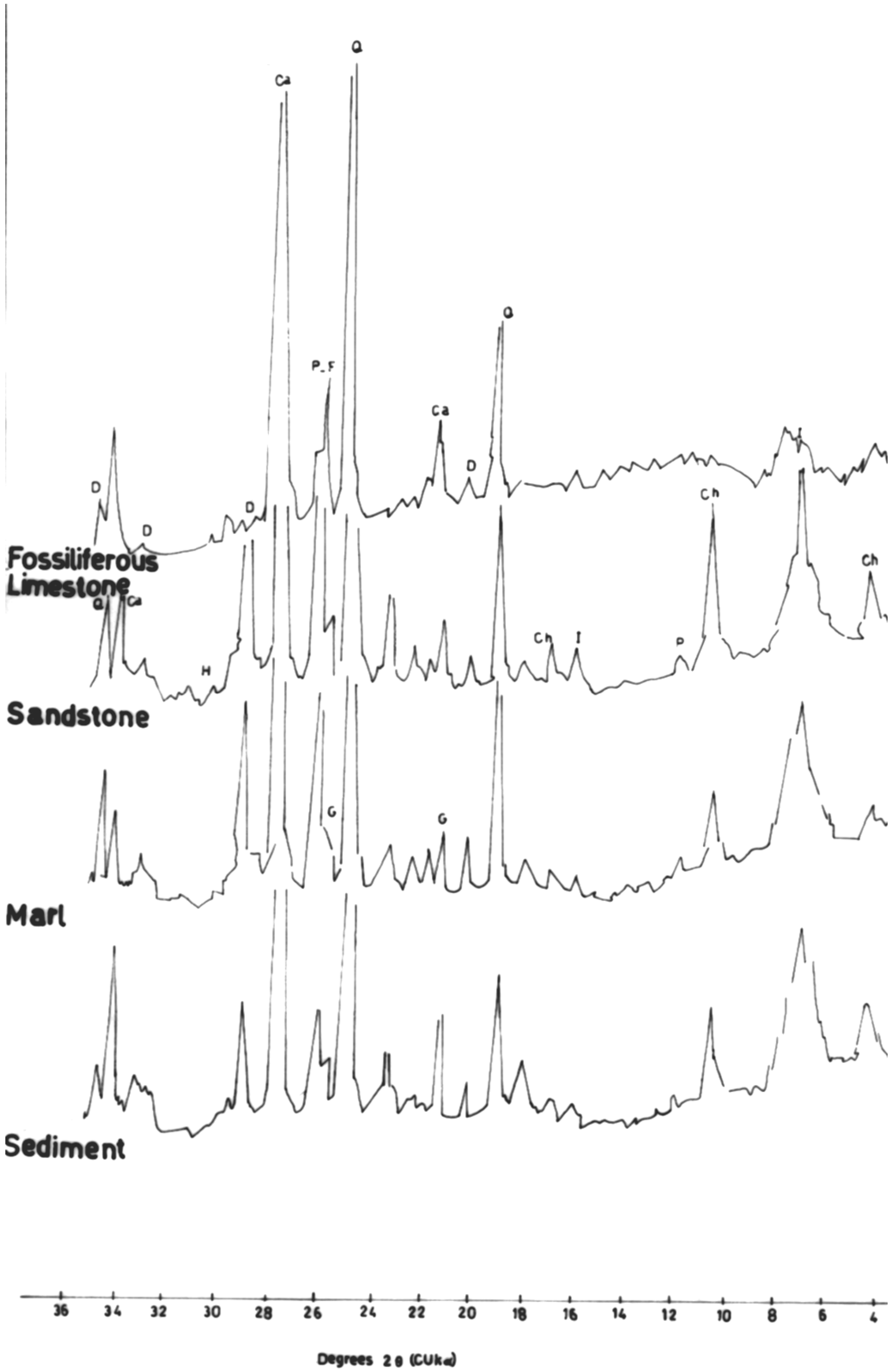

Figure 2. XRD patterns for the <2 μm fraction of powdered rocks from the watershed and the alluvium in a sedimentation basin. Ca:calcite; Ch:chlorite; D:dolomite, F: alkali feldspars; G:gypsum; H: halite; I: iIlite; P:plagioclase; Q: quartz.

drains the Basin. Elevation of the site ranges from 1585 m in the BZB to 1140 m in the lowest SB (Figure1). The BZB is a northwest -southeast syncline formed by the tectonic movements of the Zagros Mountain Ranges during the Mio -Pliocene time in the Agha Jari Formation (AJF). This formation consists of the rhythmically interbedded calcareous sandstones, and low weathering, gypsum - veined red marls and grey to green siltstones (James and Wynd, 1965). Scattered patches of the Plio-Pleistocene Bakhtyari Formation, a calcareous conglomerate with ferruginous cherts (Stocklin and Setudehnia, 1977) are also found in the BZB. A more detailed geologic and geomorphic description of the watershed has been given by Kowsar (1991).

The alluvial fan is covered with a layer of the drifting fine sand ranging in thickness from a few mm to several cm. The soil of the site has been classified as a coarse - loamy, over loamy skeletal carbonatic (hyper) thermic Typic Haplocalcids (Soil Survey Staff, 1996). Detailed descriptions of the soil present on the alluvial fan are reported elsewhere (Kowsar, 1991; Naderi *et al.,*2000).

Field Methods and Sampling

Rock samples were collected from 57 discernible beds on the southern flank of the Bisheh Zard syncline on the north 20′ due east transect. Each sample was numbered and put inside a plastic lined canvas bag. The samples were shipped to the Sedimentology Laboratory, College of Science, Tehran University, for identification and further analyses. Soil samples were collected at 0-10, 10-20, and 20-30 cm increments along two parallel transects from the 1st, 4th, and 6th SBs in the Gareh Bygone Plain ARG project site, Bisheh Zard$_1$ (BZ$_1$) system (Figure 1). Each sample was put into a tagged, plastic lined canvas bag and shipped to the Soil and Water Research lnstitute Laboratories in Tehran for analyses.

It should be realized that since the study site is an alluvial fan and the alluvium is uniform to the observed depth of 1.50 m, therefore, the sample from SB$_1$, which is the first sedimentation basin, may be considered as the freshly-laid sediment, while those from SB$_4$ and SB$_6$, which have been covered with a relatively small thickness of the suspended load, may function as the control.

Laboratory Methods

Particle size separation for mineralogical analysis was performed according to the procedures outlined by Kunze and Dixon (1986) with the following modification: $CaCO_3$ removal was attempted by the repeated washing with I *N* NaOAc at pH=5, and centrifugation for 10 min at 2500 rpm. Cation saturation, glycerol solvation and clay slurry sedimentation on glass slides were achieved according to Whitting and Allardice (1986). Furthermore, to identify the minerals in the AJF components and the freshly-laid sediment a powder sample of each was also x-rayed without any prior treatment.

XRD patterns were obtained using CuKa radiation and Ni-filter at kV=40, mA = 40, and , λ = 0.15418 nm. A drop of citrate - dithionite treated clay suspension was deposited on a Cu microgrid. The TEM studies were performed with a Phillips SM 300 operating at 80 P kV.

RESULTS AND DISCUSSION

A strong similarity among the powder XRD patterns of the three major rock types of the AJF and the alluvium derived from them (Figure 2) suggests not only the common parent materials of the rocks and their contribution to the sediment formation, but also the hereditary nature of the clay species as well.The mineralogy of the <2 μ m fraction consists of chlorite, mica (illite), quartz, dolomite, calcite, gypsum, halite, plagio-

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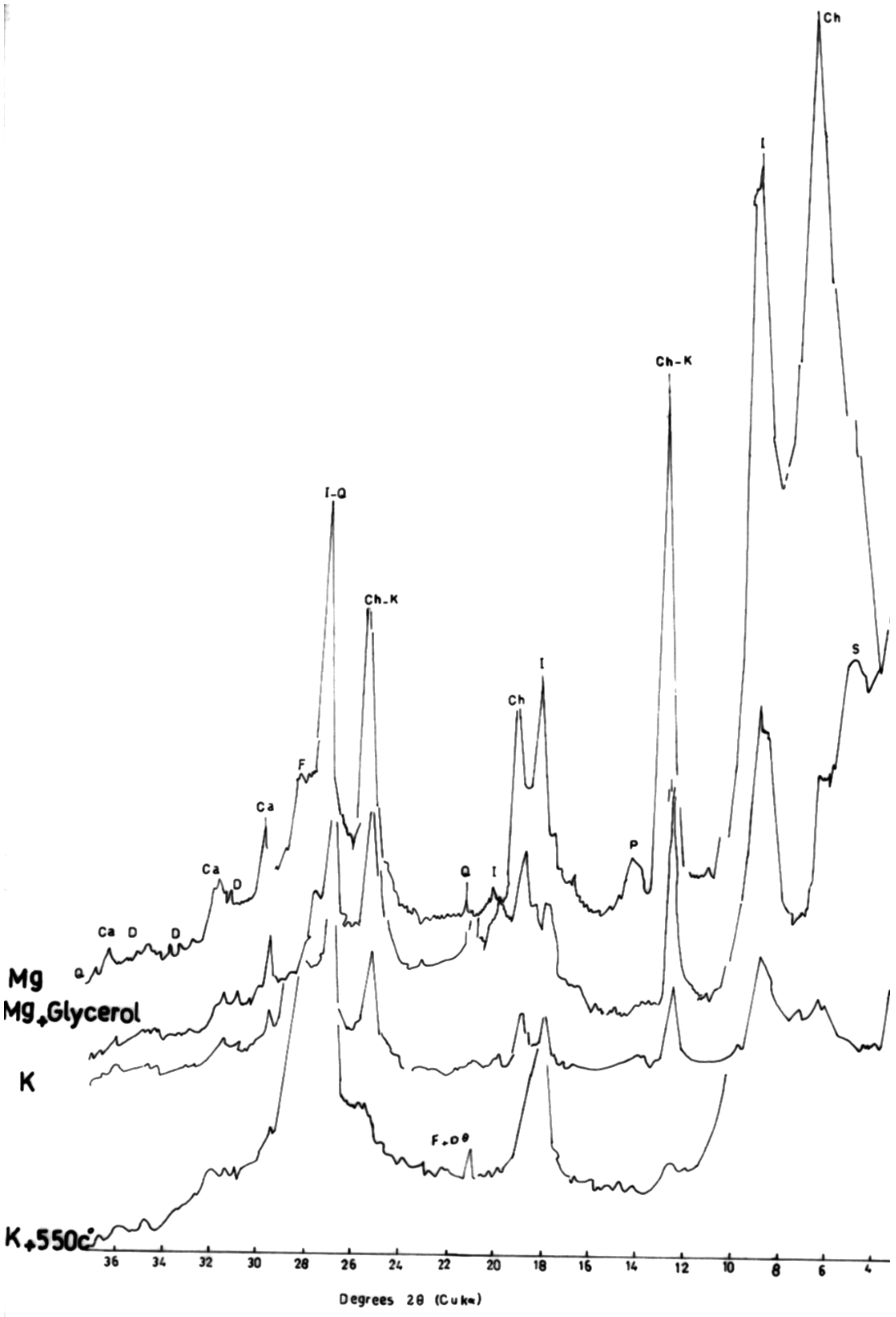

Figure 3. XRD patterns for the < 2 μm fraction of the treated alluvium from the first sedimentation basin. Ca:calcite; Ch:chlorite; D: dolomite, F: alkali feldspars; S:smectite. I: illite; K:kaolinite; P:plagioclase; Q: quartz

clase and alkali feldspars and expandable minerals.

The <2 μm fraction of the treated sediment sample consisted of chlorite, a species of smectite, mica, and possibly kaolinite. It is worth emphasizing that the XRD patterns of all the 18 samples taken from the surface 30 cm of the SBs were practically identical. Since there is no difference in the position of peaks and very little variation in their shape and intensity, therefore, only the diagrams of the upper 10 cm of the eastern end of the first SB are presented for discussion (Figure 3).

The presence of chlorite is indicated by a symmetrical, strong d (00/) spacing at about 1.42 nm which is invariant with respect to cation saturation, glycerol solvation and heating to 550^0C for 2 h. The disappearance of the strong, symmetrical 2nd, 3rd and 4th order reflections of chlorite after the heat treatment is an indication of its low Fe content, because if it were an iron rich chlorite it should have given a weak first and third-order reflections, and a strong second, and fourth-order reflections (Barnhisel and Bertsch, 1989). The first-order reflection has become only a little less intense relative to mica (illite) after the heat treatment. A shoulder on the low angle side of the chlorite peak with Mg^{2+} saturation which expanded to 1.840 nm with glycerol solvation and collapsed to 1.00 nm with K^+ saturation and heating to 550 ^{0}C for 2 h points to the presence of smectite. The collapse of 0.71 and 0.35nm after a 2- h heat treatment at 550^0C may indicate the presence of kaolinite, as the strong, symmetrical 2nd, 3rd and 4th orders of chlorite reflection had masked the presence of kaolinite. However, not a single flake of kaolinite was shown in the TEM.

The strong 1.00 nm spacing with Mg^{2+} saturation and solvation is indicative of mica (illite). The presence of a trace amount palygorskite is shown by a few fibers in a TEM (Figure 4). The electron micrograph reveals the fibers along with particles having smooth, wavy edges which may be indicative of smectite. The non-clay minerals distinguishable in the XRD patterns are: calcite, dolomite, gypsum, halite, alkali and plagioclase feldspars and quartz (Figures 2 and 3). It should be emphasized that the complete removal of $CaCO_3$ from calcareous materials is difficult; thus, the presence of calcite in the XRD patterns is not unusual.

The occurrence of these clay species in the soils of Fars province has been reported by Abtahi (1977, 1980) and Gharaee and Mahjoory (1984). However, the relative abundance of palygorskite in those soils and its scarcity at the research site is anomalous. Not a single 1.05- 1.07 nm peak, indicative of palygorskite, is evident in the X-ray diffractograms. This maybe due to the youth of the soil formed on the watershed then deposited in the SBs.

The Agha Jari clastics and its most likely parent materials, the Jahrom, Asmari, Razak and Mishan Formations, not only cover southwestern Iran, but also extend through the Persian Gulf into Saudi Arabia where they are represented by Ummer Radhuma, Dammam, Hadrukh, Dam and Hufuf Formations, respectively (James and Wynd, 1965; Power *et al.,* 1966). The presence of palygorskite in the above named formations of that country, and the soils derived from them, has been reported by Lee *et al.* (1983) and Shadfan and Mashhady (1985). Although neoformation of palygorskite in the soils of Iran and Saudi Arabia has been reported by Abtahi (1977, 1980) and Elprince *et al.* (1979), respectively, apparently, the conditions at the research site are not conductive to authigenesis of this clay mineral. Stackman and Bishay (1976) concluded from a review of the literature that palygorskite is of lacustrine origin formed by basic sedimentation in arid and semi arid periods as did occur in the fluctuating Pleistocene climate. We speculate that the position of the study site, which is close to the northeastern margin of the AJF, might have been instrumental in blocking palygorskite formation, and/or facilitated its transformation into other clay minerals. Moreover, our limited data indicate that the clay minerals found in the AJF outcrop in the Gareh Bygone Plain originated elsewhere from older formations and then redistributed by alluvial and aeolian processes in the settling basins or on the Plain. This reasoning is in agreement

Figure 4. Palygorskite fibers in assemblage with smectite. Transmission electron micrograph of the sediment sample enlarged 59400 times.

with the suggestion of Lee *et al*. (1983) for the presence of palygorskite in the soils of eastern Saudi Arabia.

PRACTICAL IMPLICATIONS

1. Due to the common origin of clay species in the Tertiary sediment in southwestern Iran, the utility of using clays as tracers in detection of sediment sources is, at most, doubtful.
2. As pore diameter may be approximated by: grain size/6.46 (Huisman and Olsthoorn, 1983), therefore, palygorskite fibers, which are 0.1-0.2 μm in diameter, could easily pass through the pores formed by the smallest silt grains. Coagulation or flocculation of these fibers deep in the profile may eventually make the soil impermeable, thus ending the useful life of infiltration ponds. Therefore, a suitable economic treatment should be designed to settle the clay-size particles in the settling basins.

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REFERENCES

1. Abtahi, A. 1977. Effect of Saline and Alkaline Groundwater on Soil Genesis in Semiarid Southern Iran. *Soil Sci. Soc. Am. J.*, **41**:583-588.
2. Abtahi, A. 1980. Soil Genesis as Affected by Topography and Time in Highly Calcareous Parent Materials Under Semiarid Conditions in Iran. *Soil Sci. Soc. Am. J.*, **44**:329-336.
3. Barnhisel, R.l., and Bertsch, P.M. 1989. Chlorites and Hydroxy-interlayered Vermiculite and Smectite. In *"Minerals in soil environments*". 2nd. Ed. (eds): Dixon, I.B., and weed, S.B., SSSA, Madison, WI.
4. Brown, M.J., Carter, D.L., and Bondurant, J.A. 1974. Sediment in Irrigation and Drainage Waters and Sediment Inputs and Outputs for Two Large Tracts in Southern Idaho. *J. Environ. Qual.*, **3**:347-351.
5. Carter, D.L. 1976. Guidelines for Sediment Control in Irrigation Return Flows. *J.Environ. Qual.*, **5**:119-124.
6. Elprince, A.M., Mashhady, A.S., and Aba-Husayn, M.M. 1979. The Occurrence of Pedogenic Palygorskite (Attapulgite) in Saudi Arabia. *Soil Sci.*, **128**:211-218.
7. Gharaee, H.A., and Mahjoory, R. 1984. Characteristics and Geomorphic Relationships of some Representative Aridisols in Southern Iran. *Soil Sci. Soc. Am. J.*, **48**:1115-1119.
8. Hsieh, Y.P. 1984. Using Clay Mineralogy to Infer Sources of Suspended Clay and Silt in a Watershed: Quantitative Approach. *Soil Sci. Soc. Am. J.*, **48**:1446-1450.
9. Huisman, L., and Olsthoorn, T.N. 1983. *Artificial Groundwater Recharge*. Pitman Advanced Publishing Program. London.
10. Ivarson, K.C.,Ross, G.J., and Miles, N.M. 1978. Alteration of Micas and Feldspars During Microbial Formation of Basic Ferric Sulfates in the Laboratory. *Soil Sci. Soc. Am. J.*, **42**:518-524.
11. Jackson, M.L. 1969. *Soil Chemical Analyses-Advanced Course*. Dep. of Soils. Univ. of Wisconsin, Madison, WI.
12. James, G.A., and Wynd, J.G. 1965. Stratigraphic Nomenclature of Iranian Oil Consortium Agreement Area. *Amer. Assoc. Petrol. Geol. Bull.*, **449**:2182-2245.
13. Kowsar, A. 1991. Floodwater Spreading for Desertification Control: An Integrated approach. *Des. Con. Bull., (UNEP)* **19**:3-1 8.
14. Kunze, G.W., and Dixon, J.B. 1986. Pretreatment for Mineralogical Analysis. In*: "Methods of Soil Analysis"*. Part I. 2nd ed. (ed): Klute, A., Agron. Monogr. 9. ASA and SSSA, Madison, WI. pp. 91-100
15. Laird, D., and Harward, M. E. 1982. Relationship of the Nature of Suspended Clay Minerals to Hydrologic Conditions. *J. Environ. Qual.*, **11**:433-436.
16. Lee, S.Y., Dixon, J.B., and Aba-Husayn, M.M. 1983. Mineralogy of Saudi Arabian Soils: Eastern Region. *Soil Sci. Soc. Am. J.*, **47**:321-326.
17. Miller, F.T., and Nichols. J.D. 1979. Soils Data. In: "Planning the Uses and Management of Land" (eds): Beatty, M.T., Petersen, G.W., and Swindale, L. D. Agron. Monogr. 21. ASA, Madison, WI. pp. 68-69.
18. Naderi, A.A., Kowsar, S.A., and Sarafraz, A.A. 2000. Reclamation of a Sandy Desert Through Floodwater Spreading. I.Sediment-Induced Changes in Selected Soil Chemical and Physical Properties. *J. Agr. Sci. Tech.,* **2**: 9-20.
19. Powers, R.W., Ramirez, L.F, Redmond, C.D., and Elbergm, Jr. E.L. 1966. Geology of the Arabian Peninsula: Sedimentary Geology of Saudi Arabia. USGS Prof. paper 560-D. US Gov. Print. Office, Washington, DC. or pp. 1-20.
20. Rhoton, F.E.,. Smock, N.E., and Wilding, L.P. 1979. Preferential Clay Mineral Erosion from Watersheds in the Maumee River Basin. *J.Environ. Qual.*, **8**:547-550.

21. Robert, M., and Berthelin, J. 1986. Role of Biological and Biochemical Factors in Soil Mineral Weathering. In:“Interactions of Soil Minerals With Natural Organics and Microbes”. SSSA Spec. Pub. 17.(Eds): Huang, P.M. and Schnitzer, M.,SSSA, Madison, WI. pp. 453-496
22. Shadfan, H., and Mashhady, A. S. 1985. Distribution of Palygorskite in Sediments and Soils of Eastern Saudi Arabia. *Soil Sci. Soc. Am. J.*,**49**:243-250.
23. Soil Survey Staff, 1996. *Keys to Soil Taxonomy*. 7th ed. USDA- NRCS. U. S. Gov. Print. Office, Washington, DC.
24. Spyridakis, D.E., Chesters G. and Wilde, S.A. 1967. Kaolinization of Biotite as a Result of Coniferous and Deciduous Seedling Growth. *Soil Sci. Soc. Am*. Proc.,**31**:202-210.
25. Stackman, W.P., and Bishay, B. G. 1976. Moisture Retention and Plasticity of Highly Calcareous Soils in Egypt. *Neth. J. Agric. Sci.*, **24**:43-57.
26. Stocklin, J., and Setudehnia, A. 1977. “*Stratigraphic Iexicon of Iran”.* 2nd ed. Geol. Sur. of Iran. Rep. No. 18. Ministry of Information and Tourism Press, Tehran.
27. Whitting, L. D., and. Allardice, W. R 1986. X- ray Diffraction Techniques. In: “*Methods of Soil Analysis”*. Part I. 2nd (ed) Klute, A.,: Agron. Monogr. 9. ASA and SSSA, Madison. WI. pp. 331-362

کاربرد گسترش سیلاب در آباد کردن بیابان‌های شنی: ۲- شناسایی کانیهای رسی در آبخیز و ته نشستهای تازه

چکیده

تغذیه مصنوعی آبخوانها، در مناطق خشک و نیمه خشکی که آب زیرزمینی آنها براثر آبکشی بیش از اندازه کاستی پذیرفته است، روشی ساده و ارزان برای بیابان زدایی به شمار می رود. هرچند رسوب زدایی سیلابهای گل آلود پیش نیاز تغذیه مصنوعی بوده و پیامد این فرایند بازسازی خاکهای فرسوده رسوبگیرها و استخرهای تغذیه است، ناتراواگشتن این سازه ها براثر انتقال رسها به زیرزمین عمر سودآور آنها را کوتاه خواهد کرد. بدین ترتیب، شناسایی کانیهای رسی برای یافتن روش ته نشین ساختن آنها در سطح زمین لازم است. از آنجا که فرسایش باروری خاک را کاسته و مواد معلق سیلاب را نیز تامین می کند، امکان ردیابی مناطق فرسایش پذیر آبخیز با استفاده از رسها، روشی که در ایالات متحد آمریکا به ثبوت رسیده است، نیز مورد ارزیابی قرار گرفت. ۵۷ نمونه سنگ از جبهه جنوبی آبخیز گر، و ۹ نمونه از ته نشستهای تازه در رسوبگیرهای طرح تغذیه مصنوعی گربایگان، فسا، تهیه شد و کانیهای رسی و غیررسی آنها با کاربرد پراش پرتو X تعیین گردید. از یک نمونه نیز TEM تهیه شد. کلریت، میکا (ایلیت) و اسمکتیت در تمام نمونه ها و جود داشت. کائولینیت در یک مورد مشاهده شد. تنها چند رشته پالی گورسکیت در TEM ملاحظه گردید. کلسیت، دولومیت، فلدسپار، گچ، کوارتز و نمک طعام دیگر کانیها بودند. از آنجا که آغاجاری سازندی آواری است، و مواد سازنده آن از فرسایش سازندهای پیرتر (آسماری، جهرم ...) گرد آمده اند، کانیهای رسی آنها کاملاً یکسان بوده و ردیابی آنها برای یافتن مناطق فرسایش پذیر در زاگرس توصیه نمی شود.

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