MINERALOGY AND MICROMORPHOLOGY =

Clay Mineralogy of the Wadi M'Goun Sediments, Their Source, and Distribution on the Southern Flank of the Central High Atlas Mountains (Morocco)

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Abstract—Clay minerals are regarded as the most critical chemical and weathering components in ground. To establish the origin and distribution of the clay mineral associations of the Wadi M'Goun watershed, we have used mineralogical analyses on a set of fifty samples from channels sediments from the M'Goun watershed draining geologically diverse southern flank of the High Atlas mountain range. The analysis included X-ray diffraction for oriented clays and randomly oriented powders. The results indicate that the clay mineral assemblages primarily comprised of chlorite, illite, vermiculite, kaolinite, palygorskite, smectite, and sometimes interstratified clay minerals. In our study area, kaolinite was certainly of detrital origin since it was relatively abundant in quartz and feldspar-rich alluvium. Palygorskite was linked to the erosion of carbonates or detrital formations only with evaporitic layers. Chlorite was abundant in the fluvial clay cortege and reflected a slight alteration of the sediments. Smectite resulted from inheritance and its abundance was controlled by water erosion. Illite presented in high amount along the rivers of the M'Goun watershed, indicates a heritage that suggested a detrital origin. The quantitative and qualitative assessments of the clay mineralogy conducted in mountain areas allowed us to establish their source and distribution, and to relate the mineralogy of clays to various factors influencing the mineralogical and geological transformation. These findings have important implications for our understanding of paleodepositional environments in a semi-arid to an arid climate.

Keywords: clay minerals, XRD analysis, paleoenvironment, provenance, semi-arid climate **DOI:** 10.1134/S1064229322080063

INTRODUCTION

The mineralogical studies of clastic sediments have been systematically undertaken to clarify the aspects of provenance, weathering, erosion, tectonic conditions, and fluvial processes, for paleoenvironmental and paleoclimatic evolutions [3, 5, 8, 9, 18, 23, 27, 30, 35]. Clastic sediments are primarily clay minerals and quartz particles, with minor quantities of feldspar, mica, and heavy minerals [11]. Clay minerals are considered to be the most important chemical and dominating weathering products in soils [8, 35]. Sedimentary clays are the main constituents of finegrained sediments which can be transported far away from their source locations over long distances, according to their sources, are divided into two major categories; authigenic clays formed by in situ precipitation from a concentrated solution in closed continental or marine sedimentary basins, and detrital clays formed by weathering processes on the continent [10, 42]. Among them, authigenic minerals can indicate the climatic and environmental characteristics of the sedimentary area. Detrital minerals can often reflect the climatic and environmental characteristics of the parent area through long-term and long-distance transportation. Clay mineral compositions are used as a clear indicator of their source region [14, 26, 33, 50], as well as providing an effective tool for tracing the path of the sediments from source to sink, monitoring their transport routes, and defining their provenance [15]. Clay minerals contained in a given soil are characterized by the nature of the parent material, climatic conditions, topography, vegetation, and the time during which these factors operated [15]. Several studies have shown that clay minerals influence the properties that control aggregation, including surface area, charge density, cation exchange capacity (CEC), dispersibity, and expandability [1, 9, 31, 36, 45, 46]. Variabilities in clay mineral compositions are thus useful for assessing the provenance and origin materials of the sediments [19, 20, 26, 36]. The mineralogy of clays is a good marker for identifying the origin of fine-grained terrigenous sediments [10]. Aggregation is primarily high in smectite and vermiculite clays, which have a high CEC, vast surface areas, and abundant soil organic carbon [25, 34, 36, 39, 41, 43, 51]. Soil compositions in terms of minerals have a strong effect on soil erodibility in arid to semi-arid climates [13, 17]. Wakindiki [49] found in their study of soil mineralogy and texture effects on crust micromorphology, erosion, and infiltration, that the soils containing smectite were more vulnerable to water erosion than those containing kaolinite. Ben-Hur and Wakindiki [3] highlighted the importance of the soil mineralogical composition on erosion modeling. The weathering regimes that regulate the dominance and intensity of pedogenetic processes in continental source regions are represented by the composition of clay minerals [10]. Weathering mainly depends on environment zonation that established the degree of chemical and/or physical weathering [10]. The analysis of clay minerals during soil formation in weathering environments is one of the most interesting tasks carried out by clay mineralogists [8, 12, 48]. Much research has shown that several soil factors like the quantity of organic matter in the soil, soil structure and texture, topography, chemical environment containing oxides, pH, and erosional processes all covary with clay mineralogy [7, 39, 43, 44]. Since the late 1960s, X-ray diffraction techniques have been used to investigate the function of clay minerals as paleoclimatic and paleoceanographic indicators all over the world [10, 32, 33].

In this study, mineralogical analyses were carried out using the X-ray diffraction technique on bulk sediments and oriented clays to understand the quantitative and qualitative evaluation of clay mineralogy. The sediment clay mineralogy of the Wadi M'Goun watershed has not been well studied and therefore requires further attention to acquire a better understanding of the mineralogical and geological evolution in a semiarid zone on the southern side of Central High Atlas Mountains. The study of the clay mineralogy in the M'Goun watershed enabled us to identify different the clay mineral species of fluvial sediments formed in the study area and to determine their source and distribution. Indeed, it has allowed us to associate the mineralogy of the clays to various factors influencing the mineralogical and geological evolution in a semi-arid to arid mountainous regions.

MATERIALS AND METHODS

Study Area. The study area is located on the southern flank of the Central High Atlas Mountains of Morocco, belonging to the upper Draa Basin upstream of the El Mansour Eddahbi dam. It is located a hundred kilometers northeast of Ouarzazate and borders the Kelaat M'Gouna municipality and the valley of Boumalne Dades. The area lies between 31°20′ and 31°40′ N latitude and 6° and 6°30′ W longitude and covers an area of approximately 1260.68 km² within

which 46290 ha are collective rangelands. The study area's elevation ranges from 1400 to 4050 m, and the catchment is dominated by north- and south-facing slopes. The Jebel M'Goun is the highest peak in the region (4071 m) [21]. The M'Goun watershed is submitted to a semi-arid to arid climate with an annual rainfall average of about 130 mm per year, which is mostly concentrated in the autumn and winter months with irregular precipitation the rest of the year. The average annual temperature is about 20°C, the temperatures are warm in summer (the absolute maximum temperature is 40°C) and cold in winter with a minimum temperature between -1 and $-7^{\circ}C$, and wind speeds can attain 25 m/s. There is an average of more than 140 days of frost per year. Snowfall occurrences are sporadic during the season, and the first modeling findings and field tests have suggested a high percentage of snow lost directly by sublimation in the higher zones [40].

Regarding the geological background, the region of interest is situated on the southern flank of the Central High Atlas Mountains that comprise diverse lithological facies which range in age from the Triassic to the Quaternary (geological map of Jbel Saghro-Dades 1/200000) (Fig. 1). The drained area consists of dolomitic limestones rocks, marly limestones, and calcareous marl of Liassic age largely dominate in the upstream of the watershed. They represent about 40%of the area. The area also hosts basalts in the western part, Jurassic continental sandstone, Cretaceous marl-limestone outcrops with gypsum, calcareousmarly layers and conglomerates of Tertiary age, and Quaternary continental deposits and alluvial terraces towards the outlet of the watershed [22]. The M'Goun catchment area is geomorphologically composed of rock faces, debris flow channels, debris fans, vast scree slopes, ancient and active landslides, and rugged gravelly riverbeds.

Methodology. To determine the clay mineralogy of the Wadi M'Goun watershed, about 50 samples of river sediments were taken along the watershed's hydrographic network. The procedure of sediment preparation determined by diffractometry will be described in this section: randomly oriented powders or oriented clays were analyzed.

The Composition of minerals was determined by X-ray diffraction using two instruments: a Rigaku monochromatized diffractometer powered at 40 kV and 50 mA, characterized by a Cu K_{α} radiation source ($\lambda K_{\alpha} = 1.54$ Å) (Center of Analysis and Characterization, Cadi Ayyad University), and a Panalytical X'Pert PRO MPD diffractometer powered at 30 kV and 20 mA, equipped with a Co tube ($\lambda K_{\alpha} = 1.79$ Å), Fe filter, and a 1-D position-sensitive detector at the Bragg–Brentano parafocusing θ – θ reflection geometry (Department of Geological Sciences, Masaryk University).



Fig. 1. Geological map of the studied area and samples localization in the M'Goun watershed, upstream north part (UNP), upstream west part (UWP), middle area 1 (MA1), middle area 2 (MA2), and downstream area (DA).

X-Ray Diffraction of Randomly Oriented Powders

For X-ray diffraction sample preparation, both qualitative and quantitative, the homogenized samples were powdered with a <200 mesh size (0.075 mm) using an agate pulverizer prior to being measured. The

powders X-ray diffraction data were measured with a step size of $0.033^{\circ} 2\theta$, time per step of 130.175 s (for a single detector active channel), angular range of 6° – $100^{\circ} 2\theta$, and a total scan duration of 49 min and 10 s. The "Qualx" program was used to determine the crys-



Fig. 2. XRD patterns of randomly oriented powders, (a): sample from the upstream north part (UNP), (b) sample from the upstream west part (UWP), (c) sample from the middle area (MA), (d) sample from the downstream area. Cal: Calcite, Dol: Dolomite, Fsp: Feldspar, Gp: Gypsum, Px: Pyroxene, Qz: Quartz.

talline phases in the X-ray diffraction patterns. To detect the better-fit mineral phases for the identified peaks, the XRD patterns were characterized through comparisons with "POW_COD" X-ray diffraction patterns which were derived an SQLite database available free of charge, and their crystallographic information was generated from the library of Crystallography Open Database (COD), which contains 454860 references (Fig. 2). The mineral abundances were determined using the "REX" program after the crystalline phases were described.

X-Ray Diffraction for Oriented Clay

The clay fraction samples were prepared using the techniques defined by Moore and Reynolds [32]. In this procedure, no chemical treatment was applied to the samples studied, since previous researchers had shown that chemical treatments such as those aiming to remove organic matter or iron and aluminum hydrox-

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ides using H_2O_2 or DCB could alter clay minerals and especially mixed-layer clays [32, 47].

The samples of clay fractions ($<2 \mu m$) were separated by settling in a water column and saturated with CaCl₂. Clay fractions were deposited on smooth glass slides as oriented aggregates. For each oriented preparation, three X-ray patterns were recorded: untreated, ethylene glycol solvated, and heated at 450°C for 3 h. Measurements were made in the $3^{\circ}-30^{\circ}$ 2 θ angular range with a scanning step size of $0.03^{\circ} 2\theta$ and a counting time of 1 s per step [20]. After the X-ray pattern's background noise was eliminated, the line location, intensity peak, and integrated area were measured. The quantitative phase analysis of the main clay species was carried out with the Rietveld method using the fundamental parameter approach [6]. The illite crystallinity index or Kübler index was measured on the peaks at 10 Å of the glycol test by choosing the width of half of the peak height [24]. The illite chem-



Fig. 3. Evolution of mineral group abundances in fluvial sediment deposits in the Wadi MGoun watershed normalized to 100%, which excludes minor and trace mineral abundances, (a) in the upstream north part, (b) in the upstream west part, (c) in the downstream area.

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Fig. 4. Scarlet plot of quartz and feldspar vs. calcite and dolomite showing the effect of sorting of detritic and carbonate materials on the Wadi M'Goun sediments.

istry index was calculated with the area ratio of the 5 Å peak and the 10 Å peak under ethylene glycol-solution conditions. It was used to evaluate the degree of chemical weathering.

RESULTS

The XRD analyses obtained from randomly oriented powders samples of the Wadi M'Goun watershed in riverbed sediments are shown in Figs. 2a-2d. We selected XRD diagrams based on samples from five major zones in the study area; samples from the upstream north part (UNP), samples from the upstream west part (UWP), samples from middle area 1 (MA1), samples from middle area 2 (MA2), and samples from the downstream area (DA). The mineralogical data of the upstream north part river sediments (UNP) revealed the predominance of calcite particles. mostly dolomitic which was the dominant phase, as well as quartz and feldspars which were the minor phases in this zone of the watershed area (Fig. 3a). Lithologically, the parent material in the north of the Wadi M'Goun watershed mainly consisted of limestone, and most of the eroded materials were highly calcareous throughout. The UWP data showed the dominance of the quartz fraction, while other minerals such as feldspar, calcite, dolomite, and gypsum were less abundant (Fig. 3b). Other compounds (pyroxene, biotite, muscovite et al.) were present in amounts that ranged from trace to a few percent. In the DA, the mineralogical results revealed the presence of all the minerals; quartz, feldspar, calcite, dolomite, pyroxene, biotite, muscovite; all these phases were observed in the alluvial cortege, and their proportions varied within the coarser channel (Fig. 3c).

Figure 4 illustrates the correlation of quartz-feldspar with the presence of calcite-dolomite; the abundance of quartz-feldspar was inversely proportional to the calcite-dolomite content. The continental provenances were distinguished mainly by the abundance of quartz and feldspar, whereas calcite abundances may have reflected the alteration of marine to submarine carbonates. When the channel networks incised the geological terrains of the Liassic carbonates, the alluviums showed a high percentage of calcite and dolomite. On the other hand, if water erosion affected Ordovician, Triassic or Jurassic detrital terrains, it was instead quartz and feldspars that formed the majority of the alluvial sediments. The alteration of late Triassic basalts provided relatively fewer amounts of micas and ferromagnesian materials compared to the other phases. The X-ray diffraction patterns on oriented clay, normal, treated with ethylene glycol and heated to 450°C, were deciphered and interpreted according to the method of Moore and Reynolds [32], giving relevant results. For each sample of fluvial sediments, three spectra facilitated the detection of peaks corresponding to the clay minerals (Fig. 5). From upstream to downstream, The X-ray diffraction analysis revealed the presence of the following clay minerals in all of the samples examined: illite, chlorite, vermiculite, kaolin-



Fig. 5. X-ray diffraction (XRD) patterns of the clay fraction in the Wadi M'Goun watershed zones, the upstream west part (UWP), middle area 1 (MA1), middle area 2 (MA2), and the downstream area (DA). Oriented mount: untreated (black), ethylene–glycol-solvated (green), and heat-treated at 450°C (red). Sme: Smectite, Chl: Chlorite, Vrm: Vermiculite, Plg: Palygorskite, Ilt: Illite, Kln: Kaolinite, Ilt-Chl: Interstratified Illite-Chlorite, Qz: Quartz.

ite, palygorskite, smectite, and sometimes interstratified clays minerals. The 10.1 Å label showed the position of illite (001). The 14.0 and 3.3 Å peaks that were unchanged by any treatments identified chlorite and quartz, respectively. Vermiculite identification can be difficult because its properties vary, there exist dioctahedral and trioctahedral vermiculites of largely unknown composition. According to Velde [48], clay mineral with a basal spacing slightly below 15 Å which shows an increase towards 15.4 Å during glycollation is considered to be a vermiculite. The 7.0 and 3.58 Å peaks indicate the presence of kaolinite, the kaolinite becomes amorphous only at 550-600°C, under 550°C it remains in its crystalline state. The 10.4 Å label showed the position of palygorskite (110). The expansion of the 14.1 peak maximum to 17.0 after ethylene glycol (EG) treatment indicated the existence of smectite. The interstratified illite-chlorite minerals were characterized by an intensive reflection between 5.0 and 4.7 Å according to Reynolds [38], after heat treatment, the (001) basal diffraction pattern was decomposed into three basic curves with their maximal at 10.8, 10.19, and 9.93 Å. The increasing proportion of chlorite components from one sample to another could affect the position of the peak from 5.01 to 4.73 Å. regarding the first-order reflection between $6^{\circ}-12^{\circ}$ 20 region, the intensity of the peak near 5.0 Å is greatly reduced when the hydroxide sheet of the chlorite component is not complete.

The distribution of clay minerals among the Wadi M'Goun watershed zones varied considerably, both



Fig. 6. Distribution of clay minerals in the Wadi M'Goun watershed: (1) Vermiculite; (2) Chlorite; (3) Illite; (4) Kaolinite; (5) Palygorskite; (6) Smectite.

qualitatively and quantitatively. The quantitative analyses of clay minerals in the studied samples are shown in Fig. 6. Chlorite and illite were predominant in all the samples in relatively high proportions. However, their percentage decreased with an increasing presence of carbonates and increased with an abundance of quartz and feldspar (Fig. 7). The same evolution was observed with kaolinite and palygorskite, but with varying amounts from 0 to 15% of the clay cortege for each clay. Alternatively, the amount of smectite increased with the abundance of carbonated alluviums and decreased with those rich in quartz and feldspar. The amount of smectite varied between 0 to 40% in the study area. Vermiculite remained without preferences with rates varying from 0 to 14%. Interstratified clays were less abundant, and they were localized in specific zones. Their constitutions were not diversified, and

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they were mainly composed of sheets of illite-chlorite or illite-smectite.

DISCUSSION

Smectite is a swelling clay that forms in basic, poorly drained soils in a hot or low temperature. It primarily originates from a source where framework silicates, it is the main constituent of Fe-Mg aluminosilicate minerals [28]. Smectite is present in the majority of the samples studied and represents up to 40% in the study area. It was mainly detected in the clay fraction of the alluvial sediment in the middle part and the downstream area of the Wadi M'Goun watershed. The occurrence of smectite suggests several origins, it may originate from volcanic activity, during pedogenesis, or inherited [4]. In the middle part of the watershed, the presence of this clay min-



Fig. 7. Scatter plots of illite and chlorite with the abundances of carbonate and quartz-feldspar alluviums.

eral may indicate a volcanic origin reliant on the Triassic basalt. In the second case, it was especially abundant near carbonate or evaporitic soils (Fig. 8), in which it may have an inherited character. The abundance of smectite is controlled by water erosion, for the reason that soils containing smectite are more susceptible to water erosion in comparison to soils containing illite and kaolinite [49].

Kaolinite can be created by weathering or the hydrothermal alteration of aluminosilicate minerals [48]. It is commonly formed under strongly hydrolyzed soils in warm, humid, or temperate climates with acidic pH and an actively leached topography. In our study area, the detrital origin of kaolinite is clear since it is relatively abundant in quartz and feldspar-rich alluvium.

Palygorskite is a clay found in soils in arid environments or shallow marine environments, commonly attributed to authigenic processes by direct precipitation from solutions with high Mg and Si concentrations and low Al-activity under alkaline pH conditions. In the M'Goun basin, it was present in small quantities and linked to the erosion of carbonate or detrital formations only with evaporitic layers.

Chlorite is considered to be a clay species derived from the physical weathering of crystalline rocks since it is often inherited from other preexisting rocks of different natures [4]. Chlorite occurred in rather small quantities in the soil because it is sensitive to weathering by transforming to vermiculites or smectites [44]. In the M'Goun catchment area, the proportion of chlorite was high in the fluvial clay cortege, which may reflect a slight alteration of the sediments.

Illite can be created by the alteration of minerals such as muscovite and feldspar [4, 48]. Illite is usually inherited from parent rocks and does not form during pedogenesis. The relatively high amount of illite along the rivers of the M'Goun watershed indicated a heritage that suggested a detrital origin. Furthermore, the presence of high percentage of illite together with the abundance of quartz, and feldspar indicates a substantial detrital input in arid and semi-aridclimates [16, 29]. However, diffractometry showed that the peaks of the illite were not always broad, which means that the indices of the latter didn't necessarily reflect the degradation of the illite crystal networks. The chemical index of illite is considered as a good indicator of weathering intensity in the provenance zone and it represents the chemical composition of illitic minerals [38]. The illite's high chemical index value indicates intense hydrolysis, while a low value indicates intense physical erosion [37]. Indeed, the chemistry index of illite for the different samples in the study area was, generally, between 0.12 and 0.40; this could translate into the absence of a hydrolysis process and intense physical erosion. The crystallinity index was between 0.1 and 0.6 with two-thirds of the values being lower than 0.4. These two indices were inversely proportional: the more the crystallinity index increased, the more the



Fig. 8. Triangular diagram of the quantitative clay composition showing the distribution of the smectite, kaolinite, and illite-chlorite sums in the Wadi M'Goun watershed (UNP: samples from the upstream north part, UWP: samples from the western part, MA1: samples from middle area 1, MA2: samples from middle area 2, DA: samples from the downstream area).



Fig. 9. Correlations of illite chemistry index with the illite crystallinity of surface sediments in the main stream and tributaries of the Wadi M'Goun watershed which indicate a linear correlation between illite chemistry index and illite crystallinity.

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Fig. 10. Evolution of illite crystallinity in the Wadi M'Goun watershed. IC: Illite crystallinity.

chemical index decreased, and vice versa (Fig. 9). The low values of the crystallinity index indicated deep diagenesis or even low metamorphism.

The most reasonable interpretation probably involved pedogenesis in specific areas where both the conditions and the geology of the terrain were favorable. In fact, for the first point, the NDVI map showed little vegetation cover. The flora consisted essentially of scattered trees. The climate was not very clement. In general, soils were not very evolved and very poor except for the alluvial valleys, where pedogenesis was relatively active by forming silty soils. Nevertheless, even these soils were poorly developed in space; diagenetic transformations could be observed and the mineralogical evolution of stable alluvial sediments may have affected the clay's mineralogy. For the geological factor, low values of the illite crystallinity index were observed, especially in the upstream western part and the median area 2 (Fig. 10). In the upstream western part, the bedrock consisted of Ordovician shales, late Triassic basalts, and Triassic siltstones. The latter can yield strongly crystallized illites [2]. Samples from the median area that showed narrow peaks of illite were all taken from a fault zone. In the M'Goun catchment area, reverse faults showing Atlasic compression are still active. Thus, tectonic can affect bedrock deformation and mineral transformations.

CONCLUSION

The quantitative and qualitative study of alluviums in the Wadi M'Goun watershed has made it possible to highlight the origins, the distribution modes, and the fine fraction evolution of sediments. According to the findings, illite, chlorite, vermiculite, kaolinite, palygorskite, and smectite were found in clay mineral assemblages of fluvial sediments in the regional catchment area, including the main channels and various branches. The recent elements were present as major clay minerals, and sometimes a smaller proportion of interstratified clay minerals were observed. The proportion of smectite is controlled by water erosion, for the reason that soils containing smectite are more susceptible to water erosion. The kaolinite content increases gradually from the upstream area towards the downstream area. It is relatively abundant in quartz and feldspar-rich alluvium that signify a detrital origin. The qualitative homogeneity of the clay processions reflects more or less the continuity in the sources of supply from the upstream to the downstream zones. However, this homogeneity was different in the tributaries, which could be explained by differences in the heterogeneous source rocks, weathering intensity, and fault zones. The tectonic, bedrock and weathering conditions in the Central High Atlas Mountains influenced the abundance of illite and chlorite in fluvial sediment samples from the study region. Besides, the mountainous landscapes spread into the region of the Wadi M'Goun watershed could have fostered stronger physical erosion and resulted in the highest illite contents. In addition, the relatively high amount of illite and chlorite along the rivers implies semi-arid to arid climatic conditions in the source regions.

From the upstream to the downstream areas, an intense physical weathering was present. The lithology of the parent rocks, tectonic activity, and the semi-arid climatic conditions of the region importantly affect the clay mineral assemblages of the Wadi M'Goun watershed draining the Central High Atlas Mountains.

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CONFLICTS OF INTEREST

The authors declare no conflict of interest.

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