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Clay mineralogy and paleoclimatic properties of the Neogene Deposits in Sinanpaşa Basin (Afyon-Western Anatolia)

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ABSTRACT

The Sinanpaşa basin, one of the Neogene basins formed in the extensional tectonic regime in Western Anatolia, is located on the eastern margin of Western Anatolia basins containing different types of evaporites, lignite, uranium and clay minerals. Miocene sequence was initiated with alluvial fan-fluvial units and followed by lacustrine sediments. River floodplains and lakeshore marshes locally contain economic coal levels. Since middle-late Miocene, the products of Afyon volcanism were deposited into a subaerial or a subaqueous environment alternating with lake sediments. Dominant clay minerals are illite and smectite within the sediments coexistence with the volcanics. These minerals were accompanied by chlorite, kaolinite, and zeolite in some samples. Feldspar, dolomite, thenardite, and gypsum are locally observed too. Sepiolite mineral enrichments are also noteworthy in shallow-coastal lacustrine environments where organic matter is abundant. Clay minerals are mostly detritic and developed due to the weathering of volcaniclastics and sedimentary rocks in temperate/humid climatic conditions. In particular, smectite and kaolinite minerals were formed in situ/authigenic under subaqueous conditions due to the acidic/basic character of tuffs. As a result, different clay mineral types have been developed due to the mineralogical composition of source rocks, climatic factors (humid/semi-humid, arid), paleotopography, depositional environment and diagenetic factors.

1. Introduction

Extruding of Anatolian block to the west caused E-W directional compression, and then N-S directional extension in Western Anatolia (Şengör and Yılmaz, 1981; Koçyiğit, 1984; Robertson and Dixon, 1984; Şengör et al., 1985; Savaşçın and Güleç, 1990; Zanchi et al., 1993; Seyitoğlu et al., 1997; Yılmaz et al., 2000; Koçyiğit and Deveci, 2007; Ersoy et al., 2011; Prelevic et al., 2012). Due to the extensional tectonic regime, NW-SE, NE-SW and E-W directional Neogene graben basins have been formed in Western Anatolia (Savaşçın et al., 1994; Bozkurt, 2003). Most of these Neogene graben basins are associated with significant economic deposits. Among these, lignite (eg Çanakkale-Çan, Muğla-Yatağan, Manisa-Soma, Kütahya-Tunçbilek, Seyitömer), evaporite (eg Balıkesir-Bigadiç, Sultançayırı, Bursa-Kestelek, Kütahya-Emet, Eskişehir-Kırka) and uranium (Manisa-Köprübaşı, Uşak-Fakılı, Aydın-Söke) deposits are the main ones. Miocene stratigraphy in all these basins was generally initiated with alluvial fan and fluvial deposits, and followed upward with

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Neogene lacustrine sediments (eg; Kaya, 1981; Helvacı and Alaca, 1991; Helvacı and Yağmurlu, 1995; Akal, 2003, 2008; Ersoy et al., 2008; Karaoğlu et al., 2010; Helvacı et al., 2017). In addition, these basins have been exposed to a strong volcanism under extensional tectonic conditions since late Cenozoic (Borsi et al., 1972; Keller and Villari, 1972; Ercan et al., 1978; Ercan, 1979; Başarır and Kun, 1982; Çevikbaş et al., 1988; Yılmaz, 1989, 1990; Savaşçın, 1990; Savaşçın and Güleç, 1990; Aydar and Bayhan, 1995; Floyd et al., 1998; Savaşçın and Oyman, 1998; Francalanci et al., 2000; Aldanmaz et al., 2000; Yılmaz et al., 2001; Akal, 2003, 2008; Çoban and Flower, 2007). The products of the volcanism were deposited in the Neogene lakes and occasionally alternated with lake deposits (Becker-Platen et al., 1977; Besang et al., 1977; Aydar and Bayhan, 1995; Aydar, 1998; Aydar et al., 2003; Akal, 2003, 2008; Prelevic et al., 2012; Akal et al., 2013, Prelevic et al., 2015). The Sinanpaşa (Afyon) basin, which is the subject of this article, is located on the eastern margin of the Western Anatolian graben system, and it is a typical example of the deposition of volcanic and sedimentary sequences in Western Anatolia.

Sinanpaşa basin is adjacent to Western Anatolian basins that contain economic borate (Balıkesir-Bigadiç, Bursa-Kestelek, Kütahya-Emet, Eskişehir-Kırka), lignite (Çanakkale-Çan, Manisa-Soma, Kütahya-Seyitömer and Tunçbilek) and also uranium (Aydın-Söke, Manisa-Köprübaşı, Uşak-Fakılı) deposits. For this reason, the geological and economic importance of the Sinanpaşa basin increases. When the basin is evaluated in terms of industrial raw materials and clay-feldspar deposits, these occurrences are operated extensively in the Akharım region (southern part of the study area, Kuşçu and Yıldız, 2012; figure 1) and its surroundings (eg. İscehisar-Anayurt, Sandıklı-Kınık). Even though, the tectonic and magmatic evolution of the Sinanpaşa basin have been investigated so far, studies regarding the sedimentology and mineralogy of the Neogene lacustrine sediments have less studied in detail. There are only a few studies about Akharım clay deposits around Sandıklı, in the southern part of the

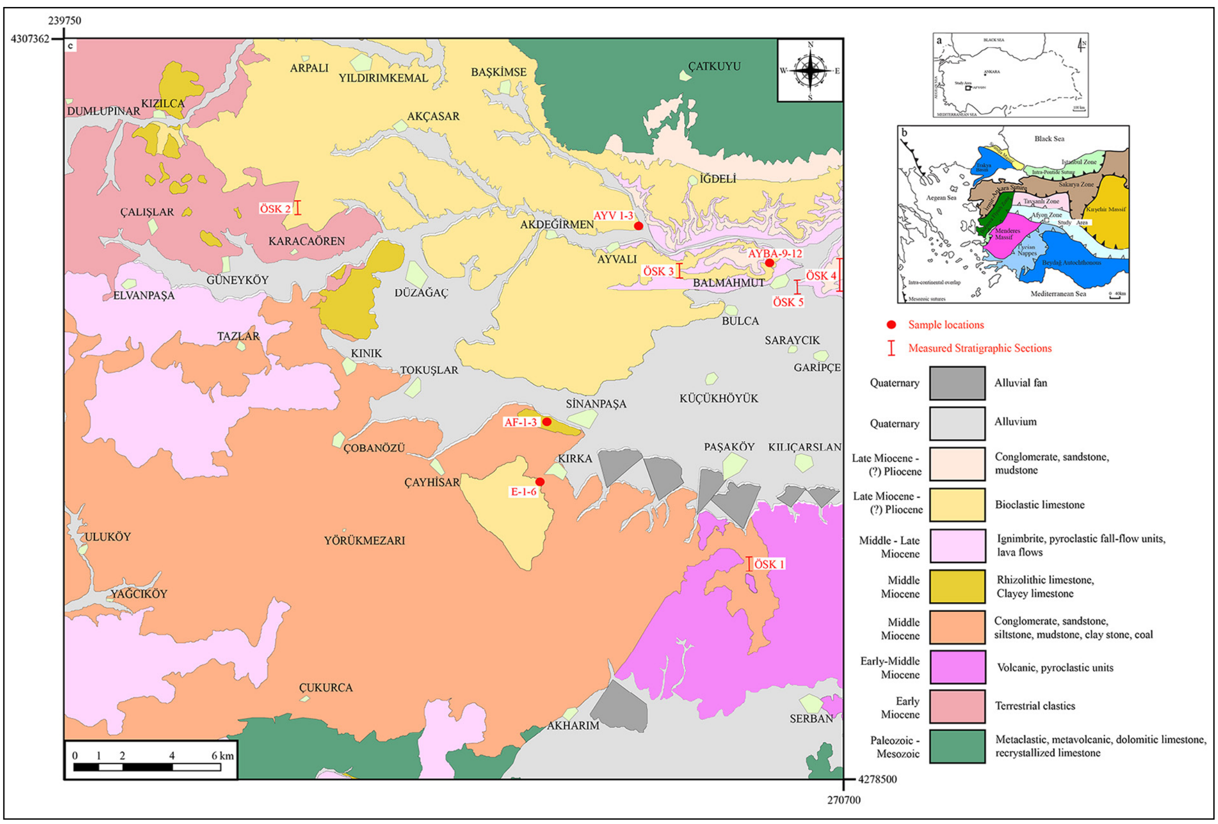

Figure 1- a) Location map of Sinanpaşa (Afyon) basin, b) generalized tectonic map of Western Anatolia and tectonic zones around Afyon Zone (simplified from Okay et al., 1996), c) geological map of Sinanpaşa basin (from Akıska and Varol (2020), who modified from Metin et al., 1987).

Sinanpaşa basin (Kuşçu and Yıldız, 2012). Therefore, this paper aims to investigate the mineralogical and petrographical properties of Neogene-aged lacustrine and volcanic/pyroclastic units accompanying the lacustrine sediments around Sinanpaşa village. In addition, it is aimed to determine the clay minerals and other accompanying mineral types deposited in different facies and also to determine their depositional environments, paleoclimate conditions and diagenesis properties.

2. Regional Geology

With the closure of the Neotethys Ocean, Anatolide-Tauride block was situated along the south of Izmir-Ankara suture zone. It was divided into four tectonic zones from north to south, such as Tavşanlı Zone, Afyon Zone, Menderes Massif and Lycian Nappes (Şengör and Yılmaz, 1981; Okay, 1984; Göncüoğlu et al., 1996; Okay and Tüysüz, 1999; figure 1a, b). Among them, Afyon Zone rocks consist of Mesozoic units that unconformably overlie a Paleozoic basement. The studied Sinanpaşa Neogene basin rests unconformably on the Paleozoic and Mesozoic basement rocks around the central part of the Afyon zone (Figure 1c). The rocks, exposed in Afyon and its surroundings, were studied in three different groups. These are; (a) Pre-Neogene basement rocks (Afyon Zone), (b) Neogene sedimentary and volcanic rocks, and (c) Quaternary fluvial/alluvial deposits (Figure 2).

(a) Basement rocks: Afyon metamorphics are the predominant basement rocks of Afyon Zone. They (possibly during Cambrian and late Silurian) consist of mica schist, garnet mica schist, albite-chlorite and quartz schist, meta conglomerate, phyllite and marbles (Metin et al., 1987; figure 2). These rocks were unconformably overlain by the Middle-Late Devonian-late Permian Anatolian carbonate platform (Güvenç et al., 1994; Tolluoğlu et al., 1997). The carbonate platform consists of recrystallized limestones which have undergone low-grade metamorphism and contains clayey and sandy layers in places. According to the paleontological data, the age of the limestones was determined as Middle-Late Devonian, Carboniferous and Permian (Öztürk, 1981; Metin et al., 1987; Bektaş, 1996). Polygenic conglomerates consisting of metamorphic rock fragments overlie the Paleozoic rocks and were unconformably overlain by white, fossiliferous limestones of Triassic-Jurassic period (Güvenç et al., 1994; Bektaş, 1996; figure 2).

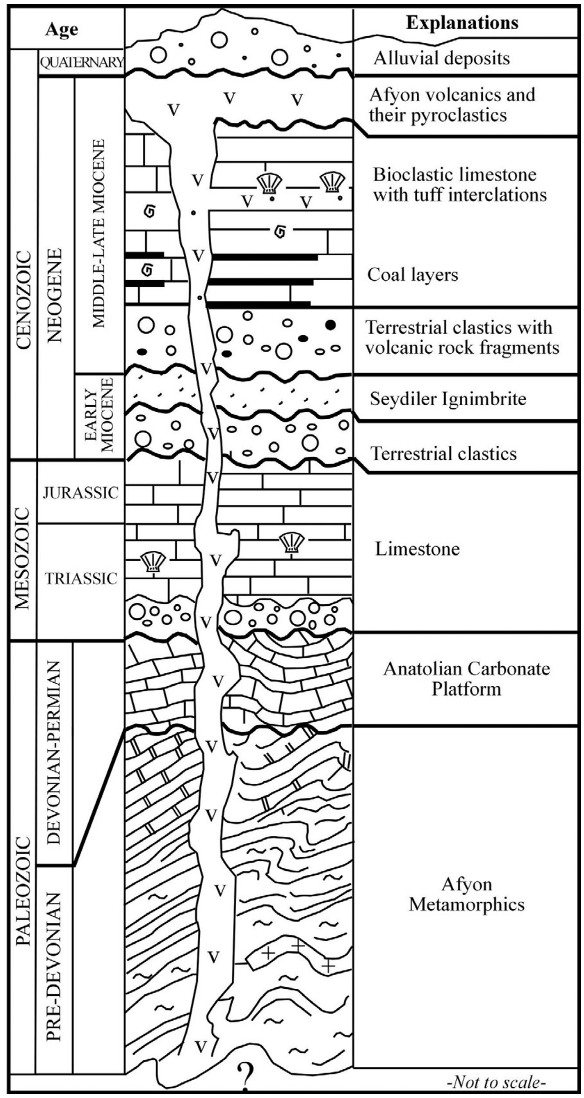

Figure 2- Generalized stratigraphic column of Sinanpaşa basin (simplified from Öztürk, 1981; Metin et al., 1987; Okay et al., 1996; Tolluoğlu et al., 1997).

(b) Neogene rocks: The basement rocks were unconformably overlain by Neogene rocks (Figure 1c). These rocks consist of a sedimentary sequence deposited in fluvial and lacustrine environments with locally economic coal layers and volcanic/pyroclastic products of Afyon volcanism (Becker-Platen et al., 1977; Besang et al., 1977; Çevikbaş et al., 1988; Metin et al., 1987; Aydar and Bayhan, 1995; Aydar et al., 2003; Akal, 2008; Akal et al., 2013; Prelevic et al., 2015). In the northern part of the basin, Neogene deposits were initiated with a polygenic conglomerate derived from the basement rocks and consist of sandstone, mudstone and claystone in the vicinity of Çalışlar-Karacaören villages. In the southern part of

the basin, around Kırka, the sequence was followed upward by a second conglomerate sequence with sandstone and mudstone deposition. This sequence contains pre-Neogene basement rock fragments and volcanic content due to its contact to early Miocene Seydiler ignimbrites (Yalçın, 1988). It was overlain by lacustrine sediments of middle Miocene. These sediments were characterized by coal-bearing mudstones situated along the Afyon-Sandıklı main road which is approximately 20 km southwest of Kırka village and by carbonates intercalated with coal levels in Kızılca village in the north and in Kırka village in the south. Meanwhile, Afyon volcanism was active during the middle-late Miocene period in the study area (Besang et al., 1977; Becker-Platen et al., 1977; Aydar and Bayhan, 1995; Akal, 2008; Prelevic et al., 2015) and the products of volcanism were either interstartified with lacustrine deposits or overly them (Aydar and Bayhan, 1995).

Afyon volcanics, exposed in the study area and its surroundings, were accepted as the products of Neogene volcanism (Keller, 1983). According to the radiometric determinations made on the lava and pyroclastics of volcanism, it was determined that Afyon volcanism, consisting of trachyandesite and trachitic lavas and their pyroclastic products (Aydar and Bayhan, 1995; Floyd et al., 1998; Akal and Helvacı, 1999; Aydar et al., 2003; Akal, 2003, 2008; Akal et al., 2013; Prelevic et al., 2015) was active between middle-late Miocene (8.5-14.5 Ma) (Besang et al., 1977; Becker-Platen et al., 1977). In the study area, while volcanic rocks were exposed around Elvanpaşa and Tazlar villages, toward the west of the basin, on the other hand, pyroclastic products were exposed around Balmahmut-İğdeli villages at the north of the basin (Figure 1c). At the end of the late Miocene, volcanism ended (Becker-Platen et al., 1977; Besang et al., 1977; Aydar and Bayhan, 1995; Akal, 2008; Prelevic et al., 2015) and the fluvial regime prevailed.

(c) Quaternary units: All Neogene units are unconformably overlain by Quaternary fluvial-alluvial deposits.

3. Material and Method

Petrographic studies have been carried out to determine the paleontological, compositional, textural and diagenetic properties of Neogene limestones and conglomerates in the study area and to obtain detailed information about the depositional environments of them. Therefore, limestone and conglomerate samples were collected along the measured stratigraphic sections and also by both point to point method. Thin sections were prepared in Thin Section and Polishing Laboratory of Department of Geological Engineering in Ankara University and they were investigated in Leica DM/LSP model polarizing microscope. In addition, in cases where petrographic determinations are insufficient, micro-structure studies were carried out by the scanning electron microscope (SEM) to collect more detailed and healthy information especially about the morphology of the clay minerals. Energy Scattering Spectroscopy (EDX) analyses were performed for the chemical determinations. Analyses were made by Zeiss Evo MA 10 SEM model electron microscope and Bruker Nano XFLash 430M Energy Scattering X-ray (EDX) detector at Leoben University (Austria).

The mineralogical composition of the samples was examined with Philips PW 1830 model X-rays (XRD) diffractometer (Anode = CuKα 1.541871 Å, Filter = Ni, voltage = 40 kV, current = 30 mA, goniometer speed = 1 or 2°/min., paper speed= 2cm/min, 2θ interval= 2θ=2,5-70°) in the Mineralogy-Petrography Laboratories of the Mineral Research and Exploration General Directorate (MTA). Representative samples were prepared for clay-mineral analysis (size fraction <2 µm) by separation of the clay fraction by sedimentation. Each sample mounted on glass were subjected to air-drying (N), ethylene-glycolating (EG) and heating (300 °C and 550 °C) procedures before the XRD studies. Diffractograms were evaluated using the American Society for Testing Material catalog (ASTM, 1972). Semi-quantitative estimates of both clay fraction and rock-forming minerals were calculated by an external standard method (Brindley, 1980; Yalçın and Bozkaya, 2002).

4. Lithology

4.1. Sedimentary units of middle Miocene

The sedimentological features of the middle Miocene units in the study area were examined in detail by Akıska and Varol (2020). Therefore, in this study, only the lithological properties of the studied units and their depositional systems will be discussed. Middle

Miocene sequence was initiated with conglomerate-sandstone sediments and continues with coal-bearing mudstones and carbonates (Figure 3). *Conglomerate-sandstone* deposits were well exposed around Kırka village at the southern part of Sinanpaşa town. These deposits were characterized by yellow-orange colored, medium-well rounded, poorly sorted, granule to block-sized clasts and cross-bedded, medium-coarse-grained sandstones. Clasts derived mainly from the basement rock fragments of Afyon metamorphics and rhyolitic lavas of Seydiler ignimbrites (early Miocene, figure 4a). The thickness of the sequence was about 100 meters. Cross stratifications observed within the sandstones, which are intercalated with the yellow-orange conglomerates and fine-grained sand matrix indicate normal stream flow units (Smith, 1986).

Coal-bearing mudstones were well exposed around 20 km southeast of Kırka village (ÖSK 1, figure 4b) at the south of the basin and also around the coal mine around Karacaören village at the NW of the basin (ÖSK 2; figure 3b). Coal layers in both regions were alternated with gray colored mudstones and their total thickness is approximately up to 150-200 meters. The sequence was passed upward by sandstone-laminated mudstones at the southeast of the Kırka village (Figure 4c). Laminated mudstones containing a variety of fossilized plant roots are yellow-gray in color. The mudstones around Karacaören village were followed upward by the carbonate rocks of late Miocene (Figure 4d). According to the palynological analysis, the coals, in both regions, point to the late middle Miocene (Akıska, 2017). Characteristics such as limited propagation and rhythmic alternation reflect the features of typical marsh coals that developed during the first stages of the lake (Miall, 1977; McCabe, 1984).

Carbonate deposits of middle Miocene overlying the coal-bearing mudstones were represented by gray-

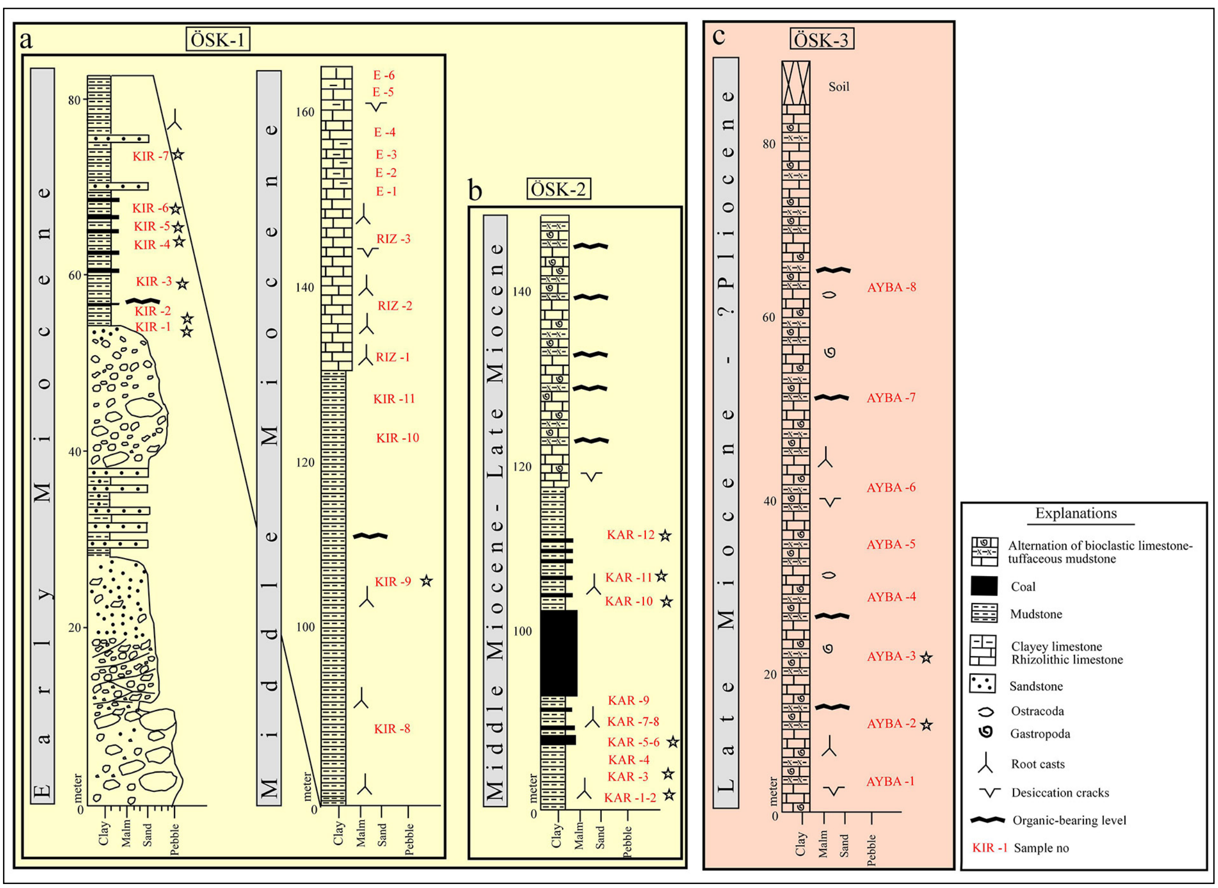

Figure 3- a) Measured stratigraphic section of middle Miocene sediments (ÖSK 1, Southeast o the Kırka village), b) measured stratigraphic section of middle Miocene sediments (ÖSK 2, Karacaören coal mine), c) measured stratigraphic section of late Miocene-?Pliocene deposits (ÖSK 3, Balmahmut village).

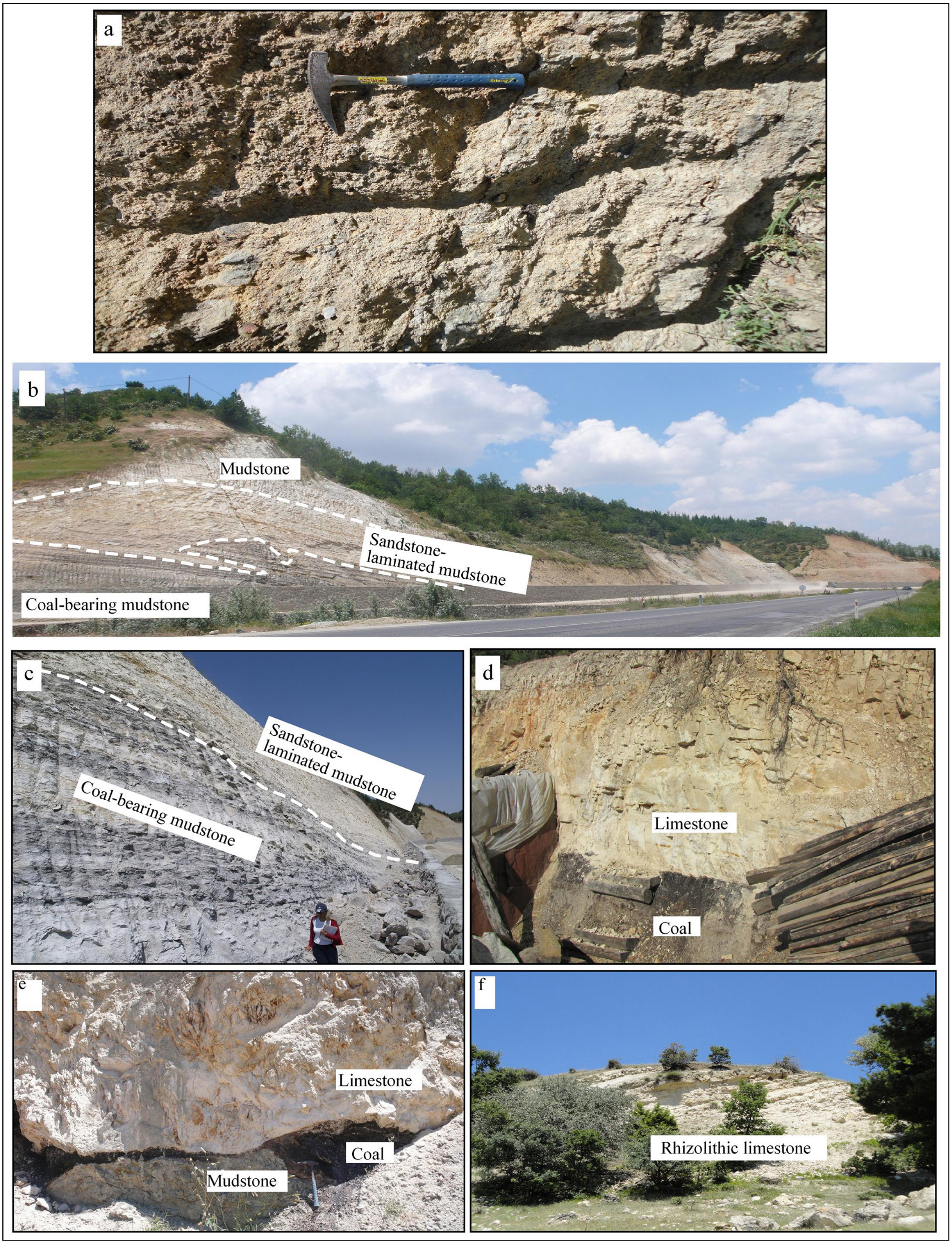

Figure 4- a) Volcanic gravels in poorly graded conglomerate, b) coal-bearing mudstone at the southeast of Kırka village, c) sandstone-laminated mudstone deposits overlying coal-bearing mudstones (Southeast of Kırka village), d) coal layers and overying late Miocene carbonate deposits (Karacaören coal mine), e) mudstone underlying the coal layers and limestone overlying the coal layers (Karacaören coal mine), f) rhizoliths (Kırka village).

beige rhizolithic limestones and white-gray clayey limestones (Figure 4e). They were exposed around Kızılca village at the north of the basin and Kırka village at the south of the basin (Figure 4f).

4.2. Deposits of Late Miocene – (?) Pliocene

This period of time was represented by bioclastic limestones and tuffaceous organic-rich mudstones (ÖSK 3; figure 3c). *Bioclastic limestones* have widespread outcrops in both the northern (around Balmahmut and İğdeli villages) and southern parts of the basin (around Kırka village). This unit which has tabular and lenticular geometry, are composed of beige colored, parallel-bedded carbonate layers ranging in thickness from several centimeters to several meters (Figures 5a and 5b). According to the petrographic studies, iron and manganese plasterings, microcrystalline calcite patches and gastrapod and ostracod macro shells with a rate of approximately 50% are abundant (Figure 5c). The limestone have nodular appearances in the lower levels and alternates upward with tuffite-bearing organic rich mudstone layers (Figure 5a and 5b). These tuffs were the products of Afyon volcanism that was active during the middle-late Miocene period in the study area having a coexistence occurrence with the limestones and exhibiting an alternating deposition character with these sediments. The *organic-rich mudstones* are represented by parallel laminated, poorly-moderate lithified gray-dark gray mudstones with plant root fragments and gastrapod shells. Lamina thickness is between 0.5-0.8 cm.

4.3. Pyroclastics of Middle-Late Miocene

Pyroclastics of Afyon volcanism well exposed in the vicinity of Balmahmut and İğdeli villages

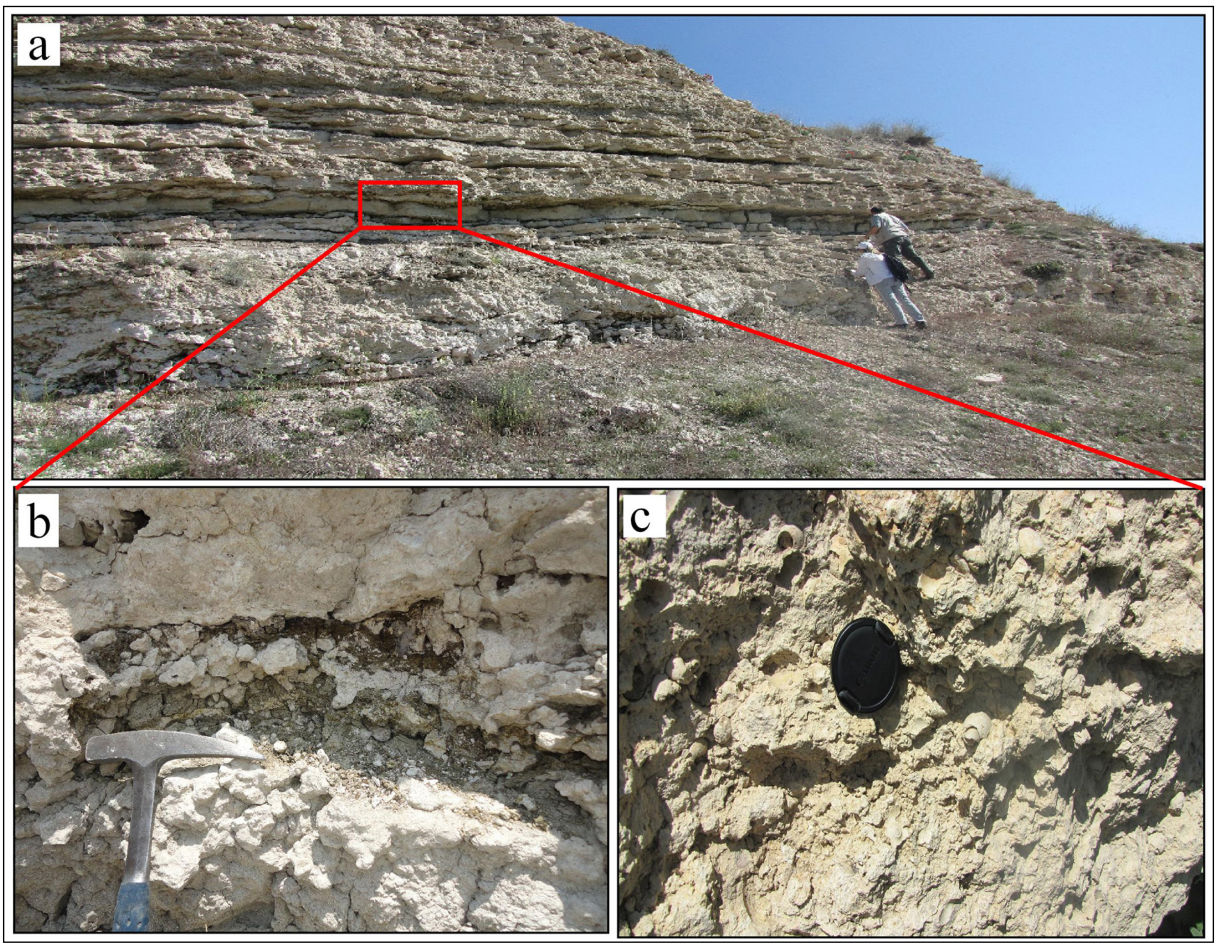

Figure 5- a) General view of alternation of tuffite bearing organic rich mudstone and bioclastic limestone, b) close up view of figure 5a center, c) close up view of macro gastrapod and ostracod shells observed in limestones (Balmahmut village).

were classified as *ignimbrites, pyroclastic flows* and *porphyric lava flows* (Figure 6a and 6b). *Ignimbrites* consist of poorly sorted, angular and semi-rounded lithic blocks mostly having trachitic composition and pumice within them with the clast sizes of lapilli to block (Figure 7a). Fairy chimney structures were formed in places due to erosion where ignimbrites are non welded.

Two measured stratigraphic sections were taken around the vicinity of Balmahmut village where porphyric lava flows were well exposed (ÖSK 4, figure 6a and 7b; ÖSK 5; figure 6b and 7e). The lava flows were underlain and overlaid by pyroclastic flows in places.

In this area, ÖSK 4 commenced with bioclastic limestones and passes upward into poorly sized and reverse graded, lava and pyroclastic flows varying from ash size to block size, and tuff layers of cream-beige color (Figures 7c and 7d). On the other hand, ÖSK 5 is initiated with calish layers (Figure 7e and 7f) and followed up by thin-medium bedded tuffs and mollusk-bearing levels. Tuff-dominated deposition packages are terminated by coarse-grained pyroclastic units (Figure 7g). Pyroclastic flow deposits have lateral and vertical extent of 50 meters in both measured stratigraphic sections.

5. Petrographic Properties of Volcanic and Pyroclastic Rocks

Volcanic rocks exposed in the investigation area around the villages of Elvanpaşa and Tazlar. These rocks are macroscopically porphyritic aphanitic textured and mainly contain biotite and coarse-grained feldspar minerals. In addition, weathering structures such as the layers of a peeled onion are remarkable.

Based on the petrographic investigations of the volcanic rocks around Elvanpaşa village, they are called as trachyandesites and mainly composed of oligoclase, andesine + biotite ± volcanic glass ± opaque mineral ± hornblend (Figure 8a). These rocks have porphyritic texture with hypocrystalline hipidiomorph groundmass and the matrix consists of microlites and volcanic glass. Furthermore, some of the mafic minerals are opacitized.

The trachyandesites around Tazlar village can be easily distinguished from other groups by their trachytic texture. Vitrophyric, porphyritic, and hypocrystalline hypidiomorphic porphyritic textures are observed in thin sections. In trachyandesites including plagioclase + biotite ± volcanic glass ± opaque mineral ± hornblende, pilotaxitic and trachytic textures formed by microlite and volcanic glass are the characteristics of this rock group (Figures 8b and 8c).

Ignimbrites consist of 28% phenocryst and 72% groundmass. Ignimbrites with banded texture are composed of plagioclase + biotite + obsidian ± opaque mineral (Figure 8d). Plagioclases whose anorthite content is calculated by the Michel Levy method generally oligoclase-andesine (An_{25}-An_{46}) in composition, and they show polysynthetic twinning and a zoned structure. In subhedral and prismatic-formed plagioclase minerals, partial sericitization and rarely argillization are observed. Subhedral biotides generally have a porphyritic texture within the matrix. Light brown and red brown-colored biotites have quite typical basal cleavages and shows parallel extinction. In the sections, hydrobiotites are rarely observed while opacitizations are common. These opacitizations are formed especially in the rims of biotite minerals. There are splinter-shaped volcanic glass fragments, volcanic glass, and pumice pieces in the groundmass that connect the minerals in the rock and constitute the majority of the sections. The volcanic glass does not present a characteristic obsidian feature. Because this volcanic glass observed in ignimbrites is more like the volcanic glass splinters formed during the rock's cooling process. In addition, partial devitrification is also observed in the splinters.

The tuffs along ÖSK 4, where the *porphyric lava flows* were observed, are mainly composed of plagioclase + biotite + lithic component + pumice + splinter ± opaque mineral components (Figure 8e). The fact that there is no welding in the tuffs, where mainly vitrophyric texture is observed, is an important detail, and these tuffs are described as glassy tuff.

Plagioclase + biotite + lithic component + pumice + volcanic glass splinters ± opaque mineral components were observed in the beige-colored tuff samples located along ÖSK 5, where *porphyric lava flows* were seen. Basaltic andesite-type rock sections constitute the tuffs, which have lithic components (Figure 8f). The vesicles in the pumices that were observed in large amounts in the rock are generally circle-shaped.

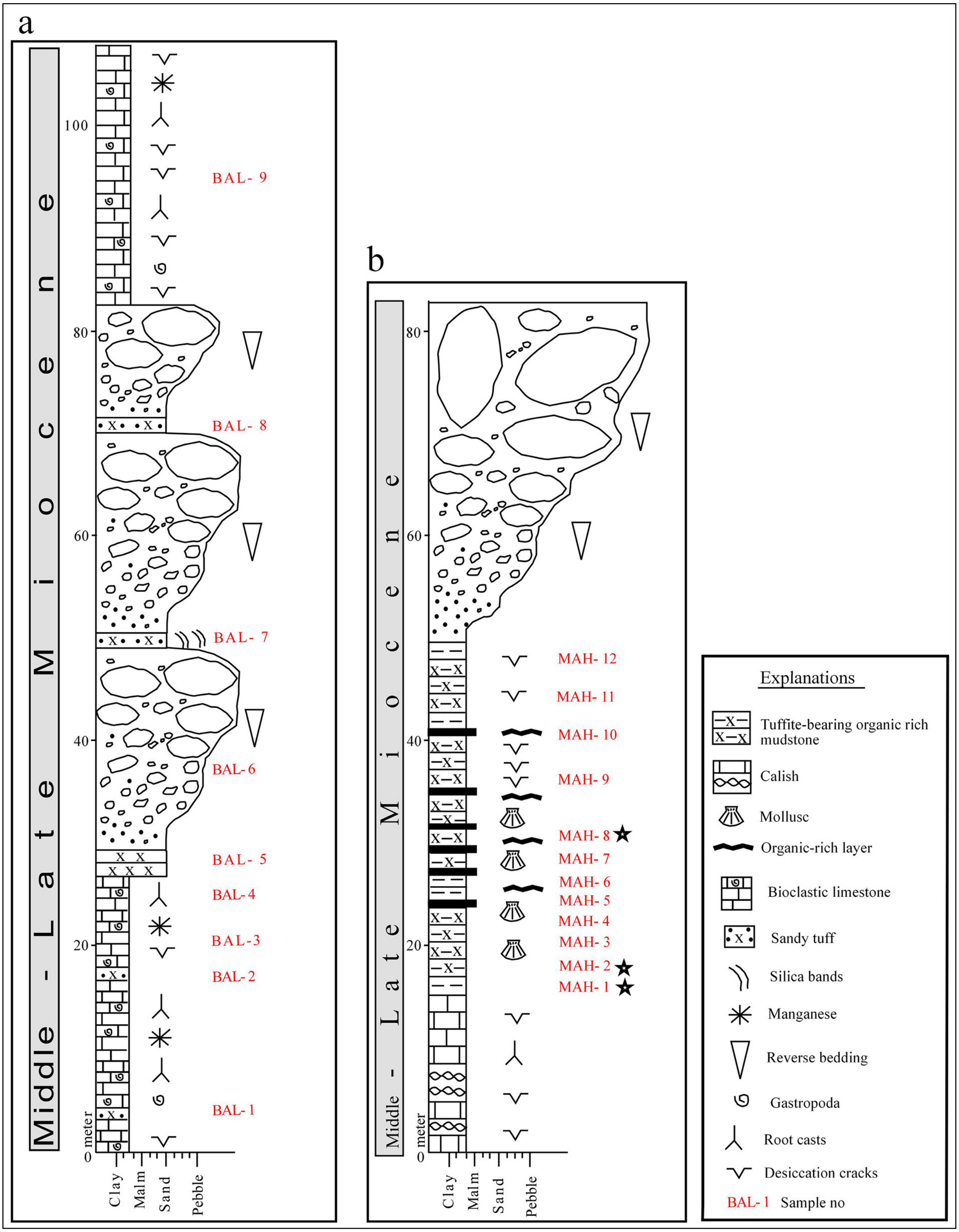

Figure 6- a) Measured Stratigraphic Section Line (ÖSK 4) with N35°E direction of pyroclastic units that exposed around the Balmahmut village. (ÖSK 4) and sample numbers for XRD analysis (middle-late Miocene), b) measured Stratigraphic Section (ÖSK 5) of pyroclastic units around the Balmahmut village and sample numbers for XRD analysis (middle-late Miocene).

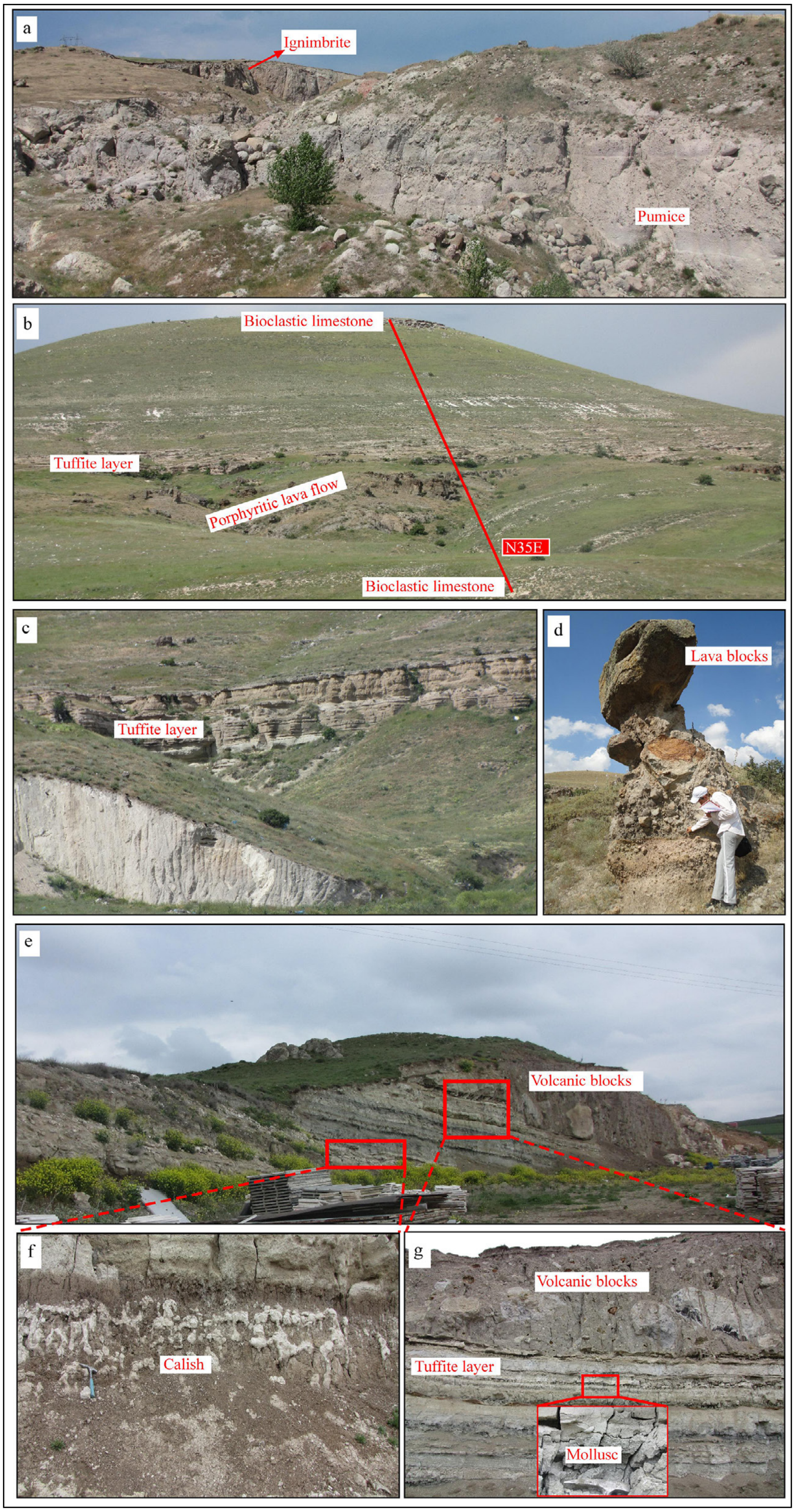

Figure 7- a) Lithic-rich ignimbrite (around the Balmahmut village, b) measured stratigraphic Section line with N35ºE direction (ÖSK 4, Balmahmut village), c) close up view of tuffite layers, d) poorly graded lava flows varying from ash size to block size, e) general view of the pyroclastics around the Balmahmut village (ÖSK 5), f) close up view of calishe layers, g) mollusc-bearing thin to medium bedded tuffs and block-sized pyroclastics.

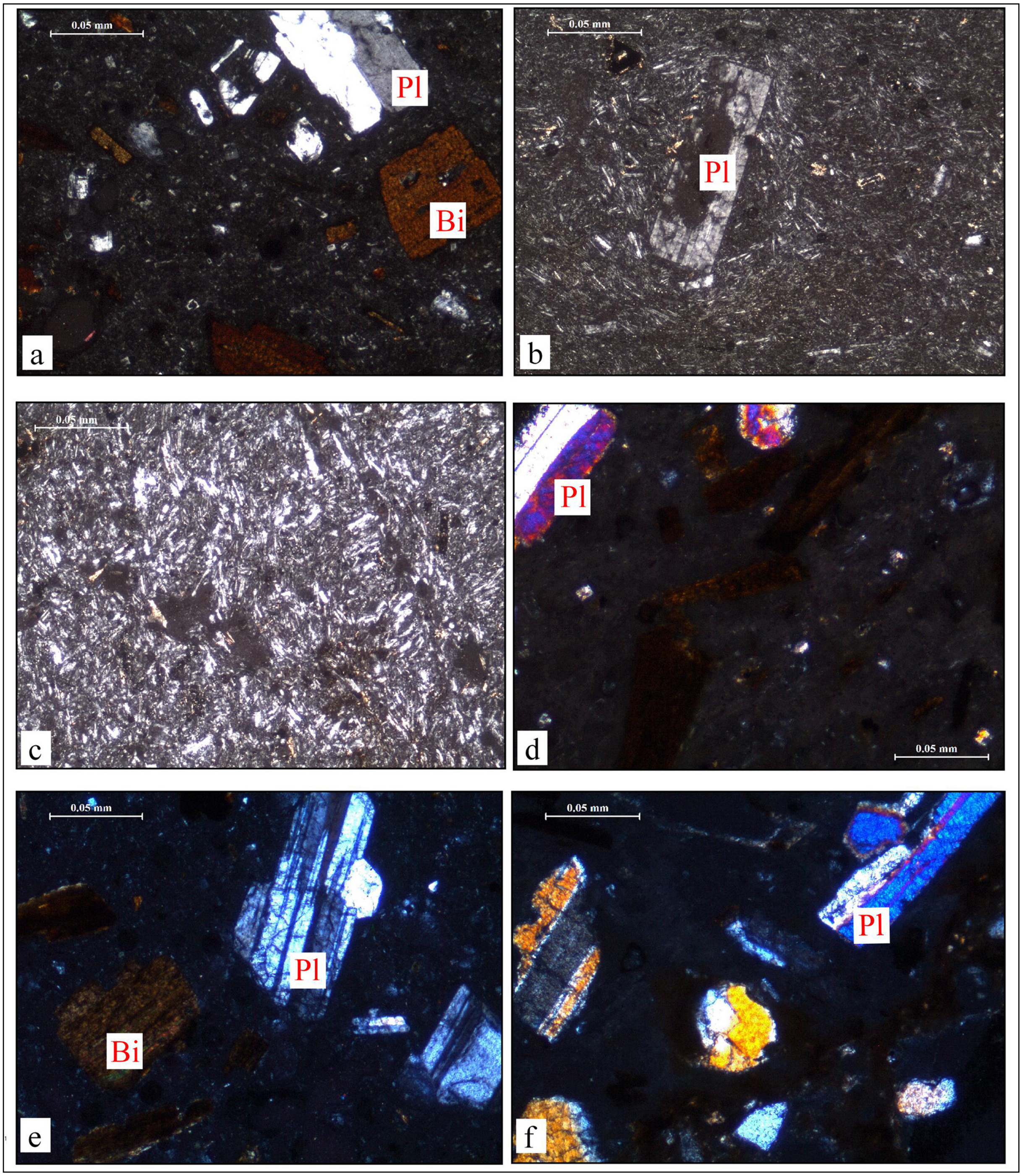

Figure 8- a) Microscopic image of trachyandesite around the Elvanpaşa village (Bt:biotite, Pl: plagioclase) (II. Nicol), b) pilotaxitic texture of the trachyandesite around the Tazlar village (II. Nicol), c) close up view of pilotaxitic texture, d) microscopic image of banded texture of ignimbrite (Çift nikol), e) microscopic image of glassy tuffs in ÖSK 4 (II. Nicol), f) microscopic image of lithic tuffs in ÖSK 5 (II. Nicol). Mineral abbreviations after Whitney and Evans (2010).

6. Mineralogical Properties of the Sinanpaşa Neogene Deposits

In order to determine the mineralogical compositions of mudstone, limestone, tuff and tuffite-bearing mudstone samples, The XRD results for the bulk samples and clay fraction analyses are presented in tables 1, 2, and 3 respectively. The lithological units are classified as: (1) middle Miocene coal-bearing mudstones, (2) middle-late Miocene tuffs, and (3) late Miocene limestone and tuffite-bearing mudstones.

Table 1- XRD results of Middle Miocene sediments (Mineral abbreviations after Whitney and Evans, 2010).

Samp. No	Lithology	Bulk Rock									Clay Fraction				
		Qz	Cal	Pl	Gp	Dol	The	Crs	Py	Amp	Sme	Ilt	Chl	Kln	Sep
KIR-1	mudstone										az	+		+	
KIR-2	mudstone											+		+	
KIR-3	mudstone	+										+		+	+
KIR-4	mudstone	+										+		+	
KIR-5	mudstone	+										+	+	+	
KIR-6	mudstone	+								+		+	+		
KIR-7	mudstone	+			+							+		+	
KIR-8	mudstone											+		+	
KIR-9	mudstone				+							+		+	
KIR-10	mudstone											+		+	
KIR-11	mudstone											+		+	
KAR-1	mudstone	+	+									+			az
KAR-2	mudstone	+	+			+					+	+			az
KAR-3	mudstone	+	+			+	+					+	+	+	
KAR-4	mudstone	+	+								+				
KAR-5	mudstone	+	+	+		+						+	+		
KAR-6	mudstone	+		+							+	+		+	
KAR-7	mudstone	+		+	+					+	+	+		+	
KAR-8	mudstone	+	+		+	+	+				+	+	+		
KAR-9	mudstone	+		+	+	+		+	+		+	+	+	+	
KAR-10	mudstone	+	+								+	+	+	az	
KAR-11	mudstone		+	+	+						+	+	+	az	
KAR-12	mudstone	+	+								+	+	+	az	
RIZ-1	limestone	+	+									+			
RIZ-2	limestone	+	+									az			
RIZ-3	limestone	+	+									az			
E -1	limestone		+												
E -2	limestone	+	+									+			
E -3	limestone		+												
E -4	limestone		+												
E -5	limestone	+										+			
E -6	limestone	+										+			

(Qz:quartz, Cal: calcite, Pl: plagioclase, Dol: dolomite, The: thenardite, Crs: cristobalite, Py: pyrite, Amp: amphibole, Sme: smectite, Ilt: illite, Chl: chlorite, Kln: kaolinite, Sep: sepiolite). Abbreviations after Whitney and Evans (2010).

6.1. Mineralogical Properties of the Middle Miocene Deposits

The rock units of this time period and their characteristic clay mineral types are given below.

Coal-bearing mudstones: Except for the clay minerals, the rock-forming minerals such as quartz, calcite and plagioclase are determined as the result of XRD analysis of samples taken from coal-bearing mudstones exposed along the main Afyon-Sandıklı road at the south of the basin and around Çalışlar-Karacaören villages at the north of the basin (Figure 9A). Quartz was determined by the reflection peaks of d(101) at 3.34 Å, d(100) at 4.25 Å, and d(112) at 1.817 Å. Calcite was characterized by the reflection peaks of d(104) at 3.02 Å, d(102) at 3.86 Å, and d(113) at 2.28 Å. Plagioclase was identified by the reflection peaks of d(040) at 3.18 Å, d(020) at 6.43 Å, and d(111) at 4.02 Å. In some samples, evaporite minerals such as dolomite, tenardite and gypsum have also

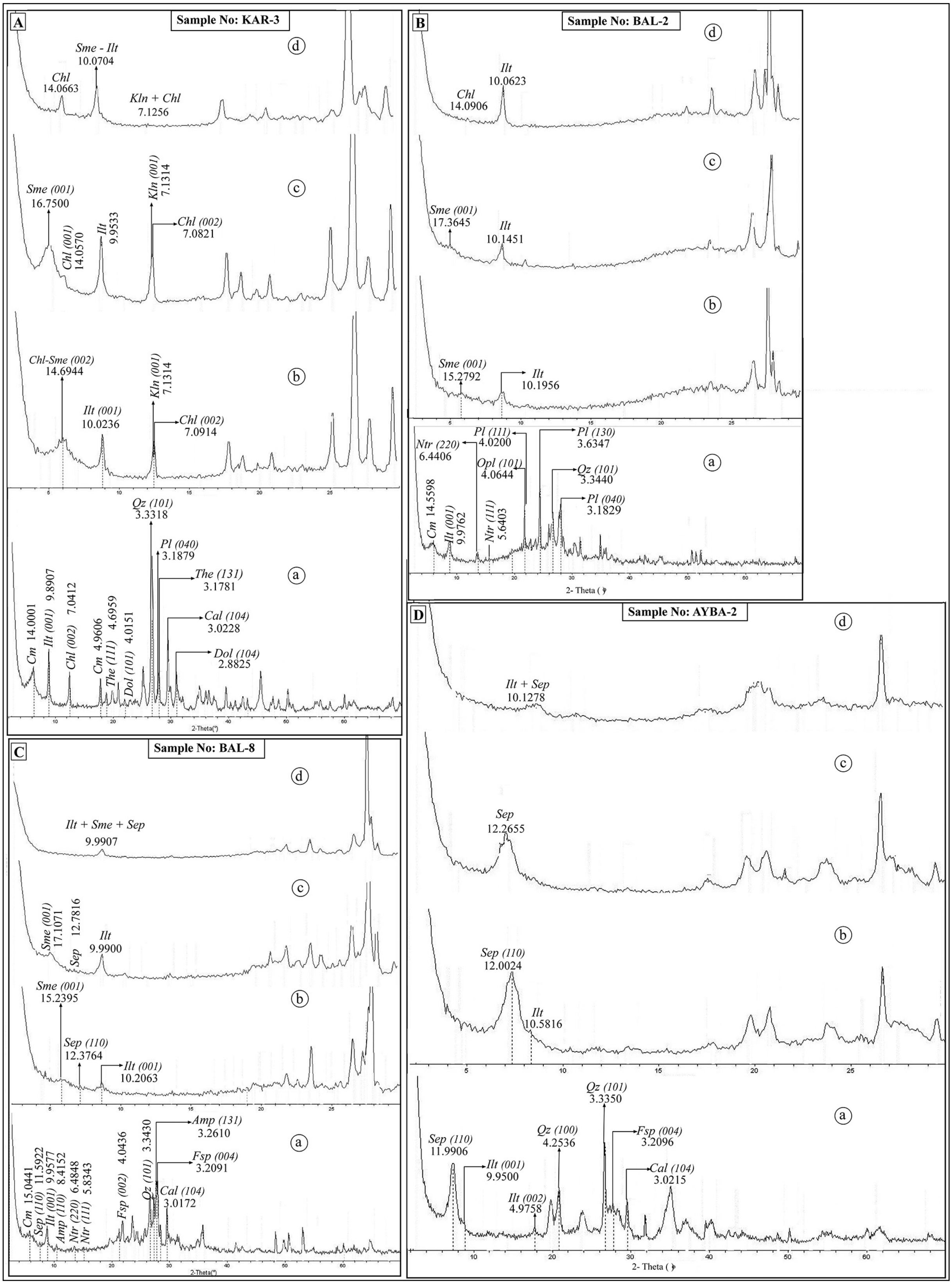

Figure 9- XRD diffractions of (A) KAR-3, (B) BAL-2, (C) BAL-8 and (D) AYBA-2 samples. Cm: clay, Sme: smectite, Kln: kaolinite, Sep: sepiolite, Chl: chlorite, Amp: amphibole, Dol: dolomite, Pl: plagioclase, Qz: quartz, Opl: opal-CT, Cal: calcite, The: thenardite, Ntr: natrolite (a = bulk rock, b = air-dried, c = ethylene glycol, d = heated at 550^{0}C. (Mineral abbreviations after Whitney and Evans, 2010).

been identified (Table 1). Accordingly, dolomite was determined by the reflection peaks of d(104) at 2.89 Å, d(101) at 4.03 Å, and d(113) at 2.19 Å. Thenardite was identified by the reflection peaks of d(020) at 2.78 Å, d(111) at 4.69 Å, and d(131) at 3.18 Å. Gypsum, which was observed in small amounts, was distiguished by the reflection peaks of d(020) at 7.67 Å, d(141) at 3.06 Å, and d(200) at 2.86 Å. Illite is the dominant clay mineral in the clay fraction analysis of the same samples. In addition to illite, smectite, kaolinite and a small amounts of chlorite were also detected. Sepiolite is especially associated with the organic rich parts (Table 1). Illite is determined by the reflection peaks of d(001) at 10.1 Å, d(002) at ~5 Å, d(003) at 3.35 Å, and d(116) at 2.55 Å. In addition, d(001) reflection peaks at 10.1 Å of illite which were not affected by air-dried, ethylene-glycolating and heating method also helped to determine the illite (Figure 9A). On the other hand, the air-dried smectites show a peak at around 15 Å with d(001) reflection which expanded to around 17 Å with d(001) reflection after saturation with ethylene glycol. Peaks at 7.14 Å and 3.57 Å with d(001) and d(002) reflections represent kaolinite. Its 7.14 Å peak collapsed after heating at 550°C (Figure 9A).

Carbonate deposits: Calcite is the dominant mineral as rock forming mineral in the rhizoliths exposed around Kırka village at the south of the basin and Kızılca village at the north and in some silicified parts it is accompanied by quartz. These samples contain ~ 5% illite. The characteristic basal peaks of the illite show reflections at ~10.1 Å d(001), ~5 Å d(002), 3.35 Å d(003), and 2.55 Å d(116) (Table 1).

6.2. Mineralogical Properties of the Middle-Late Miocene Pyroclastic Rocks

These deposits were interbedded and/or intercalated with the giant sized granular volcanic materials especially around the Balmahmut and İğdeli villages at the north of the basin, as seen in figures 6a and 6b. Pyroclastic products of Afyon volcanism which was active since middle Miocene (Aydar and Bayhan, 1995) were exposed following siliciclastic precipitation of the alluvial/fluvial fan system and they were deposited contemporaneously with the lacustrine carbonate deposition. As a result of the mineralogical investigations on tuffs and tuffite-bearing organic rich mudstone samples (Table 2), except from clay

Table 2- XRD results of Middle-late Miocene pyroclastics (Mineral abbreviations after Whitney and Evans, 2010).

Samp. No	**Lithology**	**Bulk Rock**									**Clay Fraction**			
		Qz	**Cal**	**Pl**	**Dol**	**Crs**	**Opl**	**Ntr**	**Amp**	**Sme**	**Ilt**	**Chl**	**Kln**	**Sep**
BAL-1	tuff. mudstone			+						+	+	az		
BAL-2	tuff. mudstone	+		+			+	+	+	+	+			
BAL-5	tuff. mudstone	+		+			+	+	+	+	+	az		
BAL-6	tuff. mudstone			+	+			+			+			
BAL-7	tuff. mudstone	+		+					+	+	+			
BAL-8	tuff. mudstone	+	+	+		+		+	+	az	+			az
MAH-1	tuff. mudstone	+	+	+						+	+		+	
MAH-2	tuff. mudstone						+			+	+			
MAH-3	tuff. mudstone		+	+						+	+			
MAH-4	tuff. mudstone			+						+	+	+	+	
MAH-5	tuff. mudstone		+							+	+			
MAH-6	tuff. mudstone		+					+		+	+			
MAH-7	tuff. mudstone						+			+	+		+	
MAH-8	tuff. mudstone		+							+		+		
MAH-9	tuff. mudstone		+	+									az	+
MAH-10	tuff. mudstone	+	+	+		+				+	+			+
MAH-11	tuff. mudstone	+	+	+		+				+		+	az	+
MAH-12	tuff. mudstone		+											+
MAH-13	tuff. mudstone		+						+	+	+	az		

(Q: quartz, Cal: calcite, Pl: plagioclase, Dol: dolomite, Crs: cristobalite, Opl: opal-CT, Ntr: natrolite, Amp: amphibole, Sme: smectite, Ilt: illite, Chl: chlorite, Kln: kaolinite, Sep: sepiolite, tuff. Mudstone: tuffite bearing mudstone) minerals, quartz, cristobalite, opal-CT, calcite, plagioclase, amphibole and zeolite (natrolite) have been determined (Figure 9B and 9C). Accordingly, the characteristic basal peaks of the cristobalite show reflection at 4.04 Å d(101), 2.84 Å d(102) and 2.49 Å d(200). Peaks at 8.40 Å d(110), 3.87 Å d(131) and 2.69 Å d(151) represent amphibole (Figure 9C). In addition, natrolite is characterized by the reflections at 6.55 Å d(220), 5.89 Å d(111) and 2.85 Å d(531). Smectite and illite are the dominant minerals in almost all the tuffite-bearing mudstone samples. Additionally, chlorite, kaolinite, sepiolite and a small amount of amorphous material were also determined in these samples (Table 2 and figure 9C).

6.3. Mineralogical Properties of the Late Miocene-Pliocene Deposits

According to the bulk-rock analyses of limestone and alternating tuffite-bearing mudstone samples, calcite and quartz are designated as the dominant minerals (Table 3). Apart from these minerals, illite, which was found in almost 60% of clay minerals, is determined in almost all samples, while it is accompanied by smectite and small amounts of sepiolite in some samples (Table 3 and figure 9D).

7. Scanning Electron Microscope (SEM-EDX) Determinations

Clay-dominated samples were studied by SEM in order to determine the micromorphology, textural and diagenetic characteristics of the clay minerals and their relationships with non-clay minerals. Accordingly, illite was observed in fibrous morphology (Figure 10a, table 4). Consistent with the XRD results, it is observed that illite was accompanied by quartz in SEM analysis. Subhedral-anhedral crystals of quartz support the detritic origin. Euhedral calcite crystals with rhombohedral symmetry observed as cement in the intergranular spaces (Figure 10b). In addition, the euhedral gypsum crystals (with monoclinic symmetry) around illites occasionally indicate sulphur enrichments in the diagenetic environment (Figure 10c). This enrichement which is notable particularly in coal-bearing levels can be attributed to the activity of sulfate-reducing bacteria in the diagenetic environment (Castro et al., 1999). Chlorite

Table 3- XRD results of Late Miocene-Pliocene units (Mineral abbreviations after Whitney and Evans, 2010).

Samp. No	Lithology	Bulk Rock				Clay Fraction			
		Qz	Cal	Pl	Amp	Sme	Ilt	Chl	Sep
AYV-1	limestone		+						
AYV-2	limestone		+				+		
AYV-3	limestone		+				az		
BAL-3	limestone		+						
BAL-4	limestone		+						az
BAL-9	limestone		+						
AYBA-1	tuff. mudstone			+	+	+	+		
AYBA-2	tuff. mudstone	+		+	+		+		
AYBA-3	tuff. mudstone	+		+	+		+		+
AYBA-4	tuff. mudstone	+				+	+	+	
AYBA-5	tuff. mudstone	+	+			+	+		
AYBA-6	tuff. mudstone	+				+	+	+	
AYBA-7	tuff. mudstone	+	+			+	+		
AYBA-8	tuff. mudstone	+	+			+	+		
AYBA-9	limestone		+						az
AYBA-10	limestone		+						
AYBA-11	limestone		+						az
AYBA-12	limestone		+						

(Qz: quartz, Cal: calcite, Pl: plagioclase, Amp: amphibole, Sme: smectite, Ilt: illite, Chl: chlorite, Sep: sepiolite, tuff. Muudstone: tuffite bearing mudstone)

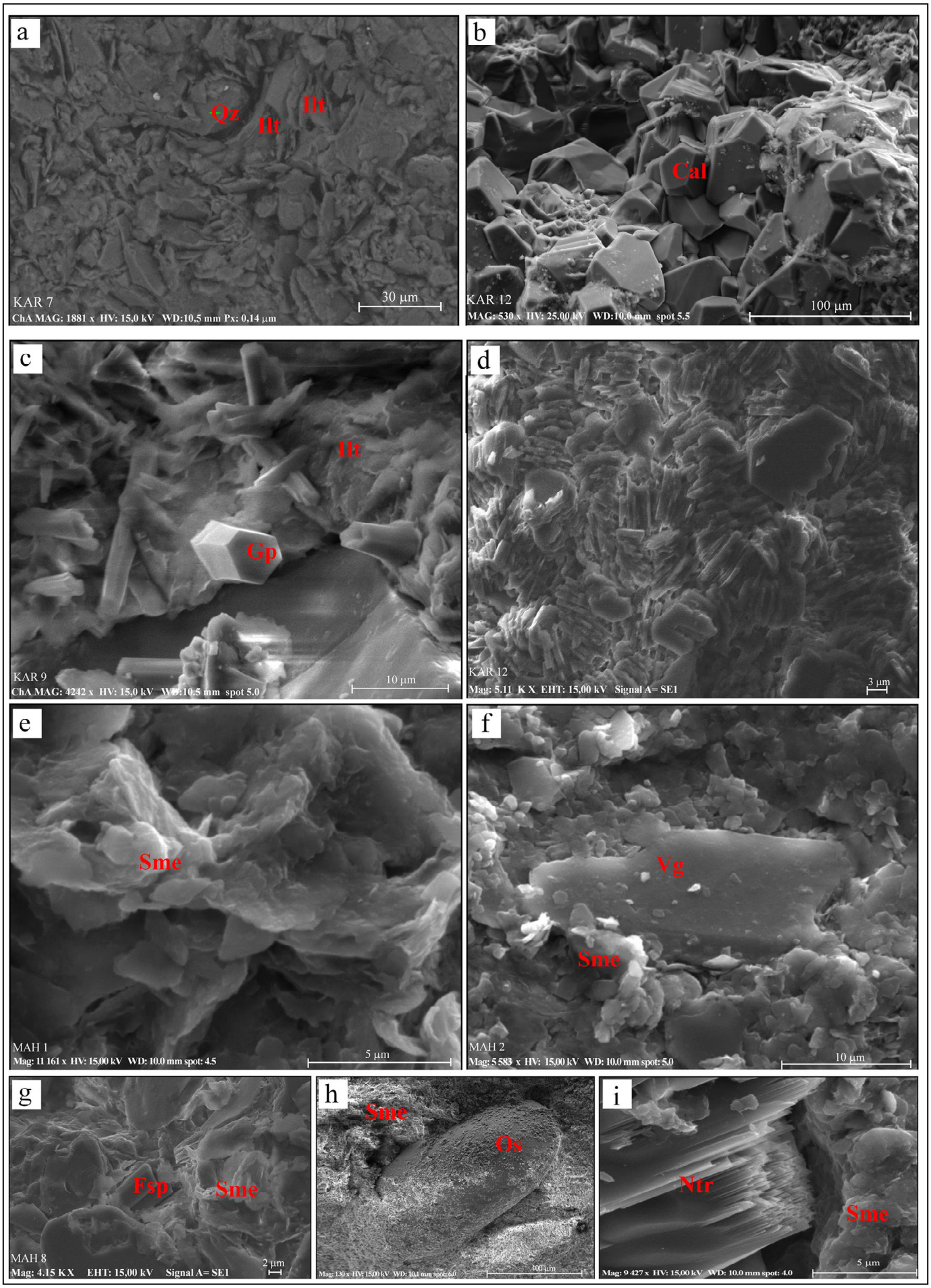

Figure 10- SEM images of a) fibrous illite (Ilt) and accompanying quartz (Qz) minerals, b) idiomorphic calcite (Cal) with trigonal symmetry, c) idiomorphic gypsum crystals (Gp) with monoclinic symmetry between illite minerals (Ilt), d) kaolinite, e) smectite mineral (Sme) with corn flakes and honeycomb texture, f) smectite mineral (Sme) developing on the edges and surfaces of fractured volcanic glasses (Vg), g) smectite mineral (Sme) on and around feldspar (Fsp), h) ostracod shell (Os) on plate-crystal of smectite, i) natrolite mineral (Ntr) and surrounding honeycomb textured smectite mineral (Sme). (Mineral abbreviations after Whitney and Evans, 2010).

Table 4- EDX analyses of illite, chlorite, kaolinite and plagioclase (wt.%).

Samp. No	SiO_2	Al_2O_3	FeO	MgO	K_2O	Na_2O	CaO	Total
KAR- 7*	53.1	33.98	1.83	1.83	9.73	-	-	99.98
KAR-3	41.78	26.26	10.99	20.96	-	-	-	99.99
KAR-12	46.92	43.68	1.94	-	7.34	-	-	99.88
BAL-2	68.71	19.82	-	-	-	11.00	0.47	100.00

*KAR-7: illite; KAR-3: chlorite; KAR-12: kaolinite; BAL-2: plagioclase.

is represented by thin stacks, pseudohexagonal and randomly oriented lath shaped plates with 5-10 µm crystal thickness. Chemical analyses by EDX determine the chlorite is iron rich (10.99%) (Table 4). The presence of pore-space filling chlorites with iron-rich composition indicates the diagenetic occurrence (Bartier et al., 1998). Kaolinite occurs as platy crystals with hexagonal shape, typically arranged in elongated stacks of books (Henning and Störr, 1986) (Figure 10d). The plates are 1-3 µm wide and generally occur as irregular forms. The EDX analysis of these plates shows similarities to kaolinite (Table 4). The concentration of kaolinite, especially in the coal-bearing levels, can be attributed to the autogenetic deposition of silicon and aluminum there (Koukouzas et al., 2009).

Smectite is observed as plate-shaped, honeycomb texture and in the form of wavy leaves (Figure 10e). Wavy morphology was described as cornflake texture by Keller (1978). Smectite leaves are developed on the surface of volcanic glass and/or in the fractures around the glass particles (Figure 10f). In addition, in some samples, smectite is also developed by replacing the partially dissolved edges of plagioclases. According to EDX analysis, sodic feldspars with high Na content (Table 4) are observed as euhedral, prismatic, and rod-shaped (Figure 10g). Smectite fills the voids between the plagioclases and the glass particles. Therefore, smectite may act as a main cement in places. Samples having plate-shaped smectites, up to 1-4 µm in diameter, are mostly observed in tuffite-bearing mudstones which also contain ostracod shells (Figure 10h). The elemental composition of the plagioclase and volcanic glass that were partially replaced to smectite are composed of Si, Al, Fe, and Na. On the other hand, EDX analysis of pure smectite represents Mg, K, Na, and Fe, as well as Si and Al (Figure 11a). This mineralogical composition indicates that smectite is occurred as a result of partial dissolution of both volcanic glass and plagioclase. It also results from replacing these minerals, as well as occurring by authigenic precipitation in microfracture-fillings of the sediments (Fischer and Schmincke, 1984).

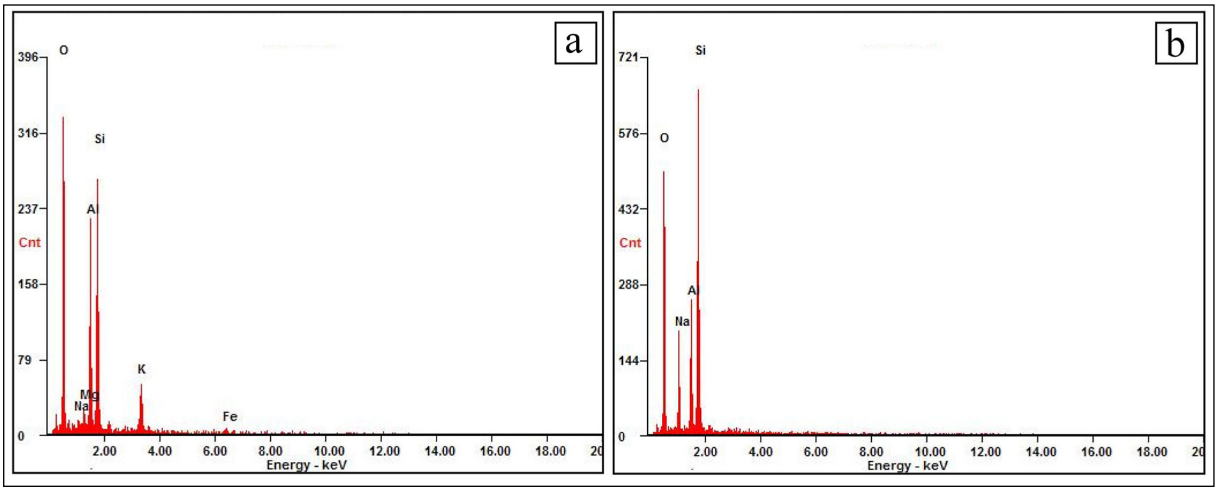

Figure 11- a) Energy Scattering Spectrometry (EDX) analyses of smectite, b) energy scattering spectrometry (EDX) analyses of natrolite minerals.

Zeolite-type minerals were also observed in SEM analysis (Figure 10i). EDX results were consistent with the element content of zeolite. Among them, natrolite, mainly accompanied by smectite, is characterized by strong Si, Al, and Na peaks (Figure 11b). Smectite has a honeycomb texture around the natrolite crystals. These mineralogical and micromorphological features indicate that the smectite and accompanying natrolite were formed as a result of the hydration of volcanic glasses and/or plagioclases (Karakaş and Kadir, 2000).

8. Discussion and Conclusions

One of the important parameters that control the formation of clay minerals in sedimentary sequences is the climatic factor. Clay mineral analysis, therefore, plays an important role in understanding ancient climate changes at different time periods (Singer and Galan, 1984; Moore and Reynolds, 1997; Thiry, 2000; Kemp et al., 2016). Similarly, the middle Miocene clay mineral diversity in the investigation area would have been influenced by climatic changes. The dominant illite-chlorite coexistence in the basin margin and marsh environments observed in the middle Miocene supports temperate and humid conditions (Weaver, 1989). Based on the palynological data, Yavuz-Işık et al. (2011) argued that almost subtropical climatic conditions were dominant in the early Miocene while temperate conditions prevailed towards the end of the middle Miocene in Western and Central Anatolia. Akkiraz et al. (2015), similarly, suggested the deposition of coal-bearing deposits in the Soma and Uşak-Güre (early-middle Miocene) basins occurred in humid and warm conditions. In addition, Kayseri and Akgün (2010) mentioned the existence of temperate climatic conditions indicate the Miocene Climatic Optimum during late Burdigalian-?Serravalian in northwestern Anatolia (Balıkesir-Gönen, Çanakkale-Çan) and during late Burdigalian-Langhian in southwestern Anatolia (Muğla-Milas). All these data are also consistent with the Miocene depositional systems of Sinanpaşa basin and their clay mineralogy. However, although evaporite minerals such as dolomite, gypsum, and thenardite are observed in the coal-bearing mudstones, these aridifications are thought to be periodic or controlled by limited changes in the diagenetic environment. For example, it is common for gypsum within the coal layers to be formed by sulfate-reducing bacteria (Castro et al., 1999). For this reason, their reflection on the diagenesis environment together with the arid and semi-arid intermediate phases in the humid climatic belt should not be neglected (Sanz et al., 1994).

It is known that compressional and extensional tectonic movements in Western Anatolia caused the development of numerous basins restricted by normal faults during Cenozoic era (Dewey and Şengör, 1979; Şengör and Yılmaz, 1981; Koçyiğit, 1984; Robertson and Dixon, 1984; Şengör et al., 1985; Zanchi et al., 1993; Bozkurt, 2000; Koçyiğit et al., 2000; Seyitoğlu et al., 2004; Koçyiğit and Deveci, 2007; Ersoy et al., 2011). These basins were filled with sedimentary, volcanic, and volcaniclastic/pyroclastic units. Sinanpaşa basin, located at the eastern margin of the Western Anatolian graben system, is one of the much-debated basins with regard to the formation and development of Western Anatolia basins. It began to form as a NW-SE trending basin in the central part of the Akşehir-Simav Fault System during early Miocene (Koçyiğit and Deveci, 2007). The lithologic associations of the middle Miocene, middle-late Miocene and late Miocene-Pliocene periods overlying the Paleozoic and Mesozoic basement were distinguished from the bottom to the top as follows:

Middle Miocene period was commenced with fluvial deposits and followed upward by lacustrine sediments. The fluvial deposits represented by conglomerate and cross-bedded sandstone are well exposed at the south of the basin. On the other hand, lacustrine deposits have widespread outcrops around both southern and also north-northwestern part of the basin. Similar to the investigated area, some other Neogene freshwater lake deposits (e.g. Uşak-Güre, Manisa-Soma, Kütahya-Seyitömer) in Western Anatolia occasionally contain economic coal levels. On the basis of radiogenic, paleontological, and palynological data, many researches accepted that coal-occurrences in the neighboring basins are early Miocene (Uşak-Güre: Karaoğlu et al., 2010; Ersoy et al., 2011) and late early-middle Miocene (Soma: Akgün and Akyol, 1987, 1999; Kaya et al., 2007; Kayseri and Akgün, 2008; Seyitömer: Nakoman, 1968; Yavuz-Işık, 2007) in age. On the other hand, based on the palynological data in the investigated area, coal-bearing layers are late middle Miocene in age (Akıska, 2017; Akıska and Varol, 2020). The coal-bearing lacustrine deposits were followed upward by volcanics. Afyon volcanism was active in the study area due to the extensional tectonic regime in Western Anatolia during the middle-late Miocene period (Keller and Villari, 1972; Besang et al., 1977; Aydar and Bayhan, 1995; Aydar, 1998; Akal et al., 2013; Prelevic et al., 2015). Materials from the volcanic activity occasionally filled the local lake areas (Aydar and Bayhan, 1995). During this time period, most common lacustrine deposits have widespread outcrops along the N-NE margin of the basin. The depositional sequence initiates with bioclastic limestone and alternates upward with lava flows and tuffite layers (Figure 1c). Volcanic activities became largely ineffective toward the end of late Miocene and ostracod and mollusk-bearing lacustrine deposits became clear. Limestones with abundant clastic (bioclast and pellet) and/or organic material contain widespread desiccation cracks. This indicates shallow local lake environments and also drying up conditions from time to time (Eugster and Kelts, 1983; Bustillo et al., 2002). In addition, laminated tuffite-bearing organic rich mudstones alternating with limestones indicate the water level fluctuations of the lake in certain climatic periods and changing to anoxic conditions (Sáez and Cabrera, 2002).

The mineralogical compositions of the lacustrine deposits have been greatly influenced by the climatic factors, mineralogy of volcanic material additives and diagenetic environment changes. According to the geological time processes, the association of calcite and quartz is dominant in the lacustrine mudstones of the middle Miocene and the limestone-clayey limestones overlying them (Table 1). These anhedral-subhedral quartz crystals indicate the detritic origin and are derived from non-volcanic basement rocks. The fact that the different degree of silicified volcanic glass observed at this level surrounded by illite also indicates the diagenetic origin. In a few mudstone samples (KAR 7-11), the association of calcite and quartz is accompanied by the feldspar mineral. The feldspars are detected as Na-rich plagioclases based on the petrographical and mineralogical studies. The limited propagation of mudstones and rhythmic alternation of coal layers in the lower and middle parts of this sequence reflect the features of typical marsh coals that developed at the basin margins before inundation of the lake (Miall, 1977; McCabe, 1984). Illite and chlorite were determined as the dominant clay minerals in these coal-bearing deposits. Except that, very rare smectite, kaolinite and sepiolite type clay minerals are noteworthy in some samples. Illite represents the clay-size component of muscovite and is a marker of detritic origin along with the chlorite minerals (Ehrmann et al., 2005; Fagel, 2007). However, the areas where Fe-rich chlorite in the coal levels are seen as pore fillings, may also be genetically linked to diagenetic origin (Bartier et al., 1998). Kaolinite, on the other hand, may have formed by the in situ / authigenic deposition of silicon and aluminum elements dominant in these environments as a result of the chemical decomposition of feldspars (mostly sodic plagioclases) in high water-active environments (Koukouzas et al., 2009) and also can be transported detritically to this environment (Keller, 1978). Non-clay minerals such as quartz, feldspar, cristobalite, opal-CT, amphibole, and zeolite (natrolite) were detected in the middle-late Miocene pyroclastic deposits. Zeolite is thought to be formed as a result of alteration of volcanic ash or tuff in the water environment (Ataman, 1977; Grim and Güven, 1978; Yalçın, 1988; Bayhan and Yalçın, 1990). In particular, smectite with a honeycomb-shaped texture around and on the zeolite (natrolite) indicates these minerals are occurred by the hydration of volcanic glasses and/or plagioclases (Karakaş and Kadir, 2000). In addition, the determination of chlorite-, kaolinite-, and sepiolite-type clay minerals accompanied by smectite in these levels may mark the result of the in-situ alteration of volcanic material in the lacustrine environment (Millot, 1964; Grim, 1968; Weaver, 1989). Therefore, the formation of smectite may have occurred by the alteration / dissolution of plagioclase and volcanic glass, and also it may occur by the transformation from amorphous to semi-amorphous structure in the alkaline conditions, and then to smectite (Karakaş et al., 2007; Karakaya et al., 2007; Bayhan and Yalçın, 1990; Gürel and Kadir, 2006). Beside, smectite is observed on the microfracture fillings and in the dissolution voids of volcanic glasses. As a result of marginal replacing of volcanic glasses and feldspars by smectite and also due to partially/wholly dissolution of these minerals, smectite may authigenically form in the sediment voids (Millot, 1970; Furnes, 1975; Tucker, 1992; Christidis et al., 1995; Kadir and Karakaş, 2000, 2002; Besbelli and Varol, 2002; Karakaş et al., 2007). It is known that the association of smectite with volcano glass observed in volcano-sedimentary lacustrinal basins and the presence of alkaline environment are important factors for sepiolite formation (Weaver and Beck, 1977; Starkey and Blackmon, 1979; Singer and

Galan, 1984; Velde, 1985). Sepiolite formation in the study area must have been controlled by the presence of volcano glass and feldspar minerals, which constitute the main component of volcanic units. In this period, due to the increase of fresh water discharge to the lake area, the drought phases that commenced following the enrichment of the lake water by the elements such as silica and magnesium transported from the volcanic and surrounding rocks, respectively should have provided suitable conditions for sepiolite formation (Weaver, 1984). In the phases following widespread smectite formation in the lake water fed by volcanic sources, the formation of zeolite (natrolite) minerals have occurred due to the enrichment of the lake water by Na, Al and K ions and the increase in pH (Gall and Hyde, 1989; Stamatakis, 1989; Hartley et al., 1991; Karakaş and Kadir, 2006). Therefore, zeolite minerals that accompany smectite minerals in the study area indicate arid or semi-arid climatic conditions (Mariner and Surdam, 1970; Gall and Hyde, 1989; Hartley et al., 1991; Renaut, 1993; Türkmenoğlu et al., 1995; Karakaş and Kadir, 2006). The smectite and kaolinite minerals in the study area must have been formed authigenically from the volcanic and pyroclastic units depending on the acidic and basic character of the environment (Chamley, 1989). As a result, the sedimentary sequence developing in the early Miocene-Pliocene range in Sinanpaşa Neogene basin and the mineralogical components that characterize them were shaped largely due to the changes in paleotopography, paleoclimate, volcanism, tectonism and diagenetic environment.

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