Paleoenvironmental conditions during deposition of Kolosh and Gercus formations in northern Iraq as deduced from clay mineral distributions

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Abstract
A mineralogical study using X-ray diffraction supported by scanning electron microscopic examination on the Paleocene-Eocene Kolosh and Gercus formations from northern Iraq is conducted to show the distribution of clay minerals and their paleoenvironmental implications. Smectite palygorskite, kaolinite, illite, and chlorite are commonly present in varying proportions within the Kolosh and Gercus formations. Association of smectite and chlorite in the claystone of the Paleocene Kolosh Formation refers to marine environment of this formation, whereas development of palygorskite fibers from smectite precursor may relate to post-depositional diagenesis. In addition, the abundance of illite and kaolinite in the Eocene Gercus Formation suggests a greater influence of terrigenous input in humid conditions, affecting the distribution of these clay minerals. The study shows vertical change in clay minerals distribution when illite and kaolinite dominate in the Eocene Gercus Formation, in comparison to chlorite and smectite abundance in the Paleocene Kolosh Formation which may relate to global warming in the Eocene.

Keywords: Clay minerals, Paleocene-Eocene, Kolosh, Gercus, Paleoenvironmental conditions, Iraq

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Introduction

Highly sensitive changes in clay mineral structures due to changes in temperature and pH in their surroundings make them useful indicators for changes in paleoenvironmental conditions [1]. Generally, clay minerals are the products of sedimentation and diagenesis under certain circumstances related to provenance, climate, and water conditions; therefore, they have important implications for interpreting the paleoenvironmental and paleoclimatic history [2-5].

In the Cenozoic sedimentary successions from northern Iraq, clay minerals of Paleocene to Eocene rocks (Figure 1) representing the Kolosh and Gercus formations were subjected to a detailed mineralogical study. The studied successions are characterized by a thick deposition of clastics and carbonates represented by the Kolosh Formation (Paleocene-lower Eocene) and the Gercus Formation (Middle Eocene). Grey to green colored shale, sandstone, marl, and thin limestone of the Kolosh Formation were deposited in a narrow rapidly subsiding trough setting.

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and represent marginal marine environment [6]. Whereas, the Gercus Formation comprises red-colored sandstone and mudstone that were deposited in continental environments such as alluvial fans, river floodplains, lakes, and deltas [7-8].

In a global context, major climatic changes were observed at the late Paleocene-early Eocene interval, including various sedimentological, mineralogical, and paleontological variations, as an intertemperate episode of global warming representing a significant impact on both marine and terrestrial ecosystems [9-10].

In the present work, X-ray diffraction (XRD) supported by scanning electron microscopic (SEM) study is achieved for the marl, shale, and mudstone units from the two formations in selected section in Shaqlawa area, northern Iraq (Figure 1). We aimed to elucidate paleoenvironmental interpretation of the Kolosh and Gercus formations (Paleocene-Eocene) succession based on data deduced from clay mineralogy.

Geological setting

Iraq is located in the northeastern part of the Arabian Plate which forms a part of the long and wide northern passive margin of Gondwana bordering the Paleo-Tethys Ocean [11]. This part is a foreland basin that was created in response to loading of the crust by thrust sheets generated due to compression [11]. The evolution of the basin in terms of sedimentary environment, succession thickness, and vertical trends is strongly dependent on the degree of compressional tectonic activity [12].

In northern Iraq, the middle Paleocene- Eocene was deposited in Megasequence AP10 according to Sharland et al. [13] during a period of renewed subduction and volcanic activity associated with the final closure of the Neo-Tethys.

The Kolosh Formation represents the deepest and mobile sedimentary basin of Paleocene-lower Eocene cycle of Iraq [14]. It is connected with gradual lateral passage with the previously described Aaliji Formation and represents its contemporaneous clastic facies.

The studied Kolosh and Gercus formations are cropping out in a narrow belt of the High Folded belts (Figure 1). In this region, Kolosh clastics represent deep sea sediments that were spilled over in the narrow Neo-Tethyan Ocean onto the passive continental margin of the Arabian Plate from the approaching active margins of the Iranian and Turkish Plates [15]. The Middle Eocene Gercus Formation represents a typical continental red bed succession. It is dominated by clastic sedimentation of conglomerates, sandstones, mudstones, and marls, with some carbonates and evaporates deposited under an arid to semi-arid climate [7, 8, 16].
Figure 1—Paleogeographic and facies maps of Late Paleocene (left) and Middle Eocene (right), after [17], showing the narrow belt of the Kolosh and Gercus formations cropping out in northern Iraq, along with the location of the studied section.

Materials and Methods
Clay mineral analysis was performed by x-ray diffraction of selected samples from the studied formations in northern Iraq (Figures 1 and 2). Thirty-five samples (20 from Kolosh and 15 from Gercus formations) were collected from the claystone members in both formations. Representative scans for x-ray diffractograms are included in Figures 3 and 4. Bulk samples were analyzed using Phillips Spellman DF3 diffractometer with Cu-α radiation at the School of Earth and Environmental Sciences of Wollongong University, Australia. SEM analysis was performed using Camscan MV 2300 at the School of Material Engineering of Wollongong University, Australia. Additional SEM analysis was conducted at the Steinmann Institute of Bonn University, Germany, using a Camscan MV 2300 SEM with a calibrated energy dispersive X-ray analysis system.
Results

X-Ray diffraction analysis revealed the presence of smectite palygorskite, kaolinite, illite, and chlorite in varying proportions between the Kolosh and Gercus formations (Figures 3 and 4).

In the Kolosh Formation, chlorite is the abundant mineral observed, in addition to kaolinite, smectite, and palygorskite (Figure 3), whereas the main clay minerals observed in the Gercus Formation are illite and traces of kaolinite and chlorite (Figure 4). Associations of these minerals are used to discuss the paleoenvironmental conditions and terrigenous input.

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**Figure 2**-Simplified lithological section for the studied Kolosh and Gercus formations in Shaqlawa area showing samples location and distribution of clay minerals

| Formation | Lithology | Sample No. | Clay minerals distribution |
|-----------|-----------|------------|---------------------------|
|           |           |            | S  | Ch | K  | I  |
| Gercus    |           | G 15       |     |     |     |     |
|           |           | G 13-14    |     |     |     |     |
|           |           | G 11-12    |     |     |     |     |
|           |           | G 10       |     |     |     |     |
|           |           | G 9        |     |     |     |     |
|           |           | G 6-8      |     |     |     |     |
|           |           | G 5        |     |     |     |     |
|           |           | G 3        |     |     |     |     |
|           |           | G 2        |     |     |     |     |
|           |           | G 1        |     |     |     |     |
| Khurmala  |           | K 20       |     |     |     |     |
|           |           | K 17-19    |     |     |     |     |
|           |           | K 16       |     |     |     |     |
|           |           | K 15       |     |     |     |     |
|           |           | K 14       |     |     |     |     |
|           |           | K 13       |     |     |     |     |
|           |           | K 12       |     |     |     |     |
|           |           | K 10-11    |     |     |     |     |
|           |           | K 9        |     |     |     |     |
|           |           | K 7-8      |     |     |     |     |
| Kolosh    |           | K 6        |     |     |     |     |
|           |           | K 5        |     |     |     |     |
|           |           | K 4        |     |     |     |     |
|           |           | K 3        |     |     |     |     |
|           |           | K 2        |     |     |     |     |
|           |           | K 1        |     |     |     |     |
| Shiranish |           | Limestone  |     |     |     |     |
|           |           | Sandstone  |     |     |     |     |
|           |           | Marl       |     |     |     |     |
|           |           | Clayston/shale |   |     |     |     |
Scanning electron micro-images (Figures 5 and 6) show that the studied clay minerals in both formations are of either terrigenous (detrital) and/or authigenic and diagenetic origin. Smectite is present in framboidal shapes, commonly with outgrowing of palygorskite fibers, reflecting its diagenetic origin from smectite precursor; however, isolated fibers of palygorskite are also common. Illite exists either in flaky plates or in fibers, whether these fibers are isolated or filling fractures. Kaolinite commonly occurs in platy hexagonal degraded forms.

**Figure - 3** Representative X-Ray diffractograms of claystones from the Kolosh Formation (Samples K6 and K13 for the upper and lower diagrams, respectively; see Figure 2 for samples location).
Figure 4 Representative X-Ray diffractograms of claystones from the Gercus Formation (Samples G3 and G11 for the upper and lower diagrams, respectively; see Figure 2 for samples location).
Figure 5-Scanning electron micrographs of clay minerals in the Kolosh Formation. A- Feldspar grain showing transformation to platy kaolinite (arrow). B- Common flaky nature of clay minerals in claystone showing degraded platy kaolinite (arrows). C- Framboidal smectite (white arrow) and a precursor for palygorskite fiber (black arrow). D- Flaky illite (white arrow) and degraded platy kaolinite (black arrow).
Figure 6 Scanning electron micrographs of clay minerals in the Gercus Formation. A- Common illite flakes and plates (arrow). B- Palygorskite in both authigenic condensed accumulation form (white arrow) and separated detrital broken fibers (black arrow). D- Fibrous illite filling and hillsing pores or fractures (white arrows). E- Degraded platy kaolinite (white arrows).

Discussion
Detrital clay minerals are the end product of continental weathering. These minerals are useful indicators for the past changes in weathering regimes. In general, continental weathering is highly affected by climatic change, which in turn affects the weathering rates, runoff, soil formation, and transport of terrigenous (detrital) material to the sea [3, 18, 19].
The sedimentation of detrital clay minerals in marine environments takes place mainly through fluvial and/or eolian pathways [20]. Marine sediments may store a record of the environmental conditions and allow comparing these with changes in oceanographic circulation and with global temperatures [21]. Presuming that geology and geomorphology of the source region remained fairly stable for the time period in consideration in a tropical region, rainfall seems to be the main factor determining the composition of clay minerals in marine sediments [22-23]. Aridity and humidity in the source area are affected by climate, which in turn affects the composition of clay minerals. Therefore, similar rock types undergoing weathering in different climatic conditions could give rise to different clay mineral assemblages [24]. The clays formed and deposited within the sequences of the Kolosh and Gercus formations (Paleocene-Eocene) in northern Iraq are indicative of the deposition of these formations in several types of environment, extending from continental to transitional environments (Gercus Formation) to deep marine environments, represented by the thick, grey to green colored deposits of the Kolosh Formation.

By tracking the results of the mineralogical analysis of clay minerals typing using the techniques of XRD and SEM, a group of clay minerals, including smectite, palygorskite, illite, chlorite, and kaolinite were recognized. It is noted that the clay minerals in the studied claystone are detrital and/or authigenic and diagenetic in origin. Clay minerals have undergone little change in the zone of weathering and are chemically unreactive in deep oceans [3].

Kaolinite and smectite could be formed by crystal growth in the basin of deposition, at the expense of muscovite, k-feldspar, and plagioclase. However, kaolinite is more likely to be inherited from kaolinitic source, since detrital kaolinite is very unlikely to form in seawater [25]. Previous studies confirmed the presence of kaolinite in the river environments (Gercus) [26], which is established in these continental environments due to the presence of acid solutions that have a suitable environment for sedimentation [1].

Detrital origin of kaolinite relates mostly to derivation from igneous rocks that are rich in potash feldspars or from reworking of older sedimentary rocks [27]. Presence of detrital kaolinite in the form of degraded hexagonal plates (Figures 5B, D and 6D) is an indication of little effect of chemical weathering in the source area [3].

Authigenic formation of palygorskite is commonly observed in lagoons and evaporitic basins [27]. Palygorskite can also be formed by transformation from precursor clays (Figure 5C) during early diagenesis [3], by direct crystallization in calcareous soils, or as results of hydrothermal alteration of basaltic glass in the open oceans in association with fore-arc basins [28].

In the weathering zones, illite commonly forms due to alteration of muscovite, biotite, and k-feldspar [29]. Presence of illite as flakes or fibers (Figures 5D and 6A, C) may indicate the altered form of illite from older feldspars or other silicate minerals.

Smectite formation is favored in marine environments with mild alkaline, available Ca, paucity of K, and high Si and Mg. Poor drainage is necessary because otherwise water can leach away ions (e.g. Mg$^{+2}$) released in the alteration reactions [3].

The changes observed in clay minerals in the studied Paleocene-Eocene succession suggest interaction of clay minerals of terrigenous and marine sources in the Kolosh Formation. The dominance of chlorite and smectite (Figure 2) signifies a marine influence as a result of rising in sea level [30], whereas the association of illite and kaolinite (Figure 2) in the Gercus Formation suggests a greater influence of terrigenous input [3]. The observed vertical change in clay minerals distribution, from abundance of chlorite and smectite in the Paleocene Kolosh Formation to illite and kaolinite dominance in the Eocene Gercus Formation, may relate to global warming in the Eocene.
Conclusions
Clay mineral distributions in the Paleocene- Eocene from north Iraq revealed the dominance of smectite and chlorite with lower amounts of illite in the claystones of the Paleocene Kolosh Formation, which refers to marine conditions as the main paleoenvironmental factor affecting such dominance. Post-depositional diagenetic reactions affect the transformation of palygorskite from precursors smectite. This mineral association may refer to dominance of warm and wet conditions that serve the preservation of these minerals. Whereas, the association of illite and kaolinite in the Eocene Gercus Formation suggests a greater influence of terrigenous input in humid conditions, such as rivers or coastal environments. This vertical distribution of clay minerals may relate to the global warming recorded in the Eocene.

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