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## **Eastern desert ware : traces of the inhabitants of the eastern desert in Egypt and Sudan during the 4th-6th centuries CE**

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## APPENDIX SEVEN Clay Minerals

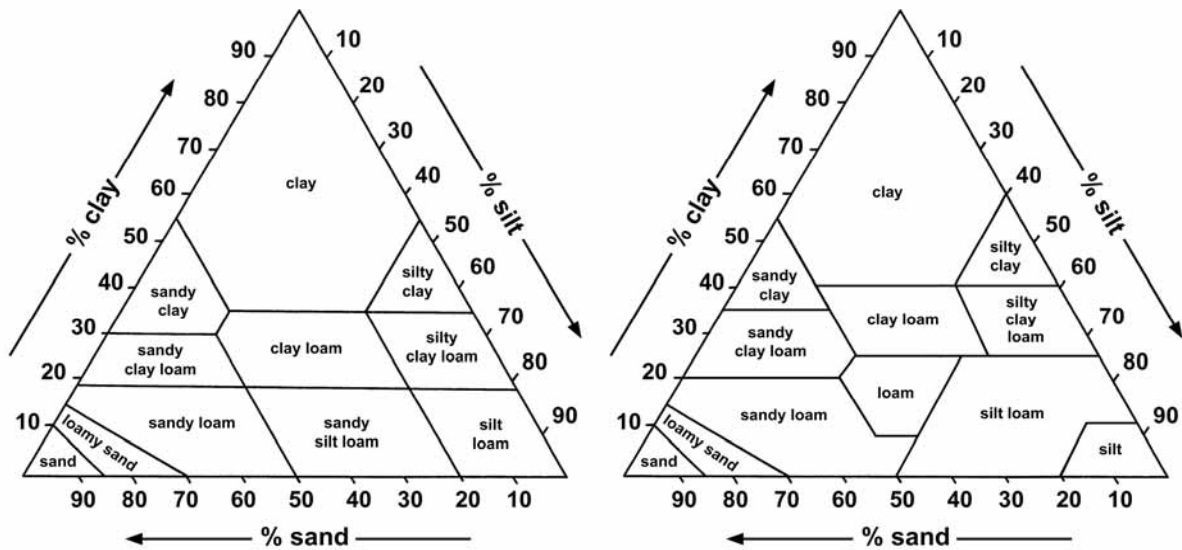


Figure 13-1: Different classification diagrams for soil texture based on composition.

|                  |                  |                  |                  |                  |                  |                  |
|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| Very coarse sand | 1000-2000        | 1000-2000        | 300-2000         | 500-2000         | 600-2000         |                  |
| Coarse sand      | 500-1000         | 500-1000         | 210-300          | 200-500          | 200-600          | 250-2000         |
| Medium sand      | 200-500          | 250-500          | 150-210          | 125-200          | 100-200          |                  |
| Fine sand        | 100-200          | 100-250          | 105-150          | 63-125           | 60-100           | 50-250           |
| Very fine sand   | 50-100           | 50-100           | 50-105           |                  |                  |                  |
| Very coarse silt | 20-50            |                  |                  | 20-63            |                  |                  |
| Coarse silt      | 10-20            | 20-50            | 16-50            | 6-20             | 20-60            | 10-50            |
| Fine silt        | 2-10             | 2-20             | 2-16             | 2-6              | 2-20             | 1-10             |
| Clay             | <2 $\mu\text{m}$ | <2 $\mu\text{m}$ | <2 $\mu\text{m}$ | <2 $\mu\text{m}$ | <2 $\mu\text{m}$ | <1 $\mu\text{m}$ |

Table 13-1: Different classification systems for soil particles based on their size (2000  $\mu\text{m}$  = 2 mm).

Clays are formed during the mechanical and (bio)chemical weathering and low-temperature hydrothermal alteration of rocks. These processes result in a variable mixture of clay, silt and sand particles (Figure 13-1), which are defined by their composition or size (Table 13-1), depending on the scientific context. Clay particles are phyllosilicates (sheet silicates) formed by sheets of tetrahedral silicates ( $\text{SiO}_4^{4-}$ , Figure 13-2), interspersed with varying amounts of  $\text{AlO}_4^{5-}$  tetrahedra; and octahedral sheets formed by oxygen ( $\text{O}^{2-}$ ) and hydroxyl ( $\text{OH}^-$ ) anions surrounding different cations, such as  $\text{Al}^{3+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Fe}^{2+}$  or  $\text{Mg}^{2+}$  (Figure 13-3, Table 13-3). Water is present, in variable amounts, as part of the structure of the minerals (the hydroxyl-groups), attracted to the negative charges on the surfaces of the mineral sheets, and free in the pores between the particles. The tetrahedral and octahedral layers are

assembled in two ways: one octahedral layer can be sandwiched between two tetrahedral layers, referred to as a 2:1 layer (Figure 13-5); or an octahedral layer can be sitting on the apices of a single tetrahedral layer, referred to as a 1:1 layer (Grim 1968; Sudo et al. 1981). Based on the composition and the structure of the minerals, many phyllosilicates can be distinguished within the much larger group of silicate minerals. These phyllosilicates can be divided into four groups: kaolinite (halloysite), smectite (montmorillonite), illite (hydrated mica) and chlorite minerals.

## Appendix VII: Clay Minerals

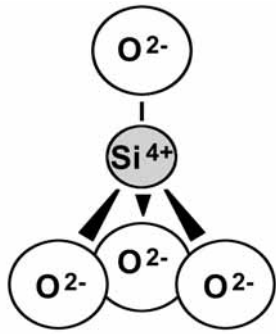


Figure 13-2: Schematic representation of a silicate ion ( $\text{SiO}_4^{4-}$ ), the basis for the silicate minerals, including the phyllosilicates (sheet silicates).

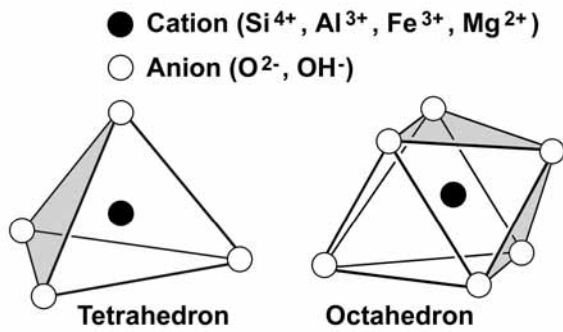


Figure 13-3: Schematic representation of the tetrahedral (left) and octahedral (right) configuration of anions around a relatively small cation.

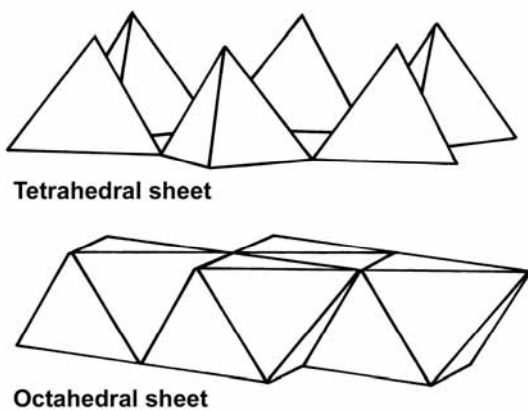


Figure 13-4: Schematic representation of a tetrahedral sheet, with one plane formed by apices and the other by sides (top); and an octahedral sheets (bottom), a they appear in phyllosilicates (sheet silicates).

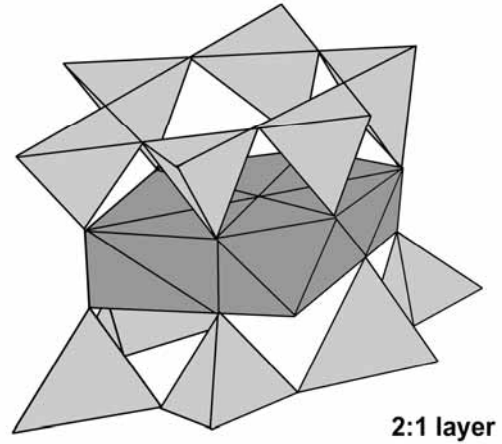
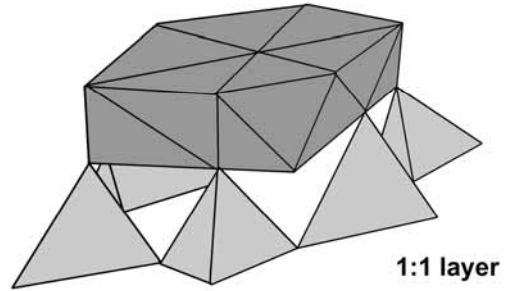


Figure 13-5: Schematic representation of the configuration of atoms in a 1:1 layer, consisting of one tetrahedral and one octahedral sheet (top), and in a 2:1 layer, consisting on one octahedral and two tetrahedral sheets (bottom).

|                   | Ions                                | Charge |
|-------------------|-------------------------------------|--------|
| Octahedral sheet  | 6 $\text{OH}^-$                     | -6     |
|                   | 4 $\text{Al}^{3+}$                  | +12    |
| Contact layer     | 4 $\text{O}^{2-}$ + 2 $\text{OH}^-$ | -10    |
| Tetrahedral sheet | 4 $\text{Si}^{4+}$                  | +16    |
|                   | 6 $\text{O}^{2-}$                   | -12    |
| Total charge      |                                     | 0      |

Table 13-2: The theoretical charge distribution in kaolinite (1:1) clay.

|                 | Tetrahedral sheet     | Octahedral sheet                             |
|-----------------|-----------------------|--|
| Kaolinite       | Si                    | Al   |
| Clinochrysotile | Si                    | Mg   |
| Greenalite      | Si                    | $\text{Fe}^{2+}$ , $\text{Fe}^{3+}$          |
| Amesite         | Al, Si                | Al, Mg                                       |
| Chamosite       | Al, Si                | Al, $\text{Fe}^{2+}$ , $\text{Fe}^{3+}$ , Mg |
| Cronstedtite    | $\text{Fe}^{3+}$ , Si | $\text{Fe}^{2+}$ , $\text{Fe}^{3+}$          |

Table 13-3: General cation composition of different kaolinite minerals.

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The general formula of kaolinite clays is  $(\text{OH})_8\text{Si}_4\text{Al}_4\text{O}_{10}$  (theoretically composed of 47%  $\text{SiO}_2$ , 39%  $\text{Al}_2\text{O}_3$  and 14%  $\text{H}_2\text{O}$ ). Kaolinite clays are 1:1 clays, composed of a single silica tetrahedral sheet and a single aluminium octahedral sheet. In the layer common to the tetrahedral and octahedral sheets, two-thirds of the atoms are shared. These shared atoms are  $-\text{O}$  instead of  $-\text{OH}$ . The charge distribution in kaolinite clay is balanced, leaving no net charge (Table 13-2). Substitution of the silica ( $\text{Si}^{4+}$ ) and aluminium ( $\text{Al}^{3+}$ ) cations by other cations results in different clay minerals within the kaolinite group (Table 13-3). As some of these cations will have a smaller positive charge, such as  $\text{Mg}^{2+}$  or  $\text{Fe}^{2+}$ , this will result in a net negative charge of the layer. This will be partly compensated by other substitutions in the same layer, but mostly by small cations, such as  $\text{Na}^+$ ,  $\text{K}^+$  or  $\text{Ca}^{2+}$ , in the interlayer (the space between the layers). These cations and their interaction with the mineral layers as well as free water molecules determine the macroscopic behaviour of the clay. As cation substitutions are rare in kaolinite clays, they have a low cation exchange capacity (the ability to exchange different cations in the interlayer) as well as a low shrink-swell capacity (the ability to change the volume of the interlayer by absorbing or releasing water).

Halloysite clay has the same general formula as kaolinite clay, but with additional structural  $\text{H}_2\text{O}$  molecules. These can irreversibly get lost at low temperatures. Pure halloysite clay naturally occurs as small cylinders (0.03 x 5  $\mu\text{m}$ ); because of this it has found applications in nano-technology. Kaolinite clays are widely used for ceramics (porcelain), but also to produce the gloss on paper and as the active ingredient in anti-diarrhoea medication. The best known variant of clinochrysolite minerals is asbestos (Table 13-3), which does not have the macroscopic properties usually associated with clays.

The general formula of smectite clays is  $(\text{OH})_4\text{Si}_8\text{Al}_4\text{O}\cdot n\text{H}_2\text{O}$ , theoretically composed of 67%  $\text{SiO}_2$ , 28%  $\text{Al}_2\text{O}_3$  and 5%  $\text{H}_2\text{O}$  (without the interlayer). Smectite is a 2:1 clay, composed of a single aluminium octahedral sheet sandwiched between two silica tetrahedral sheets, which naturally occurs in extremely small particles. Like in kaolinite clay, the charge distribution in smectite clays is theoretically balanced (Table 13-4), although smectite clays never occur in this uncharged state.

In the tetrahedral sheets of smectite clays up to 15% of the  $\text{Si}^{4+}$  ions are replaced by  $\text{Al}^{3+}$ , leaving the mineral with a net negative charge. Substitution of cations in the octahedral sheet can vary from few to complete. The replacement of aluminium by magnesium ( $\text{Mg}^{2+}$ ) results in steatite (soapstone, talc), by iron ( $\text{Fe}^{3+}$ ) in nontronite, by chromium ( $\text{Cr}^{2+}$ ) in volkonskoite, and by zinc ( $\text{Zn}^{2+}$ ) in sauconite. The negative charge of the clay minerals is compensated by small cations in the interlayer. These

will in turn attract water, resulting in the high shrink-swell and cation exchange capacities of these 'expansive' clays. In montmorillonite clay some of the aluminium in the octahedral sheet has been replaced by magnesium and the resulting negative charge is compensated by sodium ( $\text{Na}^+$ ) and calcium ( $\text{Ca}^{2+}$ ) ions in the interlayer. This specific combination results in a very high shrink-swell capacity. Smectite clays are mixed with other clays to change their ceramic properties, but are also widely used for soil improvement, for their capacity to absorb water and salts, and for thermal insulation.

|                   | Ions                                  | Charge       |
|-------------------|---------------------------------------|--------------|
| Interlayer        | $\text{H}_2\text{O} + \text{cations}$ | + (variable) |
| Tetrahedral sheet | $6 \text{O}^{2-}$                     | -12          |
|                   | $4 \text{Si}^{4+}$                    | +16          |
| Contact layer     | $4 \text{O}^{2-} + 2 \text{OH}^-$     | -10          |
| Octahedral sheet  | $4 \text{Al}^{3+}$                    | +12          |
|                   | $4 \text{O}^{2-} + 2 \text{OH}^-$     | -10          |
| Tetrahedral sheet | $4 \text{Si}^{4+}$                    | +16          |
|                   | $6 \text{O}^{2-}$                     | -12          |
| Interlayer        | $\text{H}_2\text{O} + \text{cations}$ | + (variable) |
| Total charge      |                                       | 0            |

Table 13-4: The theoretical charge distribution in smectite clays.

The basic structure of illite clays is similar to the structure of the smectite clays, with a single octahedral layer sandwiched between two tetrahedral layers. Some silica ions are replaced by aluminium ions and the charge difference is compensated by potassium ions ( $\text{K}^+$ ) in the interlayer (Table 13-5). The resulting general formula of the di-octahedral illite minerals (such as muscovite), in which only two-thirds of the possible positions in the octahedral sheet are taken, is  $(\text{OH})_4\text{K}_2(\text{Si}_6\text{Al}_2)\text{Al}_4\text{O}_{20}$  (theoretically composed of 12%  $\text{K}_2\text{O}$ , 45%  $\text{SiO}_2$ , 38%  $\text{Al}_2\text{O}_3$  and 5%  $\text{H}_2\text{O}$ ). In tri-octahedral illite minerals, such as biotite, all the possible positions in the octahedral sheet are taken, mostly by iron and magnesium in highly variable relative abundances.

Well-crystallized illite minerals, such as muscovite, margarite, biotite and clintonite, are known as micas. These do not behave as clays, but rather resemble rock or glass. The incomplete substitution of silica by aluminium and the incorporation of cations other than potassium (especially  $\text{H}_3\text{O}^+$ ) will interfere with the crystallization process and may result in non-expanding illite clays with the general formula  $(\text{K}, \text{H}_3\text{O})(\text{Al}, \text{Mg}, \text{Fe})_2(\text{Si}, \text{Al})_4\text{O}_{10}\cdot n[(\text{OH})_2, (\text{H}_2\text{O})]$ .

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|                   | Ions  | Charge |
|-------------------|---|--------|
| Interlayer        | H <sub>2</sub> O + K <sup>+</sup>                                 | + 1    |
| Tetrahedral sheet | 6 O <sup>2-</sup>   | -12    |
|                   | 3 Si <sup>4+</sup> + Al <sup>3+</sup>                             | +15    |
| Contact layer     | 4 O <sup>2-</sup> + 2 OH <sup>-</sup>                             | -10    |
| Octahedral sheet  | 4 Al <sup>3+</sup> or<br>6 (Fe <sup>2+</sup> , Mg <sup>2+</sup> ) | +12    |
| Contact layer     | 4 O <sup>2-</sup> + 2 OH <sup>-</sup>                             | -10    |
| Tetrahedral sheet | 3 Si <sup>4+</sup> + Al <sup>3+</sup>                             | +15    |
|                   | 6 O <sup>2-</sup>   | -12    |
| Interlayer        | H <sub>2</sub> O + K <sup>+</sup>                                 | + 1    |
| Total charge      |   | 0      |

Table 13-5: The theoretical charge distribution in illite minerals.

|                    | Ions   | Charge |
|--------------------|--|--------|
| Brucite-like layer | 6 OH <sup>-</sup>  | -6     |
|                    | 4 Al <sup>3+</sup> + 2 Mg <sup>2+</sup>  | +16    |
|                    | 6 OH <sup>-</sup>  | -6     |
| Tetrahedral sheet  | 6 O <sup>2-</sup>  | -12    |
|                    | 2 Si <sup>4+</sup> + 2Al <sup>3+</sup>   | +14    |
| Contact layer      | 4 O <sup>2-</sup> + 2 OH <sup>-</sup>  | -10    |
| Octahedral sheet   | 6 (Fe <sup>2+</sup> , Mg <sup>2+</sup> , Mn <sup>2+</sup> ,<br>Cr <sup>3+</sup> , Fe <sup>3+</sup> or Ti <sup>3+</sup> ) | +12    |
| Contact layer      | 4 O <sup>2-</sup> + 2 OH <sup>-</sup>  | -10    |
| Tetrahedral sheet  | 2 Si <sup>4+</sup> + 2Al <sup>3+</sup>   | +14    |
|                    | 6 O <sup>2-</sup>  | -12    |
| Total charge       |  | 0      |

Table 13-6: The theoretical charge distribution in chlorite minerals.

|                | Cations        | Fixed part  |
|----------------|----------------|---|
| Baileychlore   | Al, Fe, Mg, Zn | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Chamosite      | Fe, Mg         | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Clinochlore    | Fe, Mg         | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Cookeite       | Al, Li         | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Nimite         | Al, Mg, Ni     | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Orthochamosite | Fe, Mg         | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Pennantite     | Al, Mn         | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Ripidolite     | Al, Fe, Mg     | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |
| Sudoite        | Al, Fe, Mg     | (Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub> |

Table 13-7: General cation composition of different chlorite minerals.

The chlorite minerals are a large and diverse group that is sometimes given a separate place within the group of silicate minerals. The basic structure of all chlorite minerals comprises a series of tri-octahedral illite layers, referred to as mica-like or talc layers, alternated with brucite-like layers. These are octahedral sheets of hydroxyl-groups surrounding magnesium and aluminium ions (Figure 13-4), with the general formula (Mg·Al)<sub>6</sub>(OH)<sub>12</sub>. Brucite or Mg(OH)<sub>2</sub> is a pearly white to pale green mineral that is one of the alteration products of marble, limestone and schist, and which is commonly found in association with serpentine, calcite and talc. The brucite-like layer is unbalanced as a result of the substitution of Mg<sup>2+</sup> by Al<sup>3+</sup>. This allows it to compensate for the negative charge that is the result of the substitution of Si<sup>4+</sup> by Al<sup>3+</sup> in the mica-like layer (Table 13-6). Substitution of different cations, in different proportions, explains the large number of known chlorite minerals (Table 13-7).

Natural clays are usually a mix of the minerals discussed above, although layers of pure clay minerals, especially kaolinite clay, do occur. The relative abundance of the various minerals determines the characteristics of the raw and fired clay. Potter's clay is usually a mixture of different naturally occurring clays with other geological or organic inclusions, such as quartz, volcanic ash, dung, straw, shell, silt or ground pottery (grog). The larger inclusions in the clay, either naturally present or introduced, can be studied by (petrographic) microscopy (Appendix 8). The clay minerals themselves can only be visualized by electron microscopy or X-ray diffraction.