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Evaporite mineralogy and major element geochemistry as tools for palaeoclimatic investigations in arid regions: A synthesis

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Abstract

This paper presents a synthesis of the applications of evaporite mineralogy and the relationship between major elements for the palaeoclimatological research of arid regions, with examples from Playa Oum el Krialate in Tunisia, Wadi Natron in Egypt, East African Rift Valley, etc. The numerous evaporite minerals serving as indicators of palaeo-drylands (salinity and evaporation) include carbonates, sulfates, and Na, K, Ca, and Mg chlorides. The occurrence of double salts, such as glauberite, carnallite, kainite, gaylussite, pirssonite, burkeite, etc., suggests disequilibrium conditions. Apart from that, the presence of very rare Fe-sulfates, such as rozenite and szomolnokite, indicates anoxic conditions with higher salinity. The formation of Na-silicates, such as magadiite and kenyaite, implies a decrease in pH of a highly alkaline Na concentrated brine. The Mg-silicates (palygorskite, Mg-montmorillonite and talc) form quickly and then re-dissolve when conditions change. Identification of fulgurites in the Sahara has been related to palaeo-lightning. We have also discussed a simple geochemical approach of using the ratios of soluble/insoluble elements to identify palaeo-arid events with examples from loess-soil sequences from Feiran Oasis in the Sinai Desert (Egypt) and salty silt lacustrine sequences from Thar Desert (India).

Keywords: Tropical deserts, evaporite minerals, geochemistry, synthesis.

Resumen

En este trabajo se presenta una síntesis del uso de minerales evaporíticos y relaciones entre elementos mayores en investigaciones paleo-ambientales de zonas áridas, con ejemplos de la sabkha Oum el Krialate en Túnez, Wadi Natron en Egipto, Valle de Rift del África oriental, etc. Los minerales evaporíticos indicadores de eventos secos (salinidad y evaporación) son carbonatos, sulfatos y cloruros de Na, K, Ca y Mg. La ocurrencia de sales dobles, como glauberita, carnalita, kainita, gaylusita, pirssonita, burkeita, etc., sugiere condiciones de desequilibrio. Además, la presencia de sulfatos de Fe poco comunes, como rozenita y szomolnokita, indica condiciones de anoxia con alta salinidad. La precipitación de silicatos de Na, como magadiita y kenyaita, implica la dilución de una salmuera muy alcalina y con alta concentración de Na. Los silicatos de Mg (paligorskita, Mg-montmorillonita y talco) precipitan rápidamente para disolverse nuevamente al cambiar las condiciones. La presencia de fulguritas en el Sáhara indica la incidencia de paleo-relámpagos. Asimismo, para identificar eventos de paleo-sequía, se discute un método sencillo basado en relaciones de elementos solubles e insolubles con ejemplos de secuencias de suelo tipo loess del Oasis de Feirán (Desierto de Sinaí, Egipto) y de secuencias de sedimentos lacustres del Desierto de Thar (India).

Palabras clave: Desiertos tropicales, minerales evaporíticos, geoquímica, síntesis.

1. Introduction

The study of (palaeo-) climatic registers is highly effective in reconstructing the past climate and other conditions present at the time of sediment formation or deposition. The knowledge of old climates and the mechanisms of climatic changes may help to identify the regularities of palaeoclimatology and could possibly lead to improve present-day agriculture and horticulture as well as the management of water, soil, and natural resources. While marine sediments on continents demand a transgressive sea, the sediments in lagoonal or even in true sabkha environments require a topographically closed basin and fluvial freshwater input. After some time, these lakes become saline in (semi-) arid environments and finally evaporate in pans, accompanied by sedimentation caused by aeolian activity and erosion, *e.g.* dune and loess formation, grain sorting, and formation of fluvial and lake terraces. The criteria used to recognize these geomorphological formations and processes in old sediments and soils have been treated in hundreds of geomorphological and geological articles and textbooks (*e.g.*, Glennie, 1970; Holser, 1979; Büdel, 1982; Goudie and Pye, 1983; Lowe and Walker, 1984; Gasse *et al*., 1987; Thomas, 1989; Summerfield, 1991; Thomas and Shaw, 1991; Cooke *et al*., 1993; Kröpelin, 1993; Goudie and Wells, 1995; Lancaster, 1995; Williams and Balling, 1996; Dittmann, 1999; Glennie and Singhvi, 2002).

Diagenetic processes change the association of primary minerals (at least partly). Evaporite minerals (salts) are dissolved and transported in solution until more stable products precipitate. Physical and chemical conditions determine the transformation of salts (Braitsch, 1971; Busson and Perthuisot, 1977; Eugster and Hardie, 1978; Holser, 1979; Usdowski and Dietzel, 1998). But quite often, the development from primary limnic formations to secondary transformed products can be traced back using remnants or relict structures (Wiedemann and Smykatz-Kloss, 1981). Sophisticated physical methods may answer some questions, *e.g.* the determination of isotopic ratios of C, S, O, B, Sr, or H (see Hoefs, 1987; Heine, 1998; von Grafenstein *et al*., 1998; Eitel, 2000), including age determinations by ^{14}C methods (Geyh and Jäkel, 1974; Yadav, 1997; Geyh, 2005). Even in case of lack or absence of organic material, electron spin resonance (ESR) (Dhir *et al*., 2004), thermoluminescence (TL) or optically stimulated luminescence (OSL) methods may help (Rögner *et al*., 1999; Eitel *et al*., 2004), provided that the tolerance of the method is suitable to solve a certain problem (Zöller, 1995; Jäkel, 2004). Considering the peculiarities in formation and dissolution behaviour of salt or clay minerals, a certain reconstruction of a primary limnic environment is possible. Both the degree of interaction between sediments (or soils) and water and the development of secondary products are dependent on several environmental conditions, namely temperature, pH and Eh values, pressure, activity of water and ions, soil and sediment porosity and packing density, solubility of species and interfering (disturbing) compounds, equilibrium or disequilibrium conditions, and time.

2. Preliminary remarks on geomorphology, geology, biology, physics, and regional distribution

2.1. Geomorphological criteria

The formation of terraces by lake and sea level fluctuations is a widely observed geomorphological phenomenon (Büdel, 1982; Rust, 1989; Vogel, 1989; Heine, 1990). Similarly, palaeo-dunes are indicators of wind directions or variations in past wind currents (Bowler, 1973; Sarnthein, 1978; Lancaster, 1981; Wasson *et al*., 1983, 1984; Gaylord, 1990; Kar, 1990; Pye and Tsoar, 1990; Felix-Henningsen, 2000, 2004; Grunert *et al*., 2000; Grunert and Lehmkuhl, 2004; Heine, 2004). Lake and sea level fluctuations mirror past changes in precipitation as well as marine transgressions and regressions due to climatic and tectonic activities (Faure and Elouard, 1967; Nir, 1974; Perthuisot, 1976; Gasse *et al*., 1987; Tooley and Shennan, 1987; Koessl, 1988; Lancaster, 1989; Baumhauer, 1990; Baumhauer *et al*., 2004; Fang, 1991; Wenigwieser, 1992; Qin and Yu, 1998; Wünnemann *et al*., 1998; Eitel *et al*., 2004; Sinha and Raymahashay, 2004; Sinha *et al*., 2006; Roy *et al*., 2006, 2008a, 2009; Achyuthan *et al*., 2007). Increasing aridity leads to salt evaporation in pans and playas (Lancaster, 1981; Petit-Maire, 1986; Goudie and Wells, 1995; Eitel and Blümel, 1997; Schütt, 1998, 2004; Smykatz-Kloss *et al*., 1999/2000) and enables loess and silt accumulation (Sirocko and Sarnthein, 1989; Vogel, 1989; Derbyshire *et al*., 1995; Gallet *et al*., 1996; Smykatz-Kloss *et al*., 1998; Brunotte and Sander, 2000; Grunert and Lehmkuhl, 2004; Singhvi and Kar, 2004). Special conditions are required for the formation of fulgurites as observed by Sponholz *et al*. (1993) in the Sahara. The presence of fulgurites demonstrates the interaction of lightning and lake surfaces.

2.2. Geological and biological criteria

Climatic changes are also mirrored in geological, sedimentological, and biological events (Tricart and Cailleux, 1972; Carbonel *et al*., 1988; Sirocko and Sarnthein, 1989; Vogel, 1989; Geyh and Eitel, 1998; Smykatz-Kloss *et al*., 1998; Heine and Heine, 2002; Jain and Tandon, 2003; Mischke *et al*., 2004). The organic carbon content on the continental slope of the Arabian Sea (Indian Ocean) tells of palaeoclimatic changes and monsoon development in the adjacent continents (Berner and Lasaga, 1989; Sirocko, 1995). Sea level fluctuations show global climatic changes as well (Faure and Elouard, 1967; Einsele *et al*., 1974). Geophysical methods also contribute (Schumm, 1977; Prell and Kutzbach, 1987) and astronomic theories, like Milankovitch's, have led to the

explanation of climatic changes (Berger and Tricot, 1986; Blum and Törnquist, 2000).

Biological aspects may play an important role in reconstructing palaeontologic developments (Maley, 1983; Gasse *et al*., 1987; Schulz, 1987). The development of ostracod and diatom species, in particular, has been used for palaeoenvironment reconstructions (Carbonel *et al*., 1988; Caballero, 1997; Mezquita *et al*., 1999; Palacios Fest *et al*., 2002; Mischke *et al*., 2004; Caballero *et al*., 2005).

2.3. Physical and chemical criteria

The influence of ice and ice transport on rocky grounds and soils may be mentioned briefly. This includes all physical processes, such as freezing and thawing, and those processes related to glaciers, wind, and thunderstorms, as well as the formation of (peri-) glacial and structured soils etc. (Fink and Kukla, 1977; Washburn, 1979; Büdel, 1982; Harris, 1986; Tyson, 1986). Roberts and Spencer (1995) and Spencer *et al*. (2003) used fluid inclusions in halite (evaporitic crusts in Death Valley, California) for the measurement of palaeotemperature. When available, the exact methods of physical dating and isotope geochemistry are very helpful in fixing palaeoclimatic events and changes (Geyh and Jäkel, 1974; Delibrias *et al*., 1976; Gasse *et al*., 1987; Heine, 1990; Yadav, 1997; Geyh and Eitel, 1998; Hofmann and Geyh, 1998; von Grafenstein *et al*., 1998; Dutkiewicz *et al*., 2000; Glennie and Singhvi, 2002; Juyal *et al*., 2003). Isotopic ratios may characterize palaeoenvironments (Hoefs, 1987; Tyson, 1986; Sharp, 2007). The concentration of total organic carbon (TOC) represents the amount of organic matter preserved after sedimentation, which depends upon initial production and the degree of degradation (Meyers and Teranes, 2001; Leng *et al*., 2005). The source of organic matter can be differentiated by the ratio of organic carbon and total nitrogen (C/N) and $\delta^{13}C$. Organic matter derived from lacustrine phytoplankton has a lower C/N (<10) value compared to organic matter derived from terrestrial plants (>10) (Prasad *et al*., 1997). $\delta^{13}C$ of organic matter indicates the source of HCO_3^- and dissolved CO_2 (*i.e*. derived from C4 or C3 plants by ground water inflow, organic matter deposited as methane in the lake bottom, catchment carbonate deposits, and atmospheric CO_2). Similarly, $\delta^{18}O$ in inorganic material (carbonates and silicates) indicates palaeo-temperature during formation of the mineral phases (Leng *et al*., 2005). In water samples, deviation of both $\delta^{18}O$ and δD from the Global Meteroric Water Line suggests kinetic fractionation due to evaporation. In warmer and tropical water, both isotopes have higher δ values compared to colder and polar rainfall (Leng and Marshall, 2004).

Organic material is also used for ^{14}C measurements (Geyh, 2005) to reconstruct the chronology of the identified events. In the absence of organic material, which is the case in many Quaternary and even Tertiary arid environments, luminescence (TL, OSL) (Zöller, 1995; Rögner *et al*., 1999) or ESR techniques (Dhir *et al*., 2004) may help.

2.4. Regional distribution of drylands

Pans, playas and palaeolakes of the great dryland areas have been the objects of interests in palaeoclimatic studies. In these regions, enrichment of (earth) alkalies, iron and silica are reported. Iron enrichment is documented in (semi-) aridic ferricretes of former swamps (Nahon, 1976; Felix-Henningsen, 2000, 2004). Silica enrichment in highly alkaline lakes is reported for one of the two main silcrete formations (*i.e*. allochthonous silcrete, while the autochthonous type represents a soil formation) by Summerfield (1983), Young (1985), Joachim (1988) and Thomas and Shaw (1991). Salt enrichment is documented by Derbyshire *et al*. (1995), Mehrshabi *et al*. (2003), and Schütt (2004). Development of soils, pans, and desertic landscape of the world's largest desert, the Sahara, is reported by Petit-Maire (1986, 1987, 1991) and Baumhauer (1990). Guo *et al*. (2000) have compared the Sahara with east Asian deserts, including the Gobi and other large deserts in China, Tibet, and Mongolia (Kukla, 1987; Fang, 1991; Hofmann, 1993; Derbyshire *et al*., 1995; Jäkel, 1995; Lehmkuhl, 1995; Liu and Fu, 1996; Qin and Yu, 1998; Wünnemann *et al*., 1998; Grunert *et al*., 2000; Grunert and Lehmkuhl, 2004). While the deserts of Iran (Mehrshabi *et al*., 2003) and Arabia (Glennie and Singhvi, 2002; Barth, 2003) are rarely studied, the Indian Desert (Thar) located in the northwestern Indian state of Rajasthan has received significantly more attention from geoscientists (Wasson *et al*., 1983, 1984; Kar, 1990; Jain and Tandon, 2003; Juyal *et al*., 2003; Singhvi and Kar, 2004; Sinha *et al*., 2006; Roy *et al*., 2001, 2006, 2008b, 2009). This is also true for the world's oldest desert, the Namib, in South Africa (Rust, 1989; Vogel, 1989; Heine, 1998, 2004; Eitel, 2000; Eitel *et al*., 2004). Relative to the Namib, a relatively small desert, Sinai in Egypt, has received some attention by Nir (1974), Rögner and Smykatz-Kloss (1991), Rögner *et al*. (1999, 2004), and Smykatz-Kloss *et al*. (1998, 1999/2000, 2004). American deserts have been studied by Fahey and Mrose (1962), Bradley and Eugster (1969), Gaylord (1990), Monger and Daugherty (1991), Bischoff *et al*. (1997), Metcalfe *et al*. (2002), and Roy *et al*. (2010). The Australian desert has been studied by Young (1985), Holland *et al*. (1988), Nanson and Tooth (1989), and English *et al*. (2001).

3. Mineralogical and geochemical criteria and examples

3.1. Minerals as criteria

Among the common cations, the solution and precipitation behaviour of Na^+ and Mg^{2+} (and Fe^{2+} and Mn^{2+} in Eh-negative environments) are very suitable (Holser, 1979, Mason and Moore, 1982). Sodium is generally highly soluble (abundant in water bodies of arid environments)

and dissolves even when water activity is low. Suitable environments for preservation of Na^+ are (semi-) deserts, *e.g*. the drylands of Northern Africa as Wadi Natron in Egypt (Wenigwieser, 1992), Playa Oum el Krialate in Tunisia (Perthuisot, 1976; Koessl, 1988), and similar environments in Libya, Morocco, and Mauritania. In these environments, besides halite (NaCl), Na^+ precipitates as the highly soluble mirabilite (Na_2SO_4•$10H_2O$), but only in contact with water (*e.g*. in pools, see Smykatz-Kloss *et al*., 1992). After its formation, mirabilite transforms to thenardite (Na_2SO_4) on contact with air. In the limnic sediments of Wadi Natron, idiomorphic thenardite occurs in crystals with diameters up to a few centimetres.

Among the places where a large number of disequilibrium products occur in a short period, the Playa Oum el Krialate is one of the most favourable and well-studied localities. A short period spans a few weeks up to a few months, until the next rain occurs and dissolves the Na-salts. But during dry periods, *i.e*. times of low water activity, a number of semi-finished products ("precursors") are observed, even locally as main constituents, many of which contain either very little water or no water at all. These are in the process of becoming thermodynamically reasonable and stable end products and minerals such as mirabilite (Na_2SO_4•$10H_2O$), hexahydrite ($MgSO_4$•$6H_2O$), pentahydrite ($MgSO_4$•$5H_2O$), leonhardtite ($MgSO_4$•H_2O), nahcolite ($NaHCO_3$), trona ($Na_3(CO_3)(HCO_3)$•$2H_2O$), pirssonite ($Na_2Ca(CO_3)_2$•$2H_2O$), gaylussite ($Na_2Ca(CO_3)_2$•$5H_2O$), or burkeite ($Na_6(CO_3)(SO_4)_2$) (see Table 1).

Among these evaporites that partially occur as main constituents, some are very rare compounds and occur as traces, *i.e*. glauberite, polyhalite, carnallite, bassanite, rozenite and szomolnokite (Spencer *et al*., 2003) (see Table 1). Bassanite has been found in aridic soils by Akopodje (1985) and Smykatz-Kloss *et al*. (1985) on the surface of sunny walls in South Tunisia. Very recently, this mineral has been observed as a main component on the ground of the "White Desert" in Egypt, where it is mainly covered by a few centimetres of wind-blown carbonates and anhydrite. This cover of wind-blown weathering products is the reason for the occurrence and longer persistence of bassanite.

The very rare and instable Fe^{2+}-sulfates rozenite and szomolnikite ($FeSO_4$•$4H_2O$ and $FeSO_4$•H_2O, respectively) are found on the way from the gypsum karst, east of Tatahouine, to the Oum el Krialate playa, associated with organic material (Koessl, 1988; Smykatz-Kloss *et al*., 1992). The preservation of organic material is the reason for the negative Eh environment, which is necessary for forming Fe^{2+}-sulfates. However, the occurrence of bassanite, rozenite, and szomolnokite in the deserts of Egypt and Tunisia is exceptional among sulfate evaporites, as their occurrence requires some shelter from the (rarely abundant) rain events.

Most of the other evaporites are double salts (*i.e*. combinations of Na, Mg, Ca, and K; see Table 1). These occur only for short periods and in traces; such is the case for glauberite, kainite, carnallite, polyhalite (Smykatz-Kloss *et al*., 1992), burkeite, trona, pirssonite, and gaylussite (Fahey and Mrose, 1962; Bradley and Eugster, 1969; Holser, 1979). Trona has been a known mineral to Humanity for millennia, conspicuous in the sediments of the Wadi Natron in Egypt (Wenigwieser, 1992).

There is another special tropical environment for the preservation of Na-minerals, co-precipitated with silica, although only when very high amounts of Na and Si in the brine. Such is the case along the East African Rift Valley (East African Graben), *e.g*. in Kenya and Tanzania, and especially where high temperature and weathering rates of abundant sodic volcanics (Na-carbonatites) feed the playa lakes, such as Lake Magadi and other highly alkaline lakes (pH ~ 12) in the grabens. The alkali (Na) silicates precipitate from oversaturated brines diluted by rain water. The product is magadiite ($NaSi_7O_{13}(OH)_3$•$3H_2O$; see Eugster, 1969), that forms only in highly alkaline environments (*e.g*. Lake Magadi, with pH = 12 and concentrations of Na >100000

Table 1 Occurrence of sulfates, carbonates, and chlorides in the Playa Oum el Krialate (after Smykatz-Kloss *et al*., 1992).

Mineral	Chemical Composition	Occurrence
Gypsum	$CaSO_4 \cdot 2H_2O$	+++
Bassanite	$CaSO_4 \cdot ½H_2O$	+
Anhydrite	$CaSO_4$	++
Glauberite	$Na_2Ca(SO_4)$	+
Polyhalite	$K_2MgCa_2(SO_4)_4 \cdot 4H_2O$	+
Syngenite	$K_2Ca(SO_4)_2 \cdot H_2O$	/
Mirabilite	$Na_2SO_4 \cdot 10H_2O$	++
Thenardite	Na_2SO_4	++
Hexahydrite	$MgSO_4 \cdot 6H_2O$	++
Kieserite	$MgSO_4 \cdot H_2O$	+
Bloedite	$Na_2Mg(SO_4)_2 \cdot 4H_2O$	++
Loeweite	$Na_{12}Mg_7(SO_4)_{13} \cdot 15H_2O$	/
Picromerite	$K_2Mg(SO_4)_2 \cdot 6H_2O$	/
Langbeinite	$K_2Mg_2(SO_4)_2$	/
Kainite	$K_4Mg_4Cl_4(SO_4)_4 \cdot 11H_2O$	/
Rozenite	$FeSO_4 \cdot 4H_2O$	+
Szomolnokite	$FeSO_4 \cdot H_2O$	+
Calcite	$CaCO_3$	++
Aragonite	$CaCO_3$	+
Dolomite	$CaMg(CO_3)_2$	++
Magnesite	$MgCO_3$	+
Vaterite	$CaCO_3$	/
Nahcolite	$NaHCO_3$	+
Trona	$Na_3H(CO_3)_2 \cdot 2H_2O$	+
Halite	NaCl	+++
Sylvite	KCl	+
Carnallite	$KMgCl_3 \cdot 6H_2O$	+

+++ = main salt mineral; + + = frequent; + = abundant; / = questionable

ppm). As pH decreases (*i.e*. by dilution with rain water), magadiite transforms to kenyaite ($NaSi_{11}O_{26.5}(OH)_4 \bullet 3H_2O$) and finally to chalcedony (SiO_2) (Eugster, 1969). The formation of magadiite is also reported from a few other (similar) localities in California.

The other highly soluble cation, Mg^{2+}, forms evaporites in very special (semi-) aridic environments, and such Mg-bearing evaporites are somewhat more stable than the Na-bearing ones. Mg^{2+} compounds occur periodically (*e.g*., during evaporation in Mg-containing playa lakes) in recent playa lakes in Spain (de la Peña *et al*., 1982; Ordóñez *et al*., 1994) and Tunisia (Koessl, 1988) as Mg sulfates of the kieserite-epsomite series (Table 1), and form very soluble intermediate products such as hexahydrite ($MgSO_4 \bullet 6H_2O$), pentahydrite ($MgSO_4 \bullet 5H_2O$) or leonhardtite ($MgSO_4 \bullet 4H_2O$). These minerals are found as efflorescences on dolomitic limestones in Libya as well, together with bloedite in half caves (with stalactites and flowstones) of the Jabal Nafusah (Tripolitania; Smykatz-Kloss *et al*., 1992). The next rain dissolves these products rather quickly, unless they are covered with sediments or other authigenic formations.

There are a few more persisting evaporates of this soluble cation. There are Mg-silicates that form even in contact with water, in playa lakes and sabkha environments. This includes the occurrence of palygorskite ($(Mg,Al)_2(OH/SiO_{10}) \bullet 4H_2O$) (Millot, 1964; Yaalon and Wieder, 1976; Singer, 1979, 1984; Monger and Daugherty, 1991; Eitel, 1994), Mg-montmorillonite, (Mg_3-$x(OH)_2Si_4O_{10}$ $xAlx \bullet nH_2O$, Na_2-x) (Stengele and Smykatz-Kloss, 1995), or talc ($Mg_3(OH)_2Si_4O_{10}$) in special cases (mainly metasomatically formed). These minerals form more quickly and re-dissolve when the environmental conditions change. Thus, they represent remnants of the former arid history of that special limnic sediment (Eitel, 1994; Eitel and Zöller, 1996).

More common than simple Na- or Mg- minerals is the case of double salts (like bloedite) (Braitsch, 1971; Koessl, 1988) and carbonates. Sedimentary dolomites are reported from playa lakes in Russia, China, India, North Africa (Roy *et al*., 2001, 2006). Meta-stable proto-dolomite (Ca>Mg) typically occurs instead of dolomite in limnic and aridic regions (Smykatz-Kloss and Goebelbecker, 1992), where small amounts of Fe and Mn are incorporated into the dolomite structure (Roy *et al*., 2009).

3.2. Geochemical criteria

The behaviour of highly soluble elements like Na^+, Mg^{2+} (or K^+) is contrary to that of hydrolysates, *i.e*. Ti^{4+}, Al^{3+}, Fe^{3+} (Mason and Moore, 1982), which are resistant to normal weathering solutions. In contact with water, the soluble elements dissolve and are transported away, while the hydrolysates (interacting with solutions) become increasingly enriched in the sediment. Due to its relatively larger ionic radius and partial immobility, the behaviour of the soluble cation K^+ is different. It dissolves (like Na^+) but is adsorbed quickly, forming new compounds (*e.g*. illite) (Reheis, 1990; Pandarinath *et al*., 1999). Thus, Na/K continuously decreases with time. However, the ratios Na/Ti, Na/Al, Na/Fe, Mg/Al, Mg/Ti, and Mg/Fe (the last three, only in carbonate-free environments) strongly decrease with increasing water activity (and vice versa). Nesbitt and Young (1982) were pioneers in the use of such ratios for palaeoclimatic considerations. Other authors continued in characterising palaeoclimatic environments by using some of these relations (Sirocko, 1995; Gallet *et al*., 1996; Smykatz-Kloss *et al*., 1998, 2004; Rögner *et al*., 2004; Schütt, 2004; Roy *et al*., 2006, 2008b, 2009; Sinha *et al*., 2006).

Sponholz *et al*. (1993) observed intermittence between palaeoclimatic events in desert lakes and surrounding geomorphology (*e.g*., the water level of these lakes) when they identified relics of fulgurites (concretions in the Sahara sand produced by lightnings at former lake levels), which enabled them to reconstruct the desert lake level with time. Similarly, Roy and Smykatz-Kloss (2007) studied the REE geochemistry of evaporites and the degree of roundness of rock fragments in the clastic fractions of sediments in order to reconstruct the palaeo-fluvial conditions in the Thar Desert. Fromm *et al*. (2005) used Fe-Mn vein mineralisations and the different stages of calcrete formation as indicators of the development of the Tunisian desert. Roy *et al*. (2009, 2010) used geochemical signatures of chemical weathering and mineralogical composition of playa lake sediments to reconstruct late Holocene hydrological changes at the margins of the Thar Desert (India) and late Pleistocene-Holocene palaeo-environmental conditions at Sonora Desert (Mexico).

Impressive walls (up to 60 m high) of loess-like sediments are found in the Feiran Valley (Sinai, Egypt) (Nir, 1974; Rögner and Smykatz-Kloss, 1991; Rögner *et al*., 1999, 2004; Smykatz-Kloss *et al*., 1999/2000, 2003, 2004). Their origin has been related to fluvial terraces (Büdel, 1982), lake sediments (Nir, 1974), flood products, fluvial-torrential sediments, and alluvial loess (Rögner *et al*., 2004). The sequences consist of 3-20 cm thick loess layers intercalated with 2-8 cm thick polygonal soil layers. Table 2 presents the geochemical composition of 17 different loess-soil pairs found throughout several metres in a profile from the Feiran Oasis (data compiled from Knabe, 2000, and Rögner *et al*., 2004). The palaeoecological information is provided by the simple ideas that (1) loessian material is a product of desertic environment, and (2) subsequent soil formation requires water (humidity).

In each loess layer, the weathering of clastic minerals present in the uppermost part (feldspars, micas and, to a lesser degree, amphiboles and pyroxenes) led to the formation of the overlying soil. During this process, dissolution and transportation (*i.e*. removal) of soluble elements (Na, Mg, and K) occurs in the uppermost part of each loess layer and thus changes the ratios of soluble to (relatively insoluble) hydrolysate (TiO_2, Al_2O_3, Fe_2O_3)

Table 2. Ratios of Na_2O/Al_2O_3, Na_2O/TiO_2, and Na_2O/K_2O in 17 different loess (l) and overlying soil(s) pairs in a profile from Feiran Oasis in Sinai Desert (data compiled from Rögner *et al*., 2004).

Pairs		Na_2O/Al_2O_3	Na_2O/TiO_2	Na_2O/K_2O	Loess - Soil Na_2O/Al_2O_3	Loess - Soil Na_2O/TiO_2	Loess - Soil Na_2O/K_2O
1	s	0.056	0.75	0.47	0	0.1	0
	l	0.056	0.76	0.47			
2	s	0.057	0.8	0.45	0.015	0.16	0.16
	l	0.072	0.96	0.61			
3	s	0.043	71	0.37	0.046	0.33	0.25
	l	0.089	1.04	0.62			
4	s	0.053	0.81	0.41	0.063	0.35	0.33
	l	0.116	1.16	0.74			
5	s	0.061	0.81	0.46	0.029	0.23	0.14
	l	0.09	1.04	0.6			
6	s	0.122	1.47	0.81	0.061	0.99	0.52
	l	0.183	2.46	1.33			
7	s	0.059	0.84	0.5	0.16	1.93	1.04
	l	0.219	2.77	1.54			
8	s	0.081	0.98	0.55	0.062	0.29	0.26
	l	0.143	1.27	0.81			
9	s	0.089	1.16	0.61	0.026	0	0.12
	l	0.116	1.16	0.71			
10	s	0.078	1	0.56	0.019	0.24	0.07
	l	0.097	1.24	0.63			
11	s	70	0.9	0.54	0.042	0.3	0.19
	l	0.112	1.2	0.73			
12	s	0.064	0.83	0.51	0.072	0.42	0.32
	l	0.136	1.25	0.83			
13	s	0.077	1.01	0.6	0.058	0.21	0.21
	l	0.135	1.22	0.81			
14	s	0.062	0.82	0.55	0.059	0.34	0.17
	l	0.121	1.16	0.72			
15	s	0.061	0.82	0.5	0.129	1.38	0.79
	l	0.19	2.2	1.29			
16	s	0.062	0.91	0.5	0.057	0.22	0.22
	l	0.119	1.13	0.72			
17	s	0.082	1	0.6	0.063	0.9	0.36
	l	0.145	1.9	0.96			

elements in the soil. Humid conditions lower these ratios and increasing humidity (or intensive contact of water and loess) is mirrored by an increase in the differences of the mentioned ratios (*e.g*., amount of Na_2O/TiO_2 in loess minus the amount of Na_2O/TiO_2 in the overlying soil). These values are listed in Table 2 and shown in Figure 1.

Figure 1 shows two distinct shifts to very arid conditions (the 7th and 15th loess-soil pairs) and three smaller deviations to (semi-) aridity (the 4th, 12th, and 17th loess-soil pairs). Exact age determinations for these arid events are most desirable. The TL age (Rögner *et al*., 1999) suggests that the studied profile covers 25.7 ± 3.9 ka and more than 25 m of the profile is unexposed.

Five different loess-soil pairs from the Wadi Feiran profile (Table 3) are quite similar to those of the 17 different loess-soil pairs from Feiran Oasis (Figure1) and represent another profile in the same valley. The chemical compositions (Table 3) show that the soils are mostly enriched in Fe_2O_3 and Al_2O_3 compared to the underlying loess. The MgO contents seem to be quite constant throughout the profile. Based on the difference in the ratio between soluble and insoluble elements, two distinct aridic events were identified (Figure 2), *i.e*. the 2nd, 3rd, and

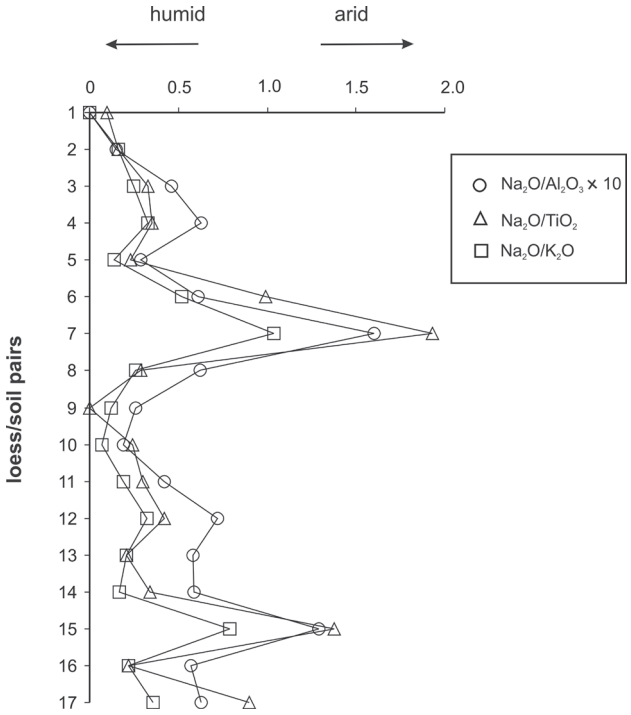

Figure 1. Difference between the relations of Na_2O/Al_2O_3, Na_2O/TiO_2 and Na_2O/K_2O in alluvial loess and overlying soil in 17 different loess-soil pairs in a profile from Feiran Oasis in Sinai.

Table 3a. Geochemical compositions of major elements (mass %) of 5 different loess (l) - soil(s) pairs in the Wadi Feiran profile in Sinai Desert (data compiled from Smykatz-Kloss *et al*., 1998). Pair 1 represents top of the profile and pair 5 is from the bottom part.

Pairs		Thickness (cm)	Na_2O	Al_2O_3	Fe_2O_3	MgO
1	s	2	0.7	10.6	5.8	3
	l	15	0.6	9.2	5.1	2.6
2	s	4	0.6	9.5	5.7	2.6
	l	13	0.7	6.1	3.4	1.8
3	s	4	0.6	9.3	5	2.5
	l	9	1.4	9.2	5.7	2.6
4	s	2	0.6	11.5	5.5	2.7
	l	10	0.7	8	4.8	2.3
5	s	4	0.7	8.3	4.8	2.3
	l	8	1	9.1	4.6	2.4

Table 3b. Values of $Na_2O/Al_2O_3 \times 10$, Na_2O/Fe_2O_3, and MgO/Fe_2O_3 in the loess (l) and soil (s) pairs and Na_2O/Al_2O_3 and Na_2O/Fe_2O_3 ratios in loess-soils of each pair in the Wadi Feiran profile.

Pairs		Na_2O/Al_2O_3	Na_2O/Fe_2O_3	MgO/Fe_2O_3	Loess-Soil Na_2O/Al_2O_3	Loess-Soil Na_2O/Fe_2O_3
1	s	0.07	0.12	0.52	0	0
	l	0.07	0.12	0.51		
2	s	0.06	0.11	0.46	0.05	0.1
	l	0.11	0.21	0.53		
3	s	0.06	0.12	0.5	0.09	0.13
	l	0.15	0.25	0.46		
4	s	0.05	0.11	0.49	0.04	0.04
	l	0.09	0.15	0.48		
5	s	0.08	0.15	0.48	0.03	0.07
	l	0.11	0.22	0.52		

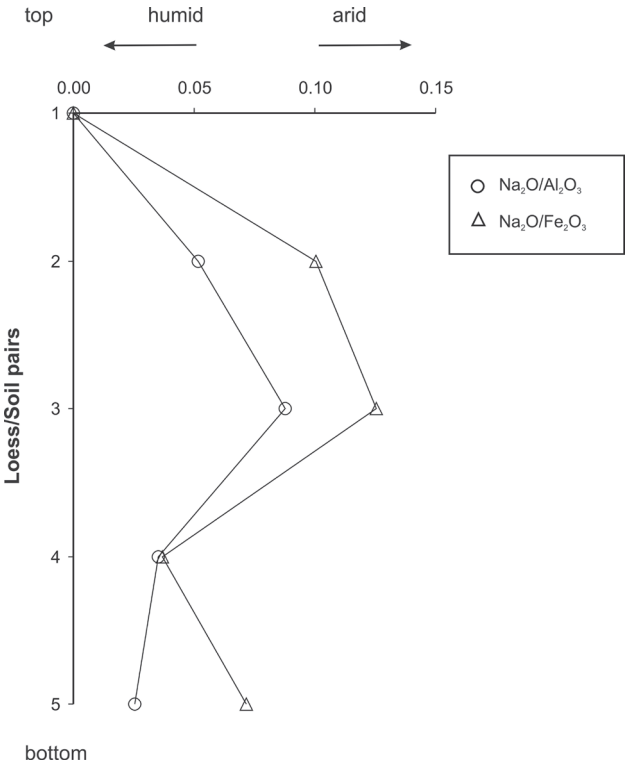

Figure 2. Difference between the relations of Na_2O/Al_2O_3, Na_2O/Fe_2O_3 and Na_2O/K_2O in alluvial loess and overlying soil in 5 different loess-soil pairs in a profile from Wadi Feiran in Sinai.

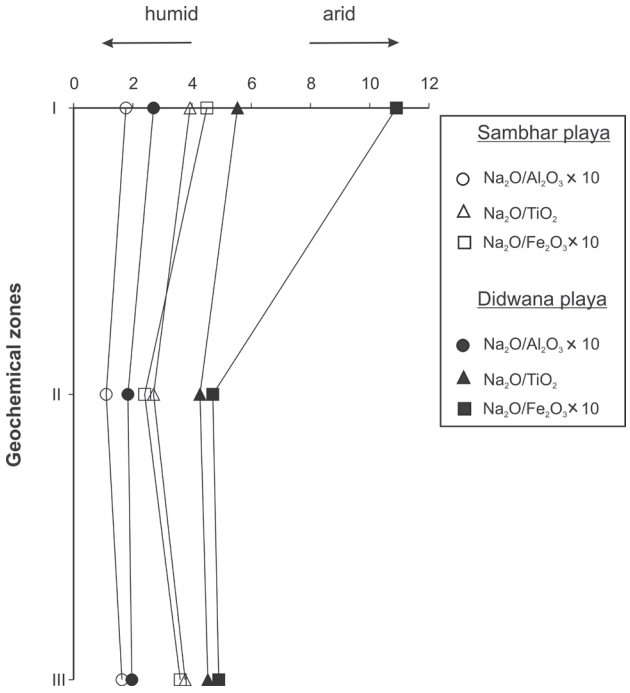

Figure 3. Comparison of Na_2O/Al_2O_3, Na_2O/TiO_2, Na_2O/Fe_2O_3 in the Sambhar and Didwana playas of the Thar Desert.

5th loess-soil pairs. Both these examples show the use of geochemical ratios in identifying arid events in the profiles of alluvial loess near the Feiran Oasis (Smykatz-Kloss *et al*., 1998; Knabe, 2000; Rögner *et al*., 2004). Two more examples of the application of geochemical ratios are given below, representing the relations in the playa lakes of the Thar Desert (Roy *et al*., 2006, 2008b). Table 4 presents the geochemical compositions in the zones (I: 0-10 cm, II: 10-130 cm, III: 130-150 cm) in a shallow profile from the Pachapadra playa lake (Roy *et al*., 2008b). The last example compares the geochemical relations between a semi-humid (desert margin) Sambhar playa and semi-arid Didwana playa (Figure 3 and Table 5).

4. Conclusions

This paper stresses the applicability of mineralogical and geochemical methods for characterising palaeoclimatological environments in tropical regions, especially in arid environments that lack preservation of biological proxies such as pollen, diatoms, and ostracods. Mineralogically, the persistence of (rare) Na- and (more common) Mg-evaporite minerals may prove the development of a former playa lake (via evaporation and transformation to more stable mineral products in the evaporitic crusts and hardpans). Transformed products, like Mg-salts or Na-, Mg-silicates such as magadiite, kenyaite, and palygorskite are reported from various palaeo-lakes.

Geochemically, the alteration of limnic sediments (*e.g*., alluvial loess in the Sinai and salty silts in the Thar) and formation of a soil cover by weathering of the underlying layer in contact with water suggest a shift from arid to (semi-) humid climate. The intensity of the humidification is observed in the evaluated differences in geochemical relations between underlying loess and overlying soils.

Table 4. Geochemical compositions (mass %) and relations in three different geochemical zones (I: 0-10 cm, II: 10-130 cm, III: 130-150 cm) from the Pachapadra playa or Thar Desert (compiled from Roy *et al*., 2008b).

	Al_2O_3	Na_2O	K_2O	TiO_2	Fe_2O_3	MgO	Na_2O/Al_2O_3	Na_2O/TiO_2	K_2O/Al_2O_3	K_2O/TiO_2	MgO/Fe_2O_3
I	12.92	7.82	1.47	0.62	7.91	3.4	0.61	12.65	0.11	2.38	0.43
II	17.08	4.53	2.6	0.75	12.64	5.07	0.27	6.03	0.15	3.46	0.4
III	11.55	4.39	1.88	0.5	7.54	4.23	0.38	8.77	0.16	3.75	0.56

Table 5. Geochemical comparison between shallow profiles of Sambhar (1.25 m, desert margin) and Didwana (1.2 m, central) playas of Thar Desert (compiled from Roy *et al*., 2006).

		Mass %						
Playa	Zones	Na_2O	Al_2O_3	Fe_2O_3	TiO_2	Na_2O/Al_2O_3	Na_2O/TiO_2	Na_2O/Fe_2O_3
Sambhar	I	2.24	12.69	4.99	0.57	0.18	3.93	0.45
	II	1.68	15.14	6.88	0.62	0.11	2.71	0.24
	III	2.1	12.89	5.84	0.56	0.16	3.75	0.36
Didwana	I	2.6	9.76	2.38	0.47	0.27	5.53	1.09
	II	2.57	13.98	5.51	0.6	0.18	4.28	0.47
	III	2.59	13.07	5.25	0.57	0.2	4.54	0.49

Especially during the humid period, Na-silicates are dissolved and partly transported and removed, while Al^{3+}, Ti^{4+}, and Fe^{3+} show to be nearly unaffected. Thus, the ratio of soluble (Na_2O) to hydrolysate elements (TiO_2, Al_2O_3, and Fe_2O_3) and their comparison report the (hydro-) geochemical development of the soil cover from the underlying loess. In suitable environments, a ^{14}C or luminescence age determination will be desirable, but in many cases the sediments (or soils) lack organic carbon for an exact age determination by ^{14}C method. Similarly, the younger sediments in arid regions are constantly mobilized by wind activity and are partially stripped of their OSL signal to be correctly dated by luminescence methods.

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