Borate Minerals and Related Authigenic Silicates in Northeastern Mediterranean
Late Miocene Continental Basins

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Abstract — Borate minerals, such as colemanite, ulexite and borax, have been reported in Tertiary continental basins of West Anatolia, Turkey, and Samos Island, Greece.

Tuffaceous rocks interbedded with the borates are rich in authigenic silicate minerals such as zeolites, potassium feldspar and opal-CT, all of which have a genetic association with the borates. Chemical analyses of the tuffaceous rocks show variation in major elements due to the varying percentage of carbonaceous, detrital, and volcanic derived material.

The volcanic activity in the vicinity of each particular lacustrine basin created ion-rich fluids from which a series of borates were precipitated under arid climate. Additionally, the saline-alkaline character of these fluids was responsible for the diagenesis of the silicic glass of the tuffaceous rocks, altering them to silicates and silica polymorphs.

Introduction

Borate minerals of commercial grade, such as colemanite, ulexite, borax, tincalconite and kernite, are known from Tertiary saline-alkaline lakes or playa (salar) deposits in the western United States, South America and western Turkey (Surdam, 1977; Helvaci, 1978; Countryman, 1979; Lefond and Barker, 1979; Alonso et al., 1988).

In some cases, other evaporite salts accompany the borates, or form independent deposits. The most common of these are celestite, trona, niter, gypsum and halite (Ericksen, 1981; Helvaci, 1977, 1984; Alonso et al., 1988; Helvaci and Inci, 1989; Helvaci et al., 1989; Stamatakis et al., 1989; Stamatakis and Economou, 1991).

Authigenic silicate minerals, such as clinoptilolite, analcime, mordenite, chabazite, boron-bearing potassium feldspar and howlite, usually accompany the borates, in a genetic association (Sheppard and Gude, 1969, 1973a, 1973b; Hay, 1966; Stamatakis, 1989a). The authigenic silicates have been diagenetically derived from the devitrification of acidic volcanic glass, usually volcanic ash, in a saline-alkaline lake environment (Surdam and Parker, 1972; Hay, 1970, 1977; Stamatakis, 1989b). They usually show a concentric orientation, with the most saline-alkaline products situated basinward, where the borate minerals are also formed (Sheppard and Gude, 1969, 1973a, 1973b).

In the eastern Mediterranean region, a variety of sedimentary environments has been reported during the Upper Miocene salinity crisis, for example lago-mare, lagoons and alkaline lakes (Cita et al., 1978). This paper describes the distribution of borate minerals and their association with certain authigenic silicates in Miocene continental basins, in the northeastern Mediterranean area.

Geological Setting

The borate deposits of the northeastern Mediterranean region are situated in western Anatolia, Turkey and Samos Island, Greece.

Western Anatolia, Turkey

The western Anatolia borate district contains five distinctly separated areas. From the west to east. They are: Bigadiç, Sultançayırı, Kestelek, Emet and Kırka (Meixner, 1965; Baysal, 1972; Helvaci, 1977) (Fig. 1). This district contains the largest borate reserves in the world (Lyday, 1982; Kistler and Smith, 1983; Helvaci, 1989).

All the deposits were formed during the Miocene in closed lacustrine basins with abnormally high salinity and alkalinity (Helvaci, 1986, 1989; Helvaci and Firman, 1976). The pre-Neogene basement of the basins is represented by Paleozoic and Mesozoic rocks, partly belonging to the Menderes massif. All these basins were partially filled with a series of tuffaceous rocks and lavas. Boron-rich fluids are presumed to have also circulated along faults into these basins (Helvaci, 1986, 1989). Although the sediments deposited in the borate lakes show some differences, they are generally represented by tuffaceous rocks, claystones, limestones and
Ca, Na, Mg, Sr-borates. Sandstones and conglomerates occur near the base of each basin (Özteker, 1969; Inan et al., 1973; Helvaci, 1989). Generally, the borates are enveloped between tuff- and clay-rich horizons (Fig. 2).

In all the above areas, intense calc-alkaline volcanic activity took place simultaneously with the borate sedimentation. Volcanic material includes acid through basic lavas and tuffaceous rocks.

**Samos Island, Greece**

On Samos Island there is an occurrence of borates in the central part of the Miocene Karlovassi basin (Stamatakis and Economou, 1991). The pre-Neogene basement of Samos Island is mainly constituted of Mesozoic marble, schist, minor quartzite, chert, and basic to ultrabasic rocks (Theodoropoulos, 1979). During the Late Miocene the Karlovassi basin was sequentially filled by limestone, tuffaceous rocks, claystone and siliceous limestone. These sediments are interpreted to be saline-alkaline lake deposits (Stamatakis, 1989b).

Acidic through basic lavas occur mainly at the eastern margin of the basin. The volcanic rocks of the Karlovassi basin seem to be very similar in their chemical characteristics to those of west-central Anatolia (Theofilopoulos and Voyatzakis, 1986).

**Analytical Techniques**

Scanning electron microscopy, X-ray diffraction analysis (Athens University, Greece), and Atomic Absorption Spectroscopy (I.G.M.E., Athens, Greece) were carried out on representative samples of the area studied. These analyses were made for the identification of the various authigenic silicates and the correlation of their major element distribution on tuffaceous material from the borate deposits.

The method described by Mumpton (1960) was used for the identification of clinoptilolite and heulandite. The results are given in Figure 3, and Tables 1 and 2.

**fig. 1. The borate districts in the northeastern Mediterranean.**

**Fig. 1.** The borate districts in the northeastern Mediterranean.
ulexite are abundant in both the borate zones, while other borates such as probertite, hydroboracite, inyoite, tunellite, and the borosilicate mineral howlite occur in lesser amounts (Meixner, 1965; Helvaci, 1989).

The tuffaceous material is represented by fine-grained ash-tuff. The volcanic glass of the ash-tuffs has been transformed to a series of authigenic silicates, such as boron-bearing K-feldspar, heulandite, clinoptilolite, illite and smectite (Figs. 1A, 1B, 1C). Volcanic-derived minerals are quartz and high sanidine. Minor amounts of chabazite and opal-CT were also identified by SEM analysis (Figs. 3B and 3D).

Kestelek Deposit

Commercial grade borates occur in various forms, such as thin layers, massive bodies and lenses, or nodules hosted within claystone. Colemanite, ulexite and probertite predominate, while hydroboracite occurs in scant amounts (Helvaci, 1978).

Yellow to brown coarse-grained tuff, up to a few centimeters is interbedded between the borate zones. Clinoptilolite is the only identified authigenic silicate mineral in this tuff which is mainly composed of oligoclase, quartz, biotite and smectite.

Sultancayiri Deposit

Commercial grade borates are represented by priceite (pandermite), accompanied by minor amounts of colemanite and howlite. Priceite layers are alternated with claystone, while howlite forms nodules within claystone. Gypsum is present in massive amounts, or as intergrowths with priceite (Helvaci, 1978).

Boron-bearing K-feldspar, clinoptilolite and opal-CT are the authigenic silicate and silica phases. High sanidine, anorthoclase, quartz, and clay minerals such as illite and smectite are also present.

Emet Deposit

Commercial grade borates are represented by colemanite with minor amounts of ulexite. Other borates found in subsidiary concentrations are tunellite, terrugite, veatchite-A, cahnite, hydroboracite and meyerhofferite. The borates are interbedded with tuffs, claystones and marlstones (Helvaci, 1984, 1986; Helvaci and Firman, 1976).

Boron-bearing K-feldspar, clinoptilolite, illite and smectite are the authigenic silicates detected in the tuffaceous samples. Volcaniclastic high sanidine and quartz are also present. Native sulphur, celestite, gypsum, realgar and orpiment occur in the borate zone throughout the area. Secondary calcite occurs in surface outcrops as a product of the weathering of borates (Helvaci, 1984).

Kirka Deposit

Borax (tinkal), colemanite and ulexite are the principal ore minerals of the commercial grade borate bodies of Kirka. Inyoite, tincalconite, kernite, tunellite, kurnakovite and indérite are minor components of the borate paragenesis (Inan...
et al., 1973; Helvacı, 1978) The borate minerals are interlayered with claystones and tuffs, while a porcellanous limestone forms a resistant cap rock for the borate zone. Celestite, gypsum, realgar, and orpiment occur in some borate layers.

Boron bearing K-feldspar, erionite, smectite, and illite are the authigenic silicates found in the tuffaceous horizons, accompanied by volcanic-derived high sanidine, albite, anorthoclase, quartz, and calcite.

Samos Occurrence

In the western part of Samos Island (Fig. 1), a small borate lens occurs which is composed of colemanite and traces of ulexite. Thin layers of gypsum occur below the borates, while celestite nodules and lenses occur within and above the borates (Stamatakis and Economou, 1991). The borate lens has been partially weathered to porous calcite.

The borate body is hosted in claystone and tuffaceous layers Late Miocene in age. The volcanic glass of the tuffaceous rocks has been altered to boron-bearing K-feldspar, smectite, albite, tridymite, cristobalite, opal-CT, clinoptilolite, analcime, chabazite, mordenite and minor amounts of other zeolites such as phillipsite and erionite (Figs. 1E and 1F, see also Stamatakis, 1989a, 1989b).
Table 1.

<table>
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<tr>
<th>Sample number</th>
<th>Mineralogical composition</th>
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</tr>
<tr>
<td>B₂</td>
<td>hs, qtz, Kf</td>
</tr>
<tr>
<td>B₃</td>
<td>cl</td>
</tr>
<tr>
<td>B₃₅ (2HCl)</td>
<td>Kf, qtz, cc, sm, ill</td>
</tr>
<tr>
<td>B₄</td>
<td>Kf, sm, ill</td>
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<tr>
<td>B₅</td>
<td>cl, heul, qtz</td>
</tr>
<tr>
<td>BE₂</td>
<td>hs, Kf, sm, qtz, ill</td>
</tr>
<tr>
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<td>ab, qtz, mc, cc, cl, er</td>
</tr>
<tr>
<td>E₁</td>
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<tr>
<td>E₁ (2µ)</td>
<td>Kf, sm, ill</td>
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<tr>
<td>K₁</td>
<td>cc, qtz, hs, sm, ab, ill</td>
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<tr>
<td>K₂</td>
<td>Kf, an, qtz, sm, ill, er</td>
</tr>
<tr>
<td>K₂ₙ (2µ)</td>
<td>Kf, sm, cc, ill, qtz, an</td>
</tr>
<tr>
<td>KES₁</td>
<td>ol, qtz, biot, cl, sm</td>
</tr>
<tr>
<td>KES₁₈</td>
<td>qtz, ol, cl, biot</td>
</tr>
<tr>
<td>KES₂</td>
<td>dol, cc, ill, plag, sm, qtz</td>
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<td>SM₄</td>
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<td>ST₈</td>
<td>o-CT, Kf, qtz, an, cl</td>
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<tr>
<td>ST₁₈</td>
<td>qtz, o-CT, cl, hs, Kf, ill</td>
</tr>
</tbody>
</table>

Explanatory notes: B = Bigadic, BE = Beypazari, E = Emet, K = Kirka, KES = Kerelek, SM = Samos, ST = Sultanpour. cc = calcite, dol = dolomite, sm = smectite, ill = illite, mc = muscovite, biot = biotite, qtz = quartz, Kf = boron-bearing K-feldspar, hs = high sanidine, an = anorthoclase, ab = albite, ol = oligoclase, plag = plagioclase, cl = clinopyroxite, er = erionite, chab = chabazite, ac = analbite, heul = heulandite, o-CT = opal-CT. 2HCl = 2 nárcotic fraction treated with HCl 1N.

The authigenic silicates and the silica polymorphs form a distinct areal zonation in the Karlovassi basin which is not observed in any of the other eastern borate basins. In these zones, clinopyroxite generally occupies the external zone of authigenic silicates, while the boron-bearing K-feldspar is the central one (Stamatakis, 1989a). This zonation of the authigenic silicates is quite common in saline-alkaline basins from the western United States (Sheppard and Gude, 1968, 1969, 1973a, 1973b).

The borates occur within the central part of the Miocene Karlovassi basin, where the salinity and alkalinity of the basin was high, as defined by the presence of the authigenic K-feldspar (Stamatakis 1989b).

In order to better correlate the authigenic silicates with the formation of evaporites in Samos Island and western Anatolia, two more basins are described that exist close to the above basins and also host authigenic silicates and evaporites.

**Beypazari Trona Deposit**

The Beypazari saline-alkaline Miocene trona deposit is situated to the east of the Anatolian borate deposits, and contains sodium carbonate and sodium sulphate evaporites such as trona, nakholite and thenardite (Helvaci and Inci, 1989; Helvaci et al., 1989). These minerals are accompanied by minor amounts of pirosmnite and thermonatrite (Helvaci and Inci, 1989).

**Mytilini Niter, Halite, Sylvite Occurrence**

The Mytilini Late Miocene basin is situated just to the east of Karlovassi basin of Samos Island. It hosts subsidi-
ary amounts of evaporite minerals, such as niter, halite and sylvite. The only authigenic silica mineral observed in the basin is opal-CT, which has been derived from a biogenic opal-A precursor (Stamatakis et al., 1989).

Although tuffaceous rocks are present, their volcanic glass has not been transformed to any zeolite, or K-feldspar.

By contrast to the adjacent Karlovassi basin, the Mytilinii basin does not contain borates, but boron concentrations were sufficient that diatom frustules have incorporated boron in values up to 2500 ppm (Stamatakis et al., 1989).

### Discussion

Chemical analyses of the tuffaceous material that is found in close association with the borate and trona deposits show variation in major element concentration which is mainly attributed to the presence or absence of carbonates (Tables 1 and 2).

Though the high sanidine and anorthoclase-rich samples have high K$_2$O concentrations, the authigenic K-feldspar-rich samples show the highest values of this oxide because the latter are poor in detrital minerals (Tables 1 and 2). Na$_2$O values are higher in the Kestelek tuffs, which contain oligoclase as a major constituent. Authigenic K-feldspar and opal-CT-rich samples have the lowest loss on ignition (LOI) values. By contrast, rocks with high percentages of carbonates, zeolites and clays yield high LOI values.

The highest SiO$_2$ concentration was detected in the tuffaceous rocks from Sultancayiri, where both opal-CT and quartz are major constituents.

High CaO and MgO content depends on the abundance of dolomite and calcite in some tuffaceous samples. Al$_2$O$_3$ content does not correlate with any other major element, although its highest concentration accompanies the high concentration of K$_2$O in the authigenic K-feldspar-rich sample SM$_3$ from Samos Island.

Generally, the particle size of the tuffaceous material from all the basins studied is fine-grained, representing ash-fall tuffs, whereas coarser-grained tuffs are found at Sultancayiri and Kestelek areas. Notably, the most fine-grained tuffs occur at Karlovassi basin which is the westernmost of all the basins studied. This may be due to the larger distance of the basin from the center of volcanic activity.

As described by Sheppard and Gude (1973b), the occurrence of authigenic boron bearing K-feldspar is a good indicator for the presence of evaporites, especially borate minerals in closed continental basins. Though the Karlovassi, Bigadic, Emet, Kirka, Sultancayiri and Beypazari basins fit that suggestion, Kestelek tuffaceous rocks that are interlayered with the borate body, do not contain K-feldspar, but only zeolites. Additionally, the Mytilinii basin contains trace amounts of evaporites, but does not contain zeolites or authigenic K-feldspar.

The presence, therefore, of the authigenic K-feldspar is not actually a prerequisite for the presence of borates. Similar results are obtained from studies of borates and related authigenic silicates of the Jarandol Miocene basin of Serbia, Yugoslavia (Obradovic et al., 1993).

As can be seen from Table 1, the major differences between the studied basins in the area concerning the kind and abundance of the various silicate and borate minerals, are probably due to variations in pore fluids and volcanic material chemistry. These could be due to: (a) differences of pH values from one lake to another, a case which is already well documented for the two Miocene lakes of Samos Island (Stamatakis, 1989b; Stamatakis et al., 1989); and (b) differences in the type and quantity of the original volcanic material which had been deposited into each particular basin (volcanic ash, coarse tuff, volcanic emanations).

Where authigenic K-feldspar is present (Karlovassi, Emet, Bigadic, Kirka, Sultancayiri), a pH greater than 9 can be anticipated basinward during the final stages of lake evolution. The pH values near the margins of these lakes may have been lower than 8. In the area of Kestelek, the presence of only clinoptilolite and opal-CT seems to reflect pH values not higher than 9 (Surdam and Parker, 1972). This obser-

### Table 2. Chemical analyses of tuffaceous rocks from western Anatolia and Samos borate districts

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>SiO$_2$</th>
<th>Al$_2$O$_3$</th>
<th>TiO$_2$</th>
<th>FeO</th>
<th>Fe$_2$O$_3$</th>
<th>MnO</th>
<th>MgO</th>
<th>CaO</th>
<th>Na$_2$O</th>
<th>K$_2$O</th>
<th>LOI</th>
<th>Total</th>
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<tbody>
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<td>B$_1$</td>
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<td>0.57</td>
<td>0.08</td>
<td>0.14</td>
<td>0.27</td>
<td>0.04</td>
<td>0.27</td>
<td>0.30</td>
<td>1.14</td>
<td>0.24</td>
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<td>0.10</td>
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<td>0.96</td>
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<td>0.03</td>
<td>—</td>
<td>0.63</td>
<td>0.04</td>
<td>0.22</td>
<td>1.30</td>
<td>2.30</td>
<td>4.65</td>
<td>5.30</td>
<td>99.57</td>
</tr>
</tbody>
</table>

B = Bigadic, BE = Beypazari, ST = Sultancayiri, E = Emet, K = Kirka, KES = Kestelek, SM = Samos — = not detected
vation points to the conclusion that a standard pH value is not necessary to precipitate borate salts but a range of values higher than 8. Ataman and Baysal’s (1978) measurements on clay minerals of western Anatolian borate deposits indicated that there are differences between the pH of the Emet, Bigadic and Kirka basins.

Similar variations in pH values have also been reported in the two evaporitic lakes of Samos (Mytilinii, and Karlovasi basins), where the degree of silica diagenesis and the formation of certain authigenic silicates are quite different (Stamatakis, 1986).

In conclusion, all the mineralogical variations of evaporitic and authigenic minerals in the Miocene continental basins of the northeastern Mediterranean indicate that the formation of either authigenic silicates, or borates, trona, gypsum, celestite, halite, niter, and sylvite was controlled by the type and density of the volcanic products in any particular Neogene lacustrine basin.

The presence of saline and alkaline conditions in each borate basin is well documented by the presence of evaporites themselves, and also by the occurrence of characteristic authigenic silicates, such as clinoptilolite, analcime and K-feldspar. The formation and the paragenetic association of the authigenic silicates require pH values ranging from around 8 to more than 10. That pH range in the borate basins indicates that the precipitation of borates could start at a pH around 9 and continue at higher alkalinities.

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