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# A boron-bearing potassium feldspar in volcanic ash and tuffaceous rocks from Miocene lake deposits, Samos Island, Greece

MICHAEL G. STAMATAKIS
Department of Geology, Section of Mineralogy, Petrology, and Economic Geology, National University of Athens, Panepistimiopolis, Ano Ilissia, Athens 15784, Greece

## ABSTRACT

The Lower Neogene Unit (LNU) of west Samos Island, Greece, consists of volcanic ash and tuffaceous rocks, claystone, marlstone, and porcelaneous limestone. The initial fresh glass of volcanic rocks has usually transformed to authigenic K-feldspar, zeolites, opal-CT, and smectite. K-feldspar is the predominant mineral in the central part of the LNU, whereas in adjacent margins, K-feldspar occurs only in the sandy tuffs. Nowhere is the K-feldspar associated with fresh glass. Chemical, SEM, and XRD analyses show the K-feldspar to be B-bearing, with B concentrations up to 2500 ppm. Statistical treatment of chemical data demonstrates a positive correlation of B with K and Rb. This B-bearing K-feldspar seems to be analogous to K-feldspars described in saline-alkaline lake deposits associated with nonmarine evaporites.

## INTRODUCTION

Authigenic K-feldspar has been reported as a diagenetic product in tuffs, sandstones, and carbonates from marine and lake environments (Hay, 1966; Kastner, 1971; Goodwin, 1973; Sheppard and Gude, 1972, 1973a, 1973b; Desborough, 1975; Starkey and Blackmon, 1979; Ali and Turner, 1982; Helvaci, 1986; Obradović and Dimitrijevic, 1987), but the B-bearing variety has been reported only from saline-alkaline lake deposits of tuffaceous rocks (Sheppard and Gude, 1968, 1969, 1972, 1973a, 1973b; Lefond and Barker, 1979). The relatively high B concentrations in this K-feldspar, compared to the very low B values of K-feldspar of volcanic origin (Harder, 1974; Stamatakis, 1986), suggest a partial substitution of B for Al in the series $KAlSi_3O_8$-$KBSi_3O_8$ in certain environments (Martin, 1971; Sheppard and Gude, 1973b). The zeolites clinoptilolite and analcime, as well as the borosilicates howlite and searlesite, and smectite are the commonest paragenetic associates of this K-feldspar (Sheppard and Gude, 1968; Hay, 1970; Surdam and Parker, 1972; Smith, 1979; Obradović and Jovanović, 1987). Usually there is a gradual diagenetic transition from unaltered fresh glass through smectite and zeolites in the moderately high saline-alkaline pore fluids to K-feldspar in the most saline-alkaline pore fluids of the lake (Sheppard and Gude, 1968).

## GEOLOGIC SETTING

On Samos Island, two main Neogene basins were developed, namely the basin of Mytilinii in the east and that of Karlovassi in the west (Fig. 1). The pre-Neogene basement of both basins consists of four tectonic units containing marbles, dolomites, quartzites, phyllites, and metamorphic basic-ultrabasic rocks and a younger nappe of diabases, peridotites, cherts, and limestones (Papanikolaou, 1979; Theodoropoulos, 1979; Mezger and Okrusch, 1985).

Neogene deposits of the Karlovassi basin start with a basal reddish-brown conglomerate overlain by the Lower Neogene Unit (LNU) of Theodoropoulos (1979). The LNU has a maximum thickness of 400 m and is subdivided into four well-recognized successions, starting from the base: (1) limestones and marly limestones with scant occurrences of thin lignitic horizons, (2) tuffaceous rocks and volcanic ash layers containing abundant authigenic silicates, (3) dolomitic clays with local occurrences of evaporites, and (4) porcelaneous limestones and porcelanites rich in silica polymorphs (Stamatakis, 1986). A 200-m-thick red and green conglomerate overlies the LNU and is interbedded with Pontian-age silts and sands (Theodoropoulos, 1979). Pliocene fresh water marlstones and limestones up to 120 m thick occur at the top of the Neogene sedimentary rocks. Deposits of the basin are presumed to be continental, Tortonian through Pliocene in age (Büttner and Kovalczyk, 1978; Besenecker and Büttner, 1978; Dermitzakis and Papanikolaou, 1981). At the margins of the Karlovassi basin there are occurrences of rhyolites, dacites, trachytes, and basalts of Neogene age (Karageorgiou, 1946; Theodoropoulos, 1979). These volcanic rocks are responsible for local silicification and kaolinitization of the surrounding LNU rocks.

Samples were taken from surface exposures and from cores of three boreholes as much as 250 m deep, drilled by the Institute of Geological and Mining Research of Greece (Fig. 2).

## ANALYTICAL TECHNIQUES

Samples were analyzed for B using atomic-absorption spectroscopy and a colorimetric method (Stanton and McDonald,

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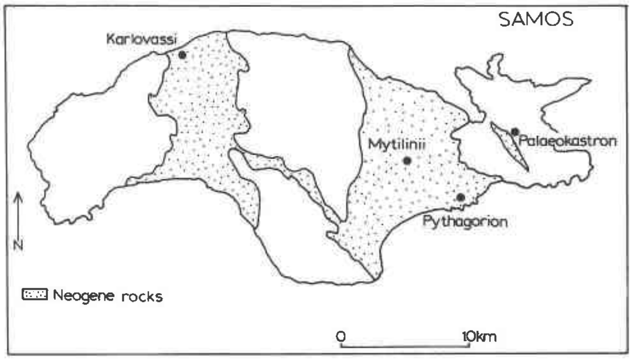

Fig. 1. The Neogene basins of Samos Island.

1966) slightly modified by Stamatakis (1986) at the Department of Geology, Athens University. Major and trace elements (Table 1) were analyzed by the plasma (ICP) method at Imperial College, London. The SEM analysis was carried out using a Cambridge S4-10 scanning electron microscope at the University of New Brunswick, Canada. The 2 μm fraction was separated by differential precipitation. A 1*N* HCl solution was used for the decomposition of carbonates and a Philips PW1010 X-ray diffractometer at the Department of Geology at Athens University was used for mineral analysis (Table 2). The correlation coefficient of B with certain trace and major elements was used for statistical analysis (Table 1).

## PETROLOGY AND MINERALOGY OF K-FELDSPAR-RICH TUFFACEOUS ROCKS

The marginal areas of the LNU are occupied by carbonate rocks, representing an older Miocene succession above the basal conglomerate. Authigenic silicates are absent in these rocks. In contrast, all three overlying successions contain authigenic silicates and silica polymorphs. Especially in the "b" succession, which mainly occupies the central part of the Karlovassi basin, authigenic K-feldspar commonly represents more than 30 wt% of the rocks. In many cases, the 2-μm fraction is composed only of K-feldspar (Table 2). The K-feldspar is usually accompanied by clinoptilolite and/or analcime, anorthoclase, or high sanidine, illite and/or muscovite, and smectite. The following minerals are detected rarely: opal-CT, erionite, chabazite, phillipsite, biotite, chlorite, and albite (Stamatakis, 1986, Table 90). The limits of K-feldspar–rich tuffaceous rocks are defined by those rocks having a higher percentage by weight of zeolites than of K-feldspar, as determined by bulk averages of authigenic silicate occurrences (Fig. 3). This spatial development of authigenic silicates in the Karlovassi basin is analogous to zoning observed in certain saline-alkaline lake deposits (Sheppard and Gude, 1968, 1969, 1973a; Hay, 1977). The color of tuffaceous rocks varies from yellowish brown to green and dull white. In some cases turbiditic layers occur having a uniform mineral composition.

The mean particle size of these K-feldspar–rich rocks is very small, and so they might be termed volcanic ash. In the rare coarse-grained samples, transparent, prismatic crystals of high sanidine or anorthoclase, up to 0.5 cm in length, are observed along with well-rounded fragments of massive tuffaceous material.

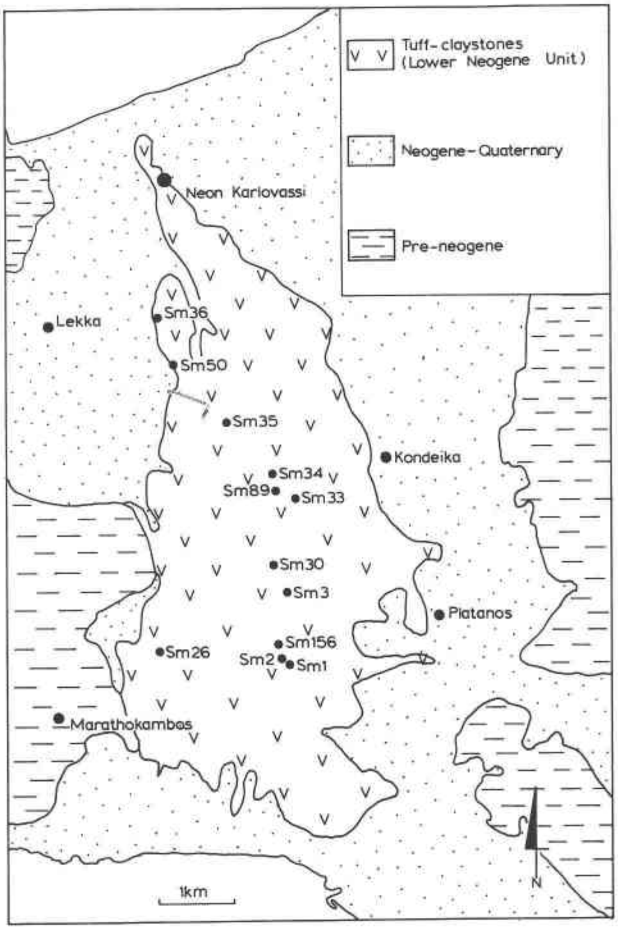

Fig. 2. Modified geologic map of the basin with sampling sites. Sm1, Sm2, Sm3 are the sites of the three boreholes.

Aggregates of K-feldspar were identified by XRD, SEM, and chemical analyses (Fig. 4, Tables 1, 2). Crystals of K-feldspar rarely surpass 10 μm in length and usually have an idiomorphic tabular or rhombohedral form (Fig. 4). The XRD pattern of this K-feldspar has the same intensities and *d* values as those described from saline-alkaline lake deposits of the western USA (Sheppard and Gude, 1965, 1973a, 1973b). The SEM microanalyses and morphological observations of the authigenic crystals of the LNU do not indicate the presence of searlesite or howlite, borosilicate minerals usually present in B-rich saline-alkaline lake deposits (Sheppard and Gude, 1968; Lefond and Barker, 1979; Smith, 1979; Smith et al., 1983; Obradović and Jovanović, 1987).

## GEOCHEMISTRY AND DIAGENETIC REACTIONS

A general characteristic of the tuffaceous rocks of the LNU is their high B contents. Material notably rich in K-feldspar has B contents ranging from 1500 to 2500 ppm (Table 1). Sheppard and Gude (1972, 1973b) re-

**TABLE 1.** Chemical analyses of samples rich in authigenic K-feldspar

| Sample | B | K | Na | Li | Rb | Mg | Ca | Sr | Ba | Al | La | Ti | Mn | Fe | Zn | Pb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sm1:15* | 1595 | 74000 | 52000 | | | | | | | | | | | | | |
| Sm156′† | 1200 | 70000 | 52000 | | | | | | | | | | | | | |
| Sm33i | 840 | 47000 | 10500 | 84 | 280 | 11200 | 11700 | 59 | 98 | 71000 | 107 | 1890 | 320 | 1720 | 73 | 109 |
| Sm33h | 800 | 47000 | 3300 | 28 | 240 | 11100 | 18800 | 56 | 24 | 57000 | 106 | 1370 | 144 | 16000 | 42 | 96 |
| Sm3:130 | 1320 | 70000 | 6200 | 123 | 420 | 11500 | 18800 | 59 | 40 | 68000 | 88 | 1550 | 490 | 17200 | 36 | 67 |
| Sm33i* | 1770 | 75000 | 5200 | | | | | | | | | | | | | |
| Sm33h* | 1760 | 75000 | 5200 | | | | | | | | | | | | | |
| Sm33g* | 2380 | 79000 | 5200 | | | | | | | | | | | | | |
| Sm36r* | 1595 | 75000 | 8900 | | | | | | | | | | | | | |
| Sm156*† | 1807 | 77000 | 5100 | | | | | | | | | | | | | |
| Sm34*† | 2464 | 85000 | 6300 | | | | | | | | | | | | | |
| Sm34″*† | 2486 | 85000 | 5300 | | | | | | | | | | | | | |
| Sm33a* | 2365 | 81000 | 5200 | | | | | | | | | | | | | |
| Sm36o* | 2050 | 80000 | 5100 | | | | | | | | | | | | | |
| Sm1:15a | 900 | 62000 | 7100 | 77 | 340 | 4900 | 4200 | 48 | 37 | 72000 | 92 | 1370 | 380 | 19500 | 99 | 79 |
| Sm1:15 | 820 | 45000 | 6000 | 340 | 330 | 16800 | 7100 | 102 | 42 | 78000 | 117 | 1400 | 149 | 17900 | 77 | 91 |
| Sm36o | 1050 | 65000 | 10900 | 172 | 430 | 11600 | 3900 | 96 | 77 | 73000 | 92 | 1710 | 1360 | 26000 | 63 | 65 |
| Sm34a | 870 | 52000 | 6700 | 105 | 320 | 13900 | 15800 | 105 | 80 | 74000 | 94 | 2100 | 670 | 15900 | 84 | 64 |
| Sm34† | 910 | 59000 | 9100 | 87 | 300 | 12220 | 14700 | 171 | 58 | 77000 | 98 | 1610 | 350 | 14900 | 76 | 84 |
| Sm34a* | 2535 | 90000 | 5300 | | | | | | | | | | | | | |
| Sm33b* | 2480 | 89000 | 5200 | | | | | | | | | | | | | |
| Sm36j | 856 | 52000 | 13500 | 104 | 290 | 14100 | 2400 | 42 | 63 | 75000 | 89 | 1770 | 82 | 13700 | 72 | 60 |
| Sm34e* | 1812 | 79000 | 5300 | | | | | | | | | | | | | |
| Sm35* | 1760 | 77000 | 11600 | | | | | | | | | | | | | |
| Sm33b | 1460 | 72000 | 4900 | 25 | 490 | 7300 | 9800 | 44 | 40 | 74000 | 95 | 1470 | 360 | 16200 | 78 | 71 |
| Sm35′* | 1038 | 71000 | 13500 | | | | | | | | | | | | | |
| Sm50j | 1405 | 67000 | 5500 | 84 | 350 | 5700 | 2600 | 50 | 80 | 75000 | 97 | 1940 | 320 | 16500 | 72 | 75 |
| Sm1:51* | 1375 | 72000 | 5900 | | | | | | | | | | | | | |
| Sm1:52* | 2250 | 83000 | 12900 | | | | | | | | | | | | | |
| Sm33* | 1243 | 70000 | 5100 | | | | | | | | | | | | | |
| Sm156† | 1341 | 69000 | 5000 | 163 | 540 | 6900 | 5300 | 530 | 50 | 75000 | 135 | 1200 | 42 | 16900 | 64 | 69 |
| Sm89 | 835 | 49000 | 5500 | 240 | 420 | 10000 | 8700 | 1530 | 42 | 74000 | 161 | 1080 | 46 | 17900 | 49 | 107 |
| Sm30b | 868 | 50000 | 5700 | 114 | 430 | 8000 | 12500 | 3900 | 67 | 76000 | 134 | 1110 | 96 | 18600 | 83 | 88 |
| Sm26h | 824 | 49000 | 4400 | 181 | 260 | 12200 | 6800 | 86 | 72 | 76000 | 101 | 1620 | 240 | 22000 | 65 | 75 |

*Note:* In all the samples, the values of additional trace elements are lower than the amounts given here (ppm): Cd 1.26, Ag 4.4, Cu 15, Co 13, Ni 33, Be 16, Cr 26, V 33, Mo 16, P 360. Sm1:15 means borehole no. 1, depth 15 m. Sm34a, Sm34b, Sm34c, etc. are samples of the same surface section at various stratigraphic levels. Correlation coefficients between B and the following trace and major elements: K 0.92, Na −0.09, Li −0.27, Rb 0.67, Mg −0.53, Ca −0.14, Sr −0.019, Ba −0.09, Al −0.07, La − 0.19, Ti 0.05, Mn 0.14, Fe 0.17, Zn −0.28, Pb −0.47.

* Samples treated with 1*N* or 6*N* in their 2 μm fraction.

† Sm*X*′ or Sm*X*″ are samples of the same tuffaceous layer at distances of 0.5 and 1.0 m, respectively.

ferred to B contents between 500 and 5000 ppm, but usually greater than 1000 ppm, in K-feldspars deposited in a saline-alkaline lake environment. In contrast, B contents in tuffaceous rocks of the LNU containing abundant volcanic or detrital anorthoclase and high sanidine (as opposed to authigenic K-feldspar) are less than 100 ppm, and the separated anorthoclase and high sanidine crystals have B contents of 30 and 10 ppm, respectively (Stamatakis, 1986, Table 85). Also, samples of the LNU with abundant clay minerals have B contents lower than 280 ppm (Stamatakis, 1986, Table 86), suggesting that the high B contents of the tuffaceous rocks of the LNU cannot be attributed to the presence of illite, the most B-rich clay mineral (Harder, 1974). Treatment of samples with 1*N* and 6*N* HCl indicates that the B is held in a form having low solubility. Statistical treatment of the chemical data showed that a statistically significant correlation exists between B and K and between B and Rb, indicating that B is associated with authigenic K-feldspar (Tables 1 and 2). The lack of significant correlation between B and Na, Ca, Sr, and Mg also indicates the lack of discrete Na, Sr, Ca, Mg borate minerals, and also howlite and searlesite, that would otherwise contribute substantial B concentrations to the samples. The moderately high values of Sr and La in the samples are characteristic of tuffaceous rocks accompanied by evaporites in saline-alkaline lake deposits (Sheppard and Gude, 1969, 1973a; Helvaci and Firman, 1976; Helvaci, 1978, 1984, 1986).

The association of K-feldspar ($KAlSi_3O_8$) with clinoptilolite ($((K,Na)AlSi_5O_{12}\cdot 3.5H_2O)$) or analcime ($NaAlSi_2O_6\cdot H_2O$) suggests that the $K^+/Na^+$, $Al^{3+}$, and $H_2O$ activities have not been constant. The predominance of K-feldspar in some areas of a lake deposit indicates higher $K^+/Na^+$ activities than in areas containing only analcime (Smith, 1979). The activities of $K^+$, $Na^+$, $Al^{3+}$, and $H_2O$ apparently were those required for K-feldspar–analcime–clinoptilolite stability (Fig. 5) but not for searlesite, as presumed from its absence.

## GENESIS OF THE B-BEARING K FELDSPAR

Three lines of field and geochemical evidence define the environment of K-feldspar formation: the zoning of authigenic silicates, their high B contents, and the development of evaporite minerals. These parameters are characteristic of saline-alkaline deposits, where B enters the structure of authigenic silicates during diagenetic re-

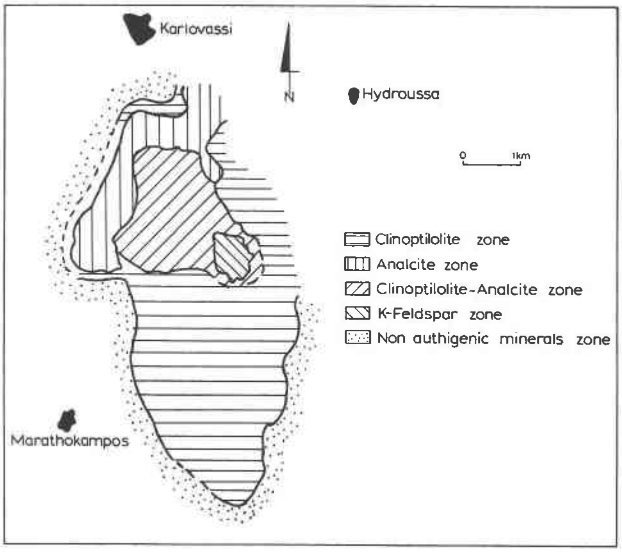

Fig. 3. Zoning of authigenic silicates in the Karlovassi basin, LNU formation. The open area in the east is covered by younger sediments.

**Table 2.** Mineral content of samples taken from volcanic ash and tuffaceous rocks

| Sample | Mineral content |
|---|---|
| Sm1:15* | K-feldspar (MJ) |
| Sm156′ | K-feldspar (MJ), anorthoclase (MD), clinoptilolite (MD) |
| Sm33i | K-feldspar (MJ), anorthoclase (MJ), smectite (TR) |
| Sm33hh | K-feldspar (MJ), opal-CT (MD), anorthoclase (MD) |
| Sm3:130 | K-feldspar (MJ), dolomite (MD), anorthoclase (MD), smectite (TR) |
| Sm33i* | K-feldspar (MJ), smectite (TR), anorthoclase (TR) |
| Sm33h* | K-feldspar (MJ), opal-CT (MD) |
| Sm33g* | K-feldspar (MJ) |
| Sm36r* | K-feldspar (MJ), anorthoclase (TR) |
| Sm156* | K-feldspar (MJ), clinoptilolite (MD) |
| Sm34* | K-feldspar (MJ) |
| Sm34″* | K-feldspar (MJ) |
| Sm33a* | K-feldspar (MJ) |
| Sm36o* | K-feldspar (MJ) |
| Sm1:15a | K-feldspar (MJ), anorthoclase (MD), muscovite (TR) |
| Sm1:15 | K-feldspar (MJ), anorthoclase (MD), muscovite (TR) |
| Sm26h | K-feldspar (MJ), clinoptilolite (MJ), anorthoclase (MD) |
| Sm36o | K-feldspar (MJ), anorthoclase (MJ) |
| Sm34a | K-feldspar (MJ), anorthoclase (MJ) |
| Sm34 | K-feldspar (MJ), anorthoclase (MJ) |
| Sm34a* | K-feldspar (MJ) |
| Sm33b* | K-feldspar (MJ) |
| Sm36j | K-feldspar (MJ), anorthoclase (MJ) |
| Sm34e* | K-feldspar (MJ), illite (TR) |
| Sm35* | K-feldspar (MJ), analcite (TR) |
| Sm33b | K-feldspar (MJ), anorthoclase (MD) |
| Sm35′* | K-feldspar (MJ), anorthoclase (MD) |
| Sm50j | K-feldspar (MJ), high sanidine (MD) |
| Sm1:51* | K-feldspar (MJ), chabazite (TR), erionite (TR), illite (TR) |
| Sm1:52* | K-feldspar (MJ), chlorite (TR) |
| Sm33* | K-feldspar (MJ), clinoptilolite (TR), illite (TR), anorthoclase (TR) |
| Sm156 | K-feldspar (MJ), anorthoclase (MD), clinoptilollite (MD) |
| Sm89 | K-feldspar (MJ), anorthoclase (MD), clinoptilolite (MD), smectite (TR), illite (TR) |
| Sm30b | K-feldspar (MJ), anorthoclase (MD), clinoptilolite (MD) |

*Note:* MJ, major; MD, medium; TR, trace. See also notes in Table 1.

actions involving volcanic ashes and tuffaceous rocks (Martin, 1971; Sheppard and Gude, 1973a). In general, borate ions remain in pore fluids and enter the structure of K-feldspar in the late stages of diagenesis (Sheppard and Gude, 1972). If higher B concentrations exist, then Ca, Sr, Na, and Mg borates can be formed along with K-feldspar, howlite, and searlesite.

It is well known that with an increase of salinity and alkalinity of the pore fluids, fresh glass is progressively altered according to the sequence glass → smectite → clinoptilolite → analcime → K-feldspar (Surdam and Parker, 1972; Hay, 1977). The abundance of K-feldspar in the Miocene lake, with no associated volcanic glass relicts and with its spatial relationship to adjacent zeolites, strongly suggests K-feldspar formation after other silicates in sites of higher salinity and alkalinity (Sheppard and Gude, 1973b). Its restricted occurrences at some sites in the distal part of the "b" succession, where sandy tuffaceous rocks were developed between massive clinoptilolite-rich tuffaceous rocks indicate alater stage of K-feldspar formation in areas of high porosity. The development of different diagenetic stages in the generation of authigenic K-feldspar has been observed in various lake basin deposits (Sheppard and Gude, 1972, 1973b). The types of authigenic minerals found in the Samos Miocene lacustrine deposits—principally K- and Na-bearing zeolites, K-feldspar, and Ca-, Sr-, and Na-bearing sulfate evaporites (Stamatakis, 1986)—permit the interpretation that the Miocene lake waters and pore fluids were enriched in borate, sulfate, and silicate anions and in K, Na, Ca, and Sr cations. Nonetheless, $H_2O$ activity was relatively low in the central part of this lacustrine system, where K-feldspar predominates over the hydrous zeolites. This low $H_2O$ activity must be attributed to the highly concentrated solutions in the central part of the Miocene lake.

Sources for these ions were probably the volcaniclastic sediments which compose the ash falls and tuffaceous deposits of the LNU rocks. Devitrification reactions would have released these various ions into solution during diagenesis. The gradual desiccation of the late Miocene Karlovassi basin, probably attributable to the Messinian salinity crisis of the Mediterranean Sea, was responsible for the decrease of water volume and the relative enrichment of total dissolved salts in the lake. The gradual desiccation of the late Miocene basin is closely related to events postulated to have taken place in both the Karlovassi and Mytilinii basins of Samos Island (Stamatakis and Zagouroglou, 1984; Stamatakis, 1986). As a result, in the Karlovassi basin, solutions rich in B and K may have altered pre-existing zeolites to B-bearing K-feldspar. This alteration probably took place in the latest stage of lake evolution in a highly saline-alkaline environment.

## POSSIBLE ECONOMIC VALUE

Aside from its uses as raw material in certain industries, K-feldspar could be used as an indicator mineral in

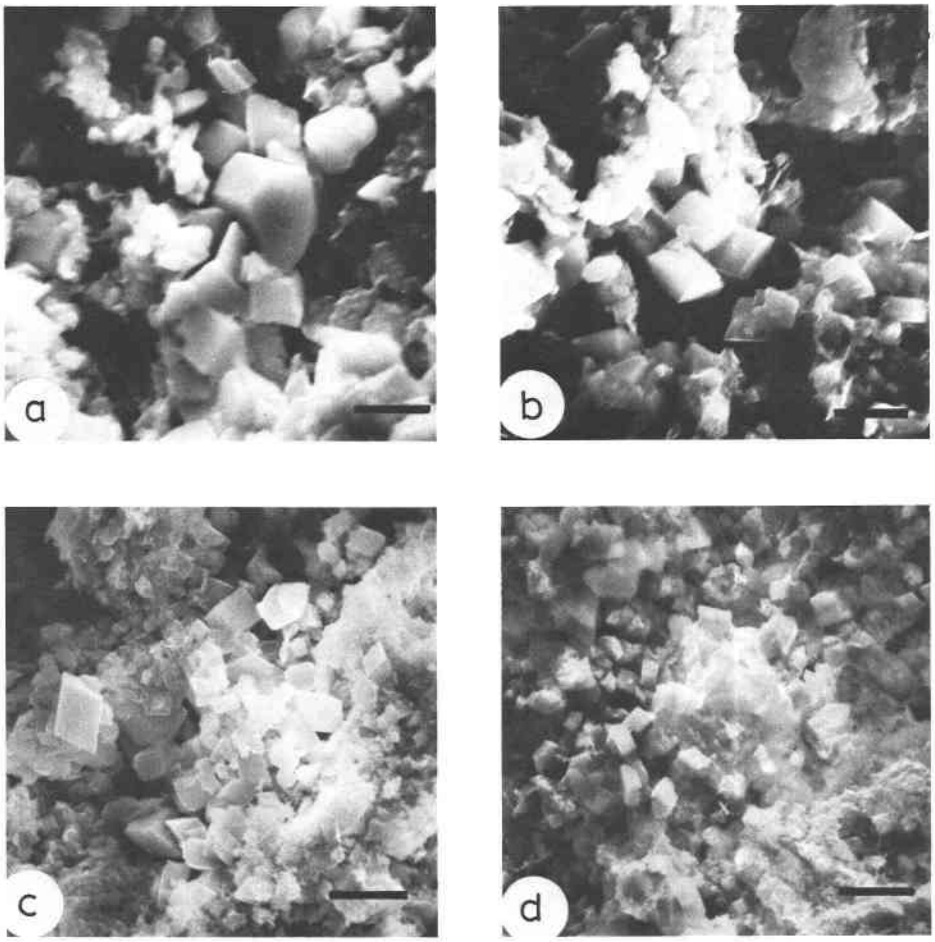

Fig. 4. SEM photomicrographs of authigenic K-feldspar from the Karlovassi basin. Idiomorphic, tabular, and rhombohedral crystals are observed. Scale bars represent 4 $\mu$m on **(a)**, **(b)**, and **(c)** and 10 $\mu$m on **(d)**.

prospecting for evaporite minerals, as it has a genetic association with sulfates, borates, soluble carbonates, and chlorides (Sheppard and Gude, 1972, 1973b; Smith et al., 1983). The K-feldspar of the Karlovassi basin displays a close spatial relationship to the gypsum, celestite, and scant thenardite found in surface exposures of the LNU (Stamatakis, 1986). It is possible that there exists a further association of B-rich K-feldspar with other evaporites, notably borates in buried saline layers, especially those in lithostratigraphic correlation with the adjacent west Anatolian borate deposits (Helvaci, 1986).

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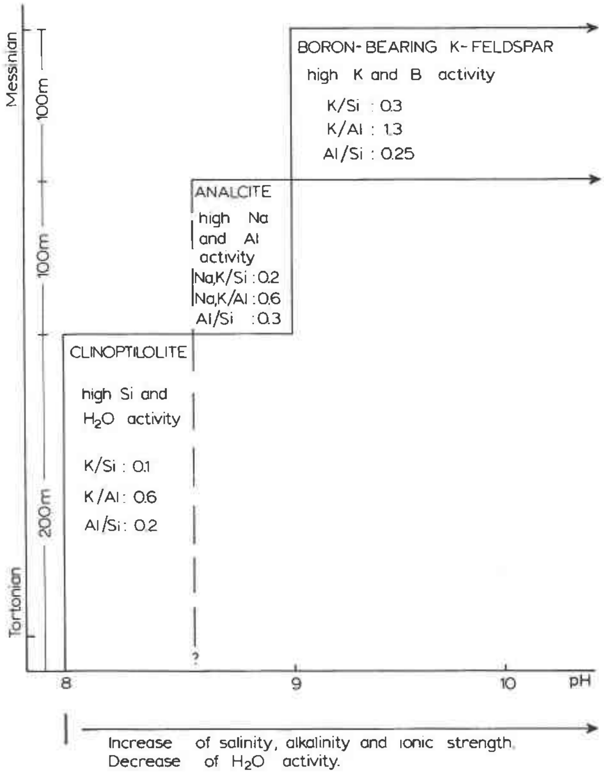

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Fig. 5. The evolution of authigenic silicates in the Karlovassi basin.

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