LINEAR ALKALI CORRELATION IN OCEANIC ALKALI BASALTS

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A strong positive correlation was found between Na_2O and K_2O in basic alkali rocks from the Hawaiian Islands and Canary Islands. The correlated variance of the variables in each province is 79 % and 76 %, respectively. Six possible explanations were examined. (1) Buffer control by phases such as a sodium-bearing clinopyroxene and a mica or a single phase such as an amphibole is possible, but each buffer has limitations. (2) The phase-equilibrium relations at high pressures may also constrain the alkali ratio in these rocks, but the relevant complex system has not as yet been investigated. (3) Metasomatism, resulting in the addition or subtraction of materials, is not considered appropriate because of the high concentration of K relative to Na at high temperatures in the fluid phase. (4) The behavior of Na and K as incompatible trace elements may result in a fixed ratio of alkalies in magmas produced from batch melting. (5) Mixing of a fractionally derived early magma with subsequent magmas, formed either at the same invariant point or from an independent source, may generate a series of mixed magmas with a linear relationship between incompatible elements. (6) The positive correlation of alkalies may result from the new closure (that is, the new set of means and variances characteristic of any set of extracts) attending separation of magma from its parent. No satisfactory explanation of the strong alkali correlation has been obtained. The feature, resulting from one or several mechanisms, may be common to other oceanic island rock series as well as to some calcalkaline sequences.

Key words: alkali correlation, Oceanic alkali basalts, Hawaiian Islands, Canary Islands.

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Introduction

Various investigators have advanced a wide variety of mechanisms by which alkali-rich basalts may be generated (Table 1). Because of the apparent necessity of deriving a liquid enriched in alkalies from a presumably alkali-poor source, *e.g.*, garnet peridotite, the mechanisms commonly involve addition of alkali-rich material to, or extraction of alkali-poor material from, the source prior to melting. The derivation of alkali basalt magma from alkali-enriched sources has recently been revived on the basis of trace-element and isotopic data. Whatever the mechanism, it should operate world wide and provide sufficient volumes of liquid to account for the alkali-rich floods of the Hebridean province, Canary Islands, Saudi Arabia, and Syria, for examples.

In spite of the considerable overlap in alkali

Table 1. Mechanisms proposed by various authors for production of alkali-rich magma.

Source Requirement	
Alkali-rich source	Becke (1903), Barth (1936)
Metasomatized parent	von Eckermann (1948)
Source Modification	
Metasomatic enrichment	von Eckermann (1942), Gummer and Burr (1943), Tilley (1952)
Biotite or amphibole decomposition	Goldschmidt (1922), Bowen (1928), Holmes and Harwood (1937)
Volatile transfer	Smyth (1913, 1927), Bailey (1966, 1970)
High H_2O pressure	Bultitude and Green (1968)
High CO_2 pressure	Eggler (1974), Bailey (1974), Brey and Green (1975), Mysen and Boettcher (1975)
Magma Modification	
Assimilation	Daly (1910), Shand (1922), Tilley and Harwood (1931)
Biotite or amphibole resorption	Bowen (1928)
Desilication	Shand (1922)
Phase separation	
Orthopyroxene extraction	Holmes and Harwood (1932), Powers (1935), Larsen (1940)
Hematite extraction	Bailey and Schairer (1966)
Immiscible liquid separation	Marshall (1914), Holgate (1954)
Physicochemical Processes	
Invariant point melting	Schairer and Yoder (1964), Yoder (1978)
Incongruent melting	Bowen (1928), Tilley (1958)
Zone refining	Harris (1957)
Fractionation	
Fractional melting	Waters (1955)
Liquid fractionation (diffusion)	Hamilton (1965), Kogarko *et al.* (1974)
of eclogite	O'Hara and Yoder (1967)
at high pressure	Yoder and Tilley (1961), Kushiro and Kuno (1963), Kushiro (1968)
Mechanical Processes	
Filter pressing	Harker (1909), Bowen (1915)
Tectonic control	Harker (1912), Geijer (1922)

content of alkali basalt and tholeiites, a distinction was made between the two magma types by Macdonald and Katsura (1964; see also Macdonald 1968) on the basis of »positive microscopic evidence.» They cited »the presence of titanian augite, of true groundmass olivine, and of interstitial alkali feldspar» as characteristic of alkali basalt. They applied these criteria to the rocks of Hawaii, and drew a definitive line between the rock types in a (K_2O + Na_2O) *vs.* SiO_2 plot (Fig. 1). Even though the line may vary in position from petrographic province to province, the alkali basalts tend to be richer than tholeiites in alkalies for a given silica content. On the other hand, there are representatives of each basalt type having the *same* alkali content. It is to be noted that on the original Macdonald-Katsura diagram compositions plotting in the vicinity of the proposed boundary for Hawaiian rocks are less abundant than those more distant from it. And, with increasing SiO_2, each basalt type appears to diverge in composition from the dividing line, beginning at a central composition near *A* of Fig. 1. Although one might wish to jump to the conclusion that such a central composition defines the common source, it should be recalled

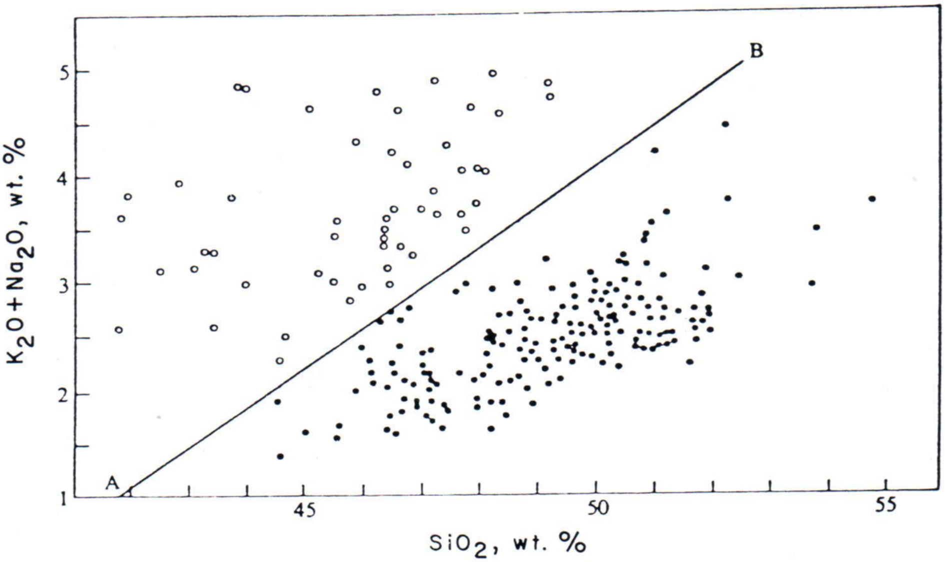

Fig. 1. Alkali vs. silica for Hawaiian basaltic rocks (Macdonald and Katsura 1964). Line *A—B* divides alkalic rocks (above *A—B*) and tholeiites (below *A—B*) on the basis of mineralogical criteria.

that each magma type may be generated at different pressures and subtle differences in physicochemical relations may have been attained at the time of magma separation.

The Macdonald-Katsura dividing line contains compositions that are subparallel with the plane of critical undersaturation as defined by Yoder and Tilley (1962). That is, the join Fo-Ab in the K-free model system used by Yoder and Tilley, which separates in principle the nepheline-normative rocks from the tholeiitic or subalkaline rocks, is close to and almost parallels the Macdonald-Katsura dividing line (*cf.* DeLong and Hoffman 1975). Because of solid solutions, especially those in the clinopyroxene, and possibly oxidation state of iron, many alkali basalts are »transitional» and have a small amount of hypersthene in the norm. In fact, more than half of the original Hawaiian Island alkalic basic rocks described by Macdonald and Katsura lack normative nepheline, and this is also true for 41 % of 221 Hawaiian basaltic rocks in RKOC76 (Chayes *et al.,* 1977) that plot above their dividing line. Nevertheless, the nepheline-normative character of alkaline rocks is a useful concept in regard to the interpretation of phase equilibria, and has recently been proposed by the IUGS Subcommission on Systematics and Igneous Petrology as a diagnostic feature of major importance. Conversely, other solid solutions result in the absence of *modal* nepheline (or analcime, alkali-rich glass) until considerable *normative* nepheline (> 7 %) is present in an alkaline basalt. For this latter reason, basanite, a term now frequently applied to rocks by definition *requiring* but in fact often lacking modal nepheline, has been included in the data reductions used here.

The close proximity of the critical plane of undersaturation, determined from a consideration of phases high in Na_2O (*i.e.,* albite, nepheline), to the Macdonald-Katsura dividing line in which Na_2O and K_2O are treated together, as apparently first proposed by Iddings (1898), implies an unusual role for K_2O. Either K is indeed a proxy for Na or its thermochemical properties are at least similar to those of Na. It was this relationship that motivated the authors initially to examine the relations between alkalies.

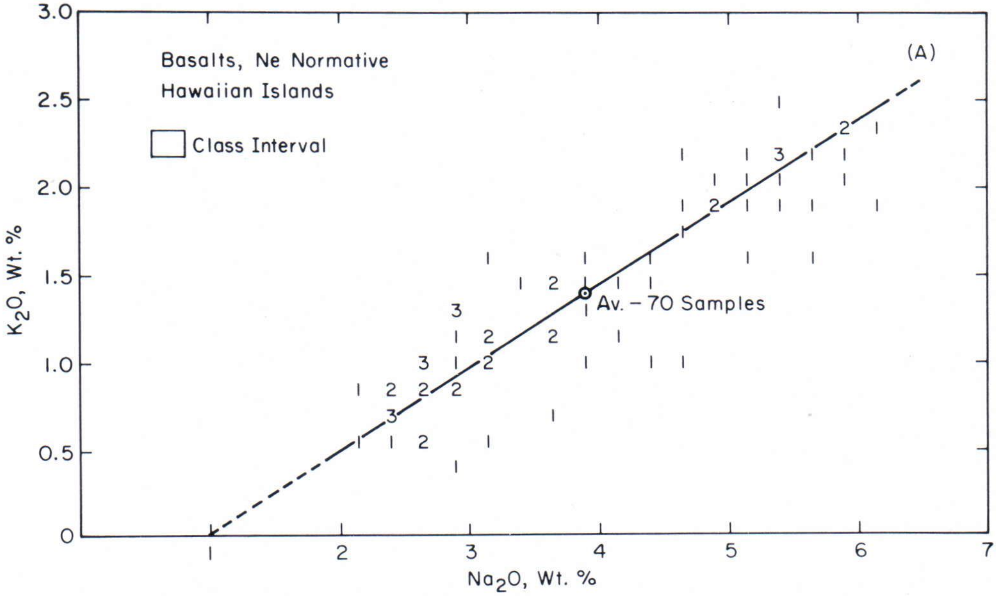

Fig. 2A. Plot of K_2O *vs.* Na_2O for Ne-normative basalts from the Hawaiian Islands. Line is reduced major-axis regression. Numbers indicate sample frequency in class interval illustrated (0.15 K_2O and 0.25 Na_2O wt. %). $r = 0.891$.

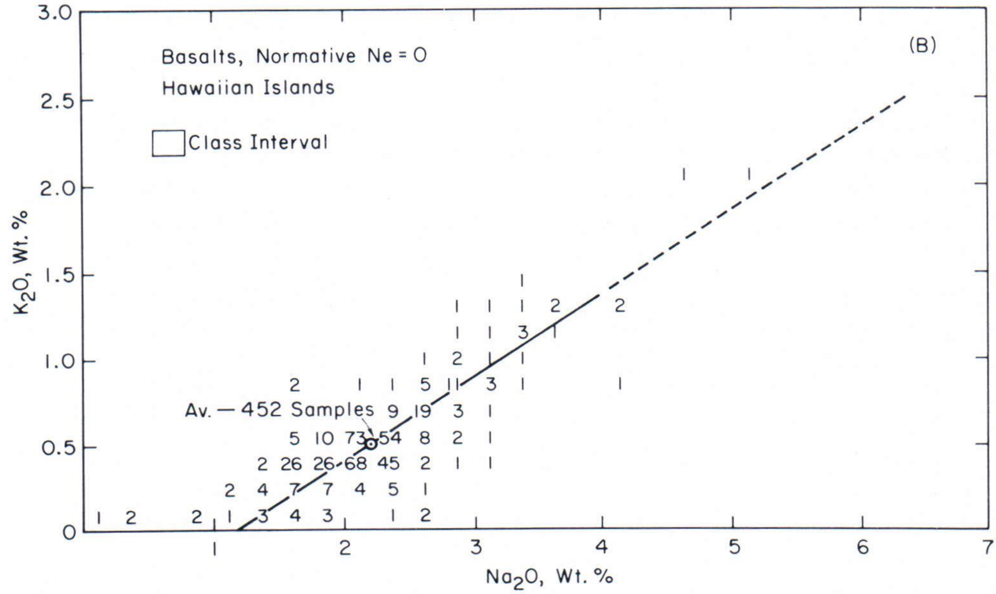

Fig. 2B. Plot of K_2O *vs.* Na_2O for Hy-normative basalts from the Hawaiian Islands. $r = 0.792$.

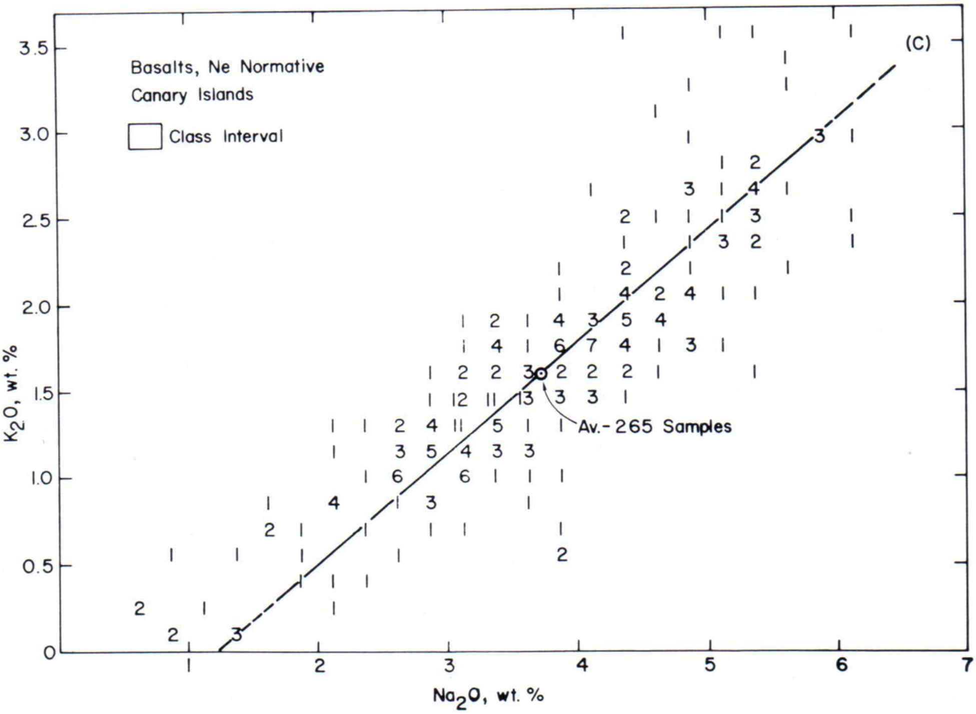

Fig. 3. Plot of K_2O vs. Na_2O for Ne-normative basalts from Canary Islands. See Fig. 2 for explanation. $r = 0.874$.

Statistical analysis

The study is confined to two areas in the world for which the greatest numbers of chemical analyses per unit area of igneous rocks are available, the Hawaiian Islands and the Canary Islands. The data bank used in the statistical analysis of the chemical analyses is RKOC76; acceptable chemical analyses were limited to those with $SiO_2 > 42$, $H_2O < 1$, and $Na_2O < 6.5$ wt. %. The data bank contains 70 such Ne-normative analyses from the Hawaiian Islands and 265 from the Canary Islands. In addition, 452 analyses lacking normative Ne were retrieved from the Hawaiian Island file. Plots of K_2O *vs.* Na_2O for these sets of chemical analyses are given in Figs. 2A, 3, and 2B, respectively. The frequency of occurrence of samples within classes 0.15 K_2O and 0.25 Na_2O wt. % wide is displayed for each data set. The reduced major axis line

$$Y - \bar{y} = \frac{\sigma_y}{\sigma_x}(X - \bar{x}),$$

where Y is K_2O, X is Na_2O, and σ is the standard deviation, is also shown in the diagrams. (The reduced major axis line is used because it describes interdependence without designation of the dependent and independent variables. There is no basis in the present petrological application for making the designation.)

The correlation of Na_2O with K_2O for the data set of Hawaiian nepheline normative rocks is 0.891, that is, the correlated variance of the variables is 79 %, whereas for alkalies in the tholeiitic rocks, it is close to 63 %. For the nepheline normative rocks of the Canary Islands, 76 % of the variance of either variable may be described as a linear function of the other. (There are too few tholeiitic rocks in the Canary Islands for statistical analysis.) The near coincidence of the regression lines for the Hawaiian rocks is noteworthy. On the other hand, the slope of the regression line for the Canary Island rocks is quite different from that of the Hawaiian petrographic province. For all three regression lines the intercept is about 1 % Na_2O, a possibly fictive feature with puzzling implications that will be discussed below.

Discussion

The substantial positive linear correlation between alkalies in both alkaline and tholeiitic rocks from the most frequently analyzed regions in the world raises a fundamental question. How can the slope of K_2O *vs.* Na_2O remain relatively constant through the generation process, or the fractionation process, or both? The early appearance of plagioclase (Yoder and Tilley 1962, Tilley *et al.* 1965, Tilley and Thompson 1972) in a relatively low-pressure fractionation process, for example, would preclude a constant relationship of the alkalies. Six possible explanations are examined to account for this relationship.

Buffer control

On the assumption that these rocks are the partial melting product of a parental material such as garnet peridotite, the Na_2O might be buffered by the jadeite component of the clinopyroxene, and to a minor extent by the Na content of pyropic garnet. The K_2O component, however, does not have an obvious buffer, even though one omphacite inclusion in diamond was found to contain 1 % K_2O (Hall and Smith 1984) — most pyroxenes from nodules contain trace amounts of K_2O. (The K in such pyroxenes may be assigned to a potassic acmite, $KFe^{3+}Si_2O_6$, or a potassic titanaugite, $KMg_{0.5}Ti_{0.5}Si_2O_6$, end member.) There are several additional potential candidates. Sanidine is stable at high pressures and is presumed to be primary in eclogite nodules found in some kimberlite pipes (Nixon 1960). Its absence as a primary phase in other nodules is probably due to the reaction

$$\mathrm{Sa} + 2\mathrm{Fo} \rightarrow \mathrm{Ks} + 4\mathrm{En}$$

at pressures less than those of the geotherm (Wendlandt and Eggler 1980b, p. 450, Fig. 10). The product kalsilite (Ks) would probably be held in solid solution in nepheline. Nepheline is believed to coexist with enstatite (En) *metastably* at 1 atm (Yoder 1952, p. 586; Schairer and Yoder 1961, p. 143; Chinner and Schairer 1962) and at 9 kbar (Yoder 1964), and *stably* from 11 to 24 kbar (Kushiro 1965); however, these latter findings were not confirmed (Windom and Boettcher 1981). Hypersthene is found in nodules in nepheline-bearing basalts, but the stable coexistence of hypersthene and nepheline at the high pressures where the nodules and basaltic liquid are presumed to have originated has yet to be demonstrated.

The high-pressure, hydrated form of sanidine, $KAlSi_3O_8 \cdot H_2O$, synthesized by Seki and Kennedy (1964) has not been observed in nodules.

Phlogopite could also serve as a buffer, but the small proportions compatible with the modes of nodules and heat-production constraints would probably be consumed in the initial melt. Possible phlogopite buffering relations are outlined schematically in a diagram (Fig. 4) deduced in part from Luth (1967) and Modreski and Boettcher (1973). Compositions representative of part of the parental material lie in the phase triangle Fo + En + Phlog, an assemblage that begins to melt at point *P*. The composition marked with a star, for example, which contains 10 % Phlog, would yield only 6 % liquid under equilibrium conditions before phlogopite would be consumed. A diagram of similar topology would probably be valid also for a potassic amphibole, e.g., kaersutite, stable at pressures less than about 35 kbar.

Dr. Eric Middlemost (personal communication, 1982) suggested that the removal of a phase such as kaersutite, or possibly melilite, some species of which contain both Na and K, from the

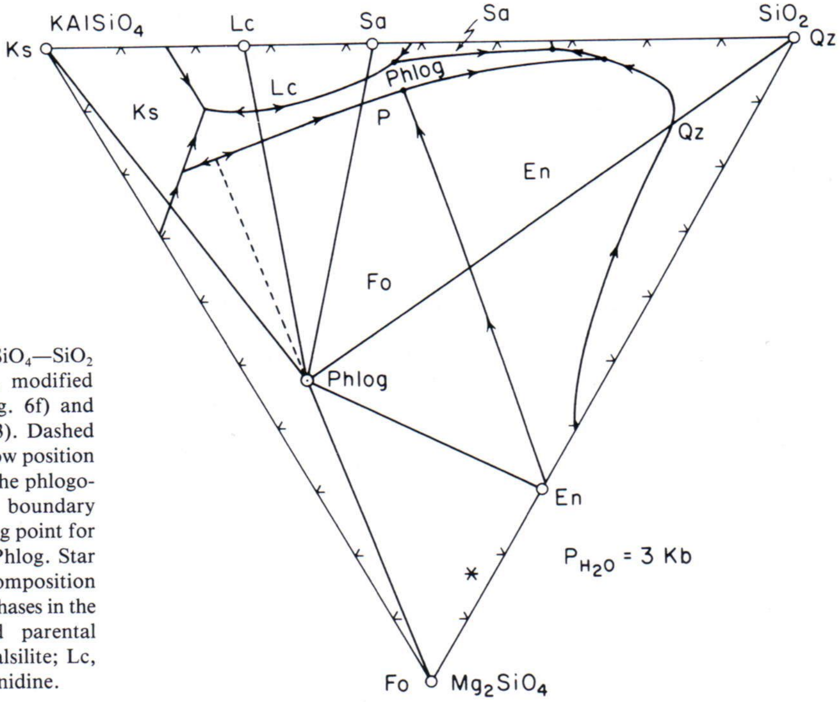

Fig. 4. The $KAlSiO_4—Mg_2SiO_4—SiO_2$ system for $P_{H_2}O$ = 3 kbar, modified from Luth (1967, p. 397, Fig. 6f) and Modreski and Boettcher (1973). Dashed line is a construction line to show position of maximum temperature on the phlogopite (Phlog)-forsterite (Fo) boundary curve. *P* is beginning-of-melting point for the assemblage Fo + En + Phlog. Star is projection of one possible composition illustrating same of the major phases in the assemblage of the assumed parental material. En, enstatite; Ks, kalsilite; Lc, leucite; Qz, quartz; Sa, sanidine.

magma may be significant in maintaining a linear relationship between Na_2O and K_2O. The amphibole has a restricted stability range, and in view of its limited content of alkalies, partitioning into the liquid would of necessity be substantial. Kaersutite is Ne and Lc normative. An amphibole such as potassic richterite, observed in some nodules, has been shown to be unstable in the presence of garnet and aluminous pyroxenes (Kushiro and Erlank 1970, p. 233), and would not be expected in the parental materials now believed to be the principal source of basaltic magmas.

The presence of H_2O, required for the formation of phlogopite or amphibole, results in partial melts that are Qz normative (Modreski and Boettcher 1973). That is, alkali enrichment in the initial liquids would be possible in the presence of those phases, but only subalkaline magmas would be formed. On the other hand, generation of silica-undersaturated melts may result from the presence of CO_2 (Kushiro 1968, p. 625, Fig. 4; Eggler 1978). For this reason, potassium-bearing carbonates may be a possible buffer. Unfortunately, CO_2 tends to destroy the critical assemblage of olivine and clinopyroxene in the mantle. According to Wyllie and Huang (1975) and Eggler (1975), the reaction Fo + Di + $CO_2 \rightarrow$ En + Dol takes place. If excess CO_2 is available, the parental assemblage then becomes olivine + orthopyroxene + garnet + dolomite. Dolomite has been searched for in nodules and appears to be absent. The two known inclusions of dolomite in diamonds are believed to be secondary. It thus seems unlikely that a potassium-bearing carbonate serves as a buffer in the generation of alkaline magmas.

Phase equilibria

Other potential buffers for potassium include nepheline and kalsilite. Both are stable to high pressures, yet neither has been found in nodules. The Buerger composition for nepheline solid

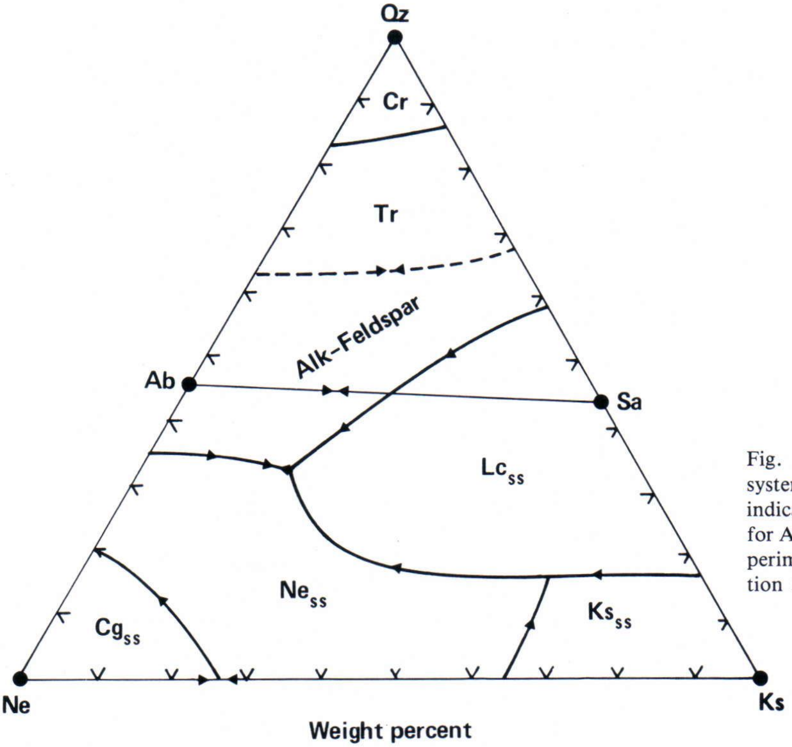

Fig. 5. The $NaAlSiO_4$—$KAlSiO_4$—SiO_2 system of Schairer (1957). Minima are indicated by opposing arrows. Minimum for Ab-Sa-Qz has not been determined experimentally and is estimated by projection for the same system determined at various water pressures.

solution contains 1K for every 3Na. This particular ratio, characteristic of plutonic rocks, is also close to the minimum melting compositions in Ne-Ks at 73 wt. % Ne and in Ab-Sa at 65 wt. % Ab (Fig. 5), and appears to hold irrespective of the silica content. It is noteworthy that Na_2O and K_2O are the only major oxides positively correlated with SiO_2 in multicomponent associations. The minima of rhyolites, trachytes, and nepheline trachytes in the nepheline-kalsilite-quartz system all have roughly the same ratio of Na_2O and K_2O. The relationship of Na_2O to K_2O in the alkali basalts may, therefore, merely reflect persistence at high pressure of the minimum melting relations in the simple systems.

The shifts of the Fo-En boundary curve and the beginning-of-melting point for the initial assemblages Fo + En + Ab and Fo + En + Sa are displayed, respectively, in Figs. 6A and 6B. It appears that, in general, the relative proportion of alkalies in the initial liquids generated from an appropriate mantle assemblage might be preserved until the pressure is reached at which jadeite becomes stable. At that pressure the initial liquid becomes picritic and, more importantly, poorer in Na. Experimental investigation of the relevant beginning-of-melting point in the Ne—Ks—Fo—Qz system at high pressures will be required to ascertain quantitatively the variation of alkalies in the initial liquids with depth. It is worth emphasizing again that the shift of normative character of the initial melts from Qz to Hy to Ne, displayed in Figs. 6A and 6B, is a pressure effect and is *not* dependent on the degree of melting, although in discussions of the generation of magmas the latter concept is often said to account for it. The continuous nature of the change in invariant points with pressure is compatible with the observed continuum in alkali content between alkali basalt and tholeiite compositions. The alkali basalts appear to be much more abundant in Cenozoic and very late Paleo-

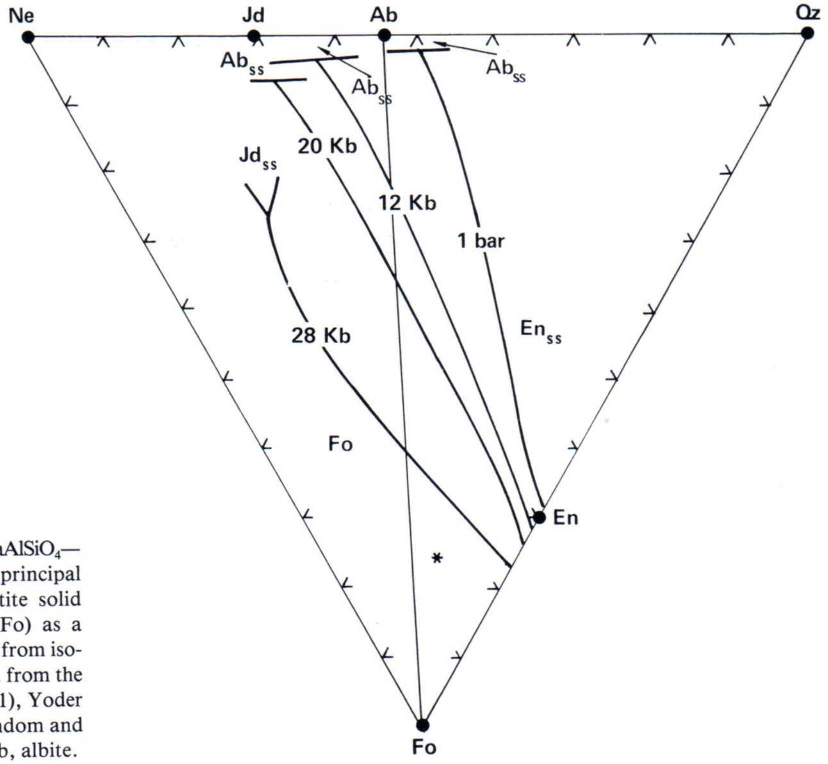

Fig. 6A. The system Mg_2SiO_4—$NaAlSiO_4$—SiO_2 illustrating the shift of the principal boundary curve between enstatite solid solution (En_{ss}) and forsterite (Fo) as a function of pressure. Completed from isobaric phase diagrams constructed from the data of Schairer and Yoder (1961), Yoder (1964), Kushiro (1968), and Windom and Boettcher (1981). Jd, jadeite; Ab, albite.

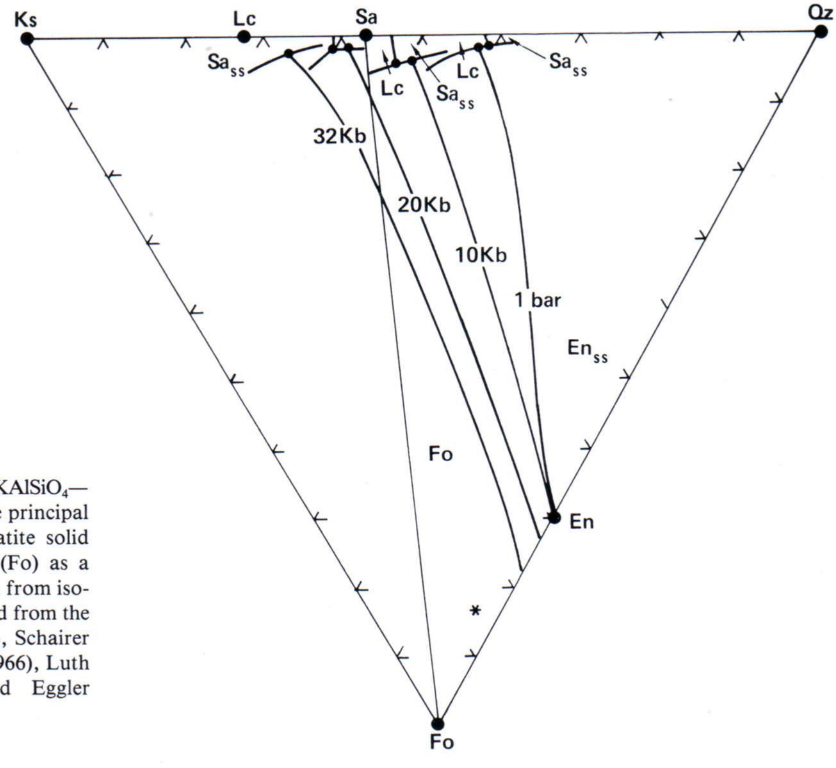

Fig. 6B. The system Mg_2SiO_4—$KAlSiO_4$—SiO_2 illustrating the shift of the principal boundary curve between enstatite solid solution (En_{ss}) and forsterite (Fo) as a function of pressure. Compiled from isobaric phase diagrams constructed from the data of Schairer (1954, p. 506), Schairer and Bowen (1955), Lindsley (1966), Luth (1967), and Wendlandt and Eggler (1980a, b).

zoic volcanic assemblages than in those of early through mid-Paleozoic age (Poldevaart 1962). If the production of alkali-rich melt fractions is enhanced by high pressures, then it would appear that some sources of younger basaltic magma are quite deep seated.

Metasomatism

The preferential partitioning of alkalies into the gas phase may also account for the correlation of K_2O and Na_2O in magmas. Although the partitioning coefficients between crystal, liquid, and gas in alkaline magmas are not known, Mysen (1981, pp. 347—349; personal communication, 1982) has suggested, on the basis of the enrichment of light rare earths, that at high temperatures the gas phase is enriched in K relative to Na with increasing pressure. Metasomatism, therefore, might lead to the more exotic varieties of potassium-rich rocks but not necessarily to alkali basalt. Furthermore, Wendlandt and Eggler (1980a, p. 409) noted that in the Lc-normative rocks there is little correlation between K_2O and Na_2O, as might be anticipated in a metasomatic process dependent on highly variable parameters such as permeability and porosity. Metasomatic processes, therefore, probably would not result in products with the observed correlation between alkalies.

Trace element behavior

A strong correlation of Na and K may result if they are essentially incompatible elements during the partial melting of the parental material. On the assumption that both almost completely partition into the liquid, the correlation of these elements is fixed primarily by their ratio in the parental material (*cf.* Frey *et al.* 1980). Continuous invariant melting where the liquid remains in equilibrium with the parental phases would merely result in dilution. If a magma were removed from the batch, it would retain the initial ratio with a concentration dependent on the stage of dilution. With extreme incompatibility, fractional melting — that is, continuous separation of liquid from the parental phases — would yield only one alkali-enriched magma; subsequent magmas would be essentially devoid of alkalies. Such a partial melting process would not yield a series of alkaline magmas.

The positive intercepts for Na_2O in the Na_2O—K_2O regressions indicate that Na is less incompatible than K. As K tends toward zero, apparently one or more phases persist for which the partitioning of Na into the liquid is of a value such that some Na is retained in the parental material. It is interesting to note that the volatile-free compositions of chondritic meteorites also reflect the same constraints, both oxides tending toward zero only after Na_2O drops below ~ 1 wt. % and K_2O, below ~ 0.1 wt. % (Haramura *et al.* 1983, p. 115).

The similarity of the intercepts for the Hawaiian and Canary Island data suggests that the alkali compositions of the source regions of the two areas are similar. The dissimilarity of the slopes indicates that the parental assemblages, the proportions of phases, or the partitioning coefficients are different as a consequence of differing conditions of magma separation. The near coincidence of slope and intercept for the Hawaiian alkaline and tholeiitic rocks, however, presents a dilemma. The implication is that the sources are similar, the parental assemblages are the same, and the effective partitioning coefficients are essentially the same — even though there are many reasons to believe that the two magma series were generated as a result of major differences in one or all of the cited circumstances. Those differences may be sufficiently subtle at the site of generation to be obscured by the relatively lower correlations of alkalies of the Hawaiian tholeiitic rocks. Other petrographic provinces yielding adequate analytical data with high correlations for both rock series may provide resolution of this dilemma. On the other hand, the proposed hypothesis wherein the alkalies are assigned the role of a trace element may not be applicable.

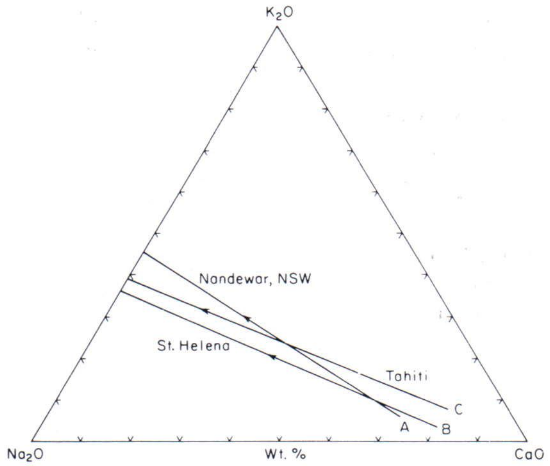

Fig. 7. Trend of Na_2O, K_2O, and CaO content of rocks from Nandewar, New South Wales (Abbott 1969, p. 130); St. Helena Island, South Atlantic (Baker 1969); and Tahiti (McBirney and Aoki 1968, p. 545). The arrows indicate the direction of liquid fractionation on the basis of iron enrichment, if that process were applicable. The ends of the lines are representative of compositions of magmas required for mixing, if that process were applicable.

Magma mixing

A linear relationship between elements has led many investigators to conclude that some magmas are derived by the periodic or continuous mixing of two separate magmas. The strong linear component of variation of CaO in the ternary CaO—Na_2O—K_2O illustrated in Fig. 7 is a feature common to other oceanic island rock series and applies as well to some continental calcalkaline sequences. Fractional melting of the type described above, in which the alkalies behave as incompatible trace elements, for example, may yield an early magma highly enriched in alkalies that could then after separation mix in various proportions with a subsequent batch melt produced at the same invariant point, or with a magma from an independent source. The volume relations would not necessarily be appropriate, but the concept appears to account for a linear correlation of incompatible elements. It remains to be demonstrated, however, that the alkalies are indeed incompatible elements. The linear correlation seems all the more remarkable in the light of the many variables that may influence the mixing of magmas after they leave the site of generation (see, *e.g.*, O'Hara 1977; O'Hara and Mathews 1981).

Closure correlation

It has been shown (Chayes 1982, p. 245; see also Chayes and Kruskal 1966, p. 700; Chayes 1971, pp. 39—40) that there may be strong positive correlation between any pair of trace elements, or other variables of small initial variance, in successive melt fractions even if these elements are uncorrelated with each other in the parental material. The correlation results because the partial melting imposes new closure conditions — a new set of means and variances — on the melt fraction. It is even possible that pairs of trace elements characterized by a strong negative correlation in the source may be strongly positively correlated in melt fractions. Chayes emphasizes that strong positive correlations are to be expected among trace elements and may convey little or no information about covariances between these variables in the parent. The argument fails if successive magmas are formed strictly at an invariant point; if the melts vary in major element composition, however, the positive covariances between trace elements may be merely the numerical consequence of the closure inherent in the process by which the magma is separated from its parent. On these grounds the strong correlation between K_2O and Na_2O may be a direct result of magma separation.

Summary

The slope of Na_2O *vs.* K_2O appears to be relatively constant in basalts from the Hawaiian Islands and the Canary Islands. This relationship in basic alkaline magmas may result from one, or a combination, of the following factors:

1) buffering by appropriate Na- and K-bearing phases in the parental material;

2) phase relations governing the proportion of Na- and K-bearing phase(s) stable in basic alkaline magma;

3) behavior of Na and K as strongly incompatible elements,

4) magma mixing from a common or independent source, or

5) positive correlation imposed by the new closure attending partial melting and magma separation.

Such a regular relationship is not believed to result from the addition or subtraction of materials in a metasomatic process. At this time, there does not appear to be an unambiguous common solution to the alkali-ratio problem; however, one or some combination of several solutions may apply to a particular petrological province.

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