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LOW GRADE ALTERATION OF BASIC ROCKS IN HOKKAIDO, JAPAN.

by

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(with 9 text-figures)

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Abstract

Mesozoic basalt flows, tuffs and diabases are widely distributed in Hokkaido and show a continuous compositional change from initial low temperature sea water – basalt chemical exchange to the development of rocks with spilite characteristics during very low grade prehnite-pumpellyite burial metamorphism. This change involves net addition of K, P, Na, Si, H_2O and oxidation of FeO and net loss of Fe, Mg and Ca. More extensive alteration (i.e. spilitization) occurs when calcic plagioclase inverts to albite and free Ca, in particular, combines with CO_2 and is redistributed as calcite. The least altered rocks are olivine-hypersthene or quartz normative tholeiites and have chemical characteristics of oceanic-type basalts. The association of serpentine and metagabbro with some of the basic rocks indicates that they are part of an obducted and disrupted ophiolite suite.

Introduction

Basaltic rocks in Hokkaido are associated with a thick sequence of pelitic and psammitic sediments of probable Lower Triassic – Upper Jurassic age which form the axial belt of Hokkaido extending north into Sakhalin. The distribution of the basic rocks is given in Fig. 1. The geosynclinal sequence has been broadly divided into three main groups; Nakanogawa, Kamui and Sorachi Groups in ascending order. Basic rocks associated with the Nakanogawa sediments are represented by diabase dykes and sills and are best developed as the sheeted diabase complex at Shimokawa (Fig. 1), (Miyake, 1965; Hunahashi et al. 1969) extending for some 20 km in length and attaining a maximum thickness of about 1.5 km. Such rocks represent shallow intrusive equivalents of the overlying lavas and tuffs. Basic rocks in the Upper Kamui and Sorachi Group sediments are characterized by a great predominance of frequently pillowed lavas over hyaloclastics and are associated with radiolarian cherts, slate, sandstone and fossiliferous limestone. Due to intensive folding and faulting associated with serpentinite intrusion in the western part of the

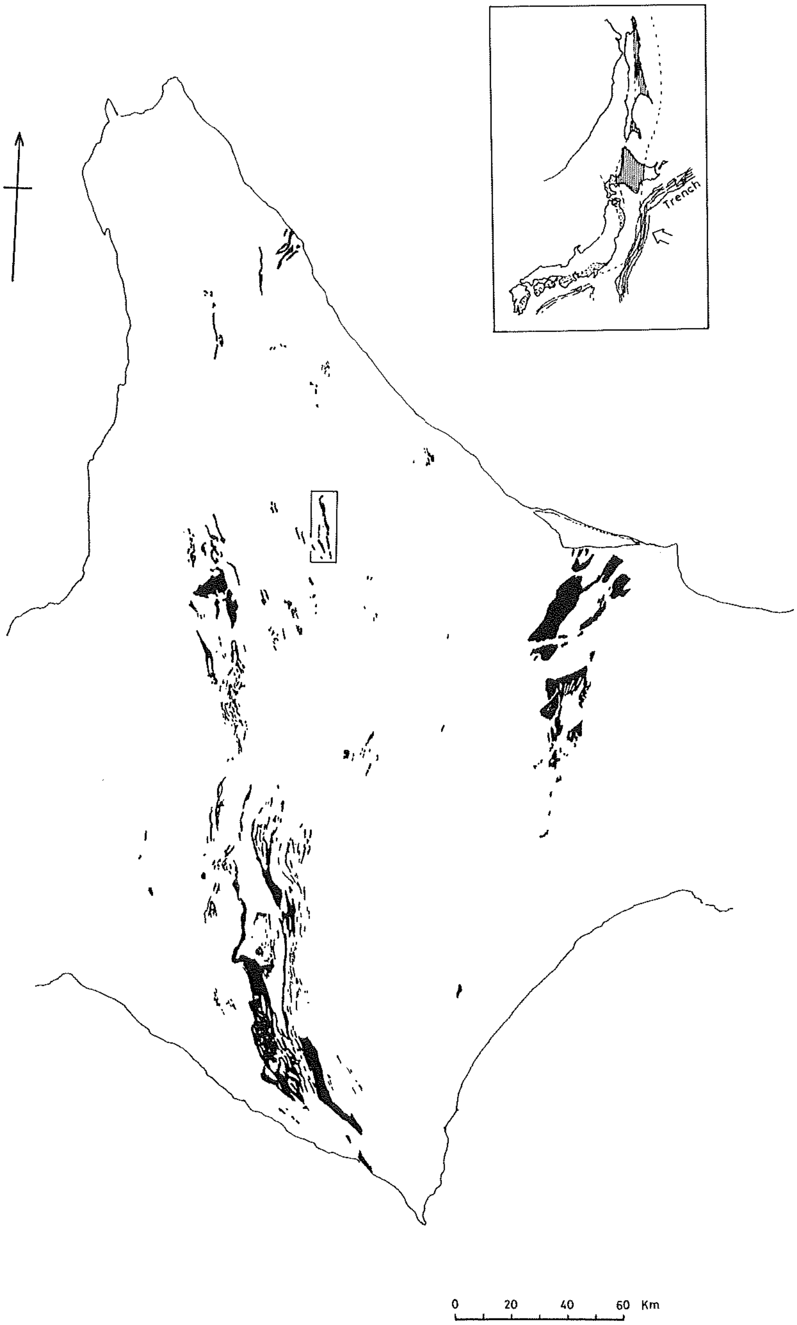

Fig. 1. Map of the distribution of Mesozoic basic rocks in Hokkaido. The square marks the exposure of the sheeted diabase complex at Shimokawa.
In the inset map the dotted area represents the present distribution of the Sambosan rocks and the striped area represents the present distribution of rocks of the Hokkaido-Sakhalin trough.

geosyncline during Tertiary time the thickness of the basic flows and tuffs is unknown. The Upper Jurassic basaltic rocks in N.E. Hokkaido are relatively undeformed and are estimated to be about 2,300 m thick (Teraoka et al. 1973).

The distribution, mineralization and general chemical characteristics of the basic rocks have been discussed by numerous authors, e.g. Suzuki (1954), Igi (1956), Suzuki (1963); Bamba and Sawa (1967), Bamba and Maeda (1969), Sawada and Kanmera (1973), Bamba (1974). This paper deals with the chemical changes of the rocks during submarine weathering and burial metamorphism in a geosynclinal pile. Later polyphase metamorphism accompanied by the tectonic intrusion of serpentinite and localised metamorphism of the basic rocks to greenschist (+ glaucophane schist) or epidote amphibolite and/or thermal metamorphism to brown hornblende amphibolite during the Tertiary Alpine-type orogenic phase in Hokkaido will be discussed in another paper now in preparation.

Sequence of Progressive Alteration

Primary textures are generally well preserved in the basic flows and diabases although varying amounts of mineralogical reconstitution is nearly always present, particularly in the hemicrystalline parts of the rocks. On the basis of mineralogy and bulk chemistry a continuous change from early stage low temperature submarine weathering to the development of metabasites with spilite characteristics is recognized.

Initial Stage Low Temperature Alteration

Initial stage low temperature alteration (halmyrolysis) of the basic rocks is demonstrated in Fig. 2 where the ratio of the element concentration in the glass rim, variolitic transition zone and core of relatively fresh and more altered pillow lavas are plotted. The diagram clearly indicates the *direction of alteration* with increasing (net addition of) K, P, Na and H_2O, oxidation of Fe^{2+} and decreasing (net loss of) Fe, Mg, and Ca. Al and Si do not vary significantly during this stage of alteration. It is notable that Ti and Mn are depleated in the glassy rims.

These trends are essentially similar for all three zones and more pronounced alteration occurs in the glassy rims. The results are in harmony with other studies, both natural and experimental, of low temperature chemical exchange between sea water and basaltic lavas, e.g. Miyashiro et al. (1969), Hart (1970), Thompson (1975) and Scott and Hajash (1976).

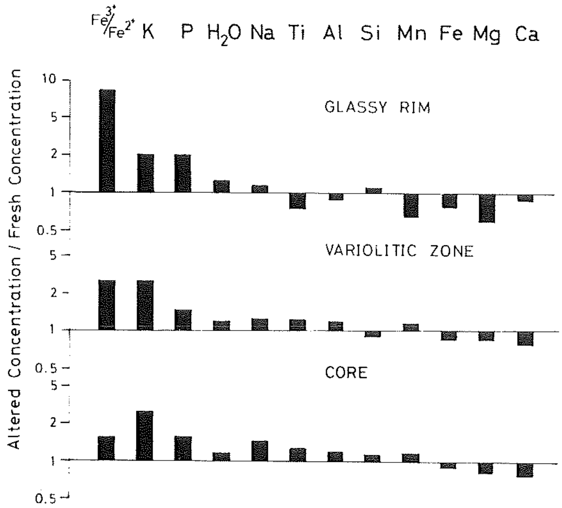

Fig. 2. Comparison of low temperature alteration effects on major element concentration in altered pillow lava relative to unaltered pillow lava from N.E. Hokkaido. Data from Bamba and Sawa (1967) and Bamba and Maeda (1969).

Burial Metamorphism and Spilitization

Progressive alteration of the basic rocks during burial is indicated by the invariable presence of chlorite (largely after glass), albitized, non-recrystallized plagioclase (in some cases containing prehnite, calcite, clinozoisite and pumpellyite) and calcite. These phases may, or may not be accompanied by prehnite and/or pumpellyite (typically in veins and amygdules), white mica, epidote and sphene. Augite is either fresh (?metastable) or is replaced in varying degree by chlorite, occasionally actinolite and rarely (see Sawada and Kanmera, 1973) by sodic amphibole.

It is now generally accepted that spilite genesis is a secondary alteration process controlled by the original basaltic mineralogy and texture (e.g. Vallance, 1965, 1969; Cann, 1967; Graham, 1975) and involves redistribution of chemical components in crystalline basalt and dolerite during burial. Although the effects of sediment cover in preventing chemical exchange between sea water and basalt is little known it is probable that at the substantial depths at which these basalts were presumably erupted and the fluxing ability of water in such low grade mineral reactions, sea water – basalt reactions will, to some extent, take place. In fact the 'spilite' reaction is governed by the instability of calcic plagioclase under low grade burial conditions in the presence of H_2O and CO_2 and inverts to albite by the coupled substitution of NaSi $\rightleftharpoons$ CaAl. As this reaction is a constant volume diffusive replacement the original texture is retained while bulk composition changes appreciably, the *amount of change* depending on the modal amount of plagioclase present.

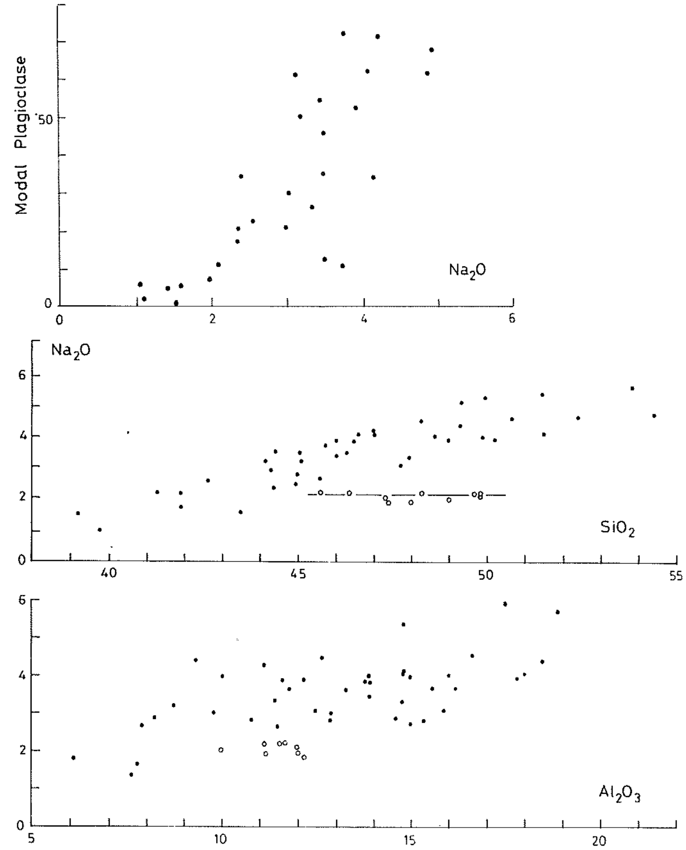

Fig. 3. Plots of modal plagioclase, SiO_2 and Al_2O_3 against Na_2O in Hokkaido basic rocks. Open circles represent relatively unaltered basalt and dolerite with unalbitized plagioclase.

In the Hokkaido basic rocks the above mentioned features are illustrated in Fig. 3. While there are positive correlations between Na and Si and modal plagioclase and bulk Na content, the positive correlation between Na and Al indicates that, unlike Ca, Al largely remains insitu during alteration to form prehnite, epidote, white mica and pumpellyite. It is notable that in unaltered rocks (i.e. calcic plagioclase is preserved) Si, and to a lesser extent Na, vary as a result of initial bulk composition rather than by any alteration process.

Major element histograms for 82 Hokkaido basic rocks are given in Fig. 4.

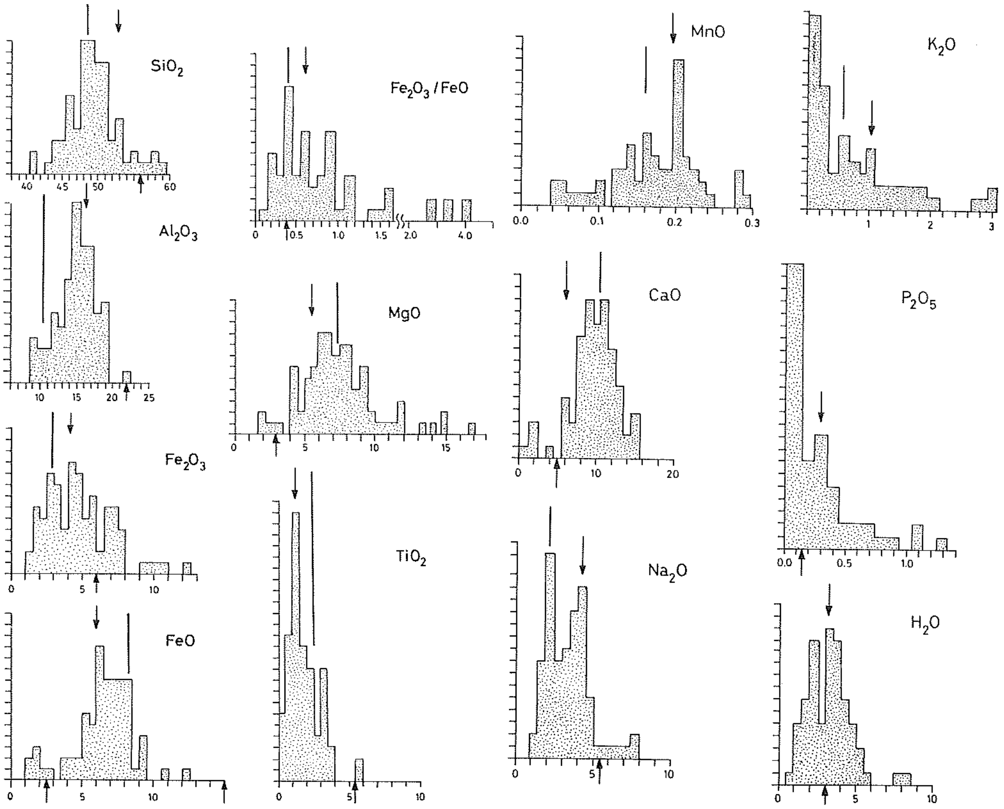

Fig. 4. Histograms of major element distribution in 82 basic rocks of Hokkaido (excluding keratophyres).
Horizontal axis, wt% oxides (volatile-free basis). Vertical axis, frequency of analyses.
Fe_2O_3/FeO scale is not linear at high ratio values. Upward pointing arrow(s) along the horizontal axis indicates the concentration limit for rocks of basaltic composition within the 'chemical screen' of Manson (1967). Downward pointing arrow indicates element concentration in an average analysis of 225 'spilites' (Vallance. 1969). Vertical bar indicates element concentration in an average analysis of 282 oceanic tholeiites (Manson, 1967).

For the most part chemical variation is within the limits of the 'basaltic screen' of Manson (1967) although some overlap occurs, particularly with regard to the higher spread of H_2O, P, Na, Fe^{3+} and Si values, notably higher Fe^{3+}/Fe^{2+} ratios, and lower spread of Ca, Mg and Fe^{2+} values. The bimodal distribution of Na, P and H_2O is significant with the higher maxima almost exactly corresponding to the average spilite values given by Vallance (1969). The large, relative spreads of K and P with respect to the basalt chemical screen indicates that these elements were readily redistributed during burial (see also Fig. 2).

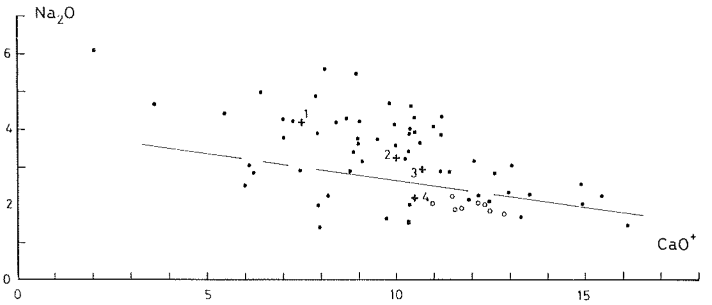

Fig. 5. Plot of Na_2O verses CaO in the Hokkaido basic rocks (excluding keratophyres).
* CaO adjusted for the amount of $CaCO_3$ present.
Open circles represent unaltered basalts and dolerites.
1. Av. of 225 'spilites' (Vallance, 1969).
2. Av. of 84 alkaline dolerites (Manson, 1967).
3. Av. of 24 Weathered oceanic ridge basalts (Hart, 1970).
4. Av. of 282 oceanic tholeiites (Manson, 1967).

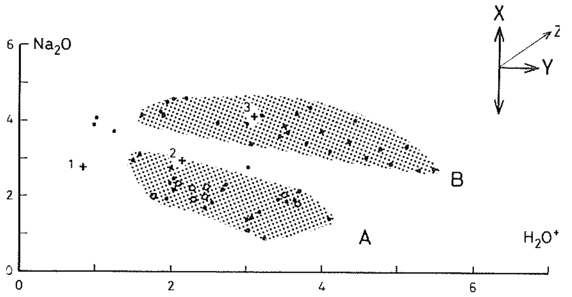

Fig. 6. Plot of Na_2O verses H_2O^+ in the Hokkaido basic rocks (excluding keratophyres).
Field A = < 20% sodic plagioclase or unalbitized plagioclase.
Field B = > 20% sodic plagioclase.
Open circles represent unaltered basalts and dolerites.
1. Av. of 45 fresh ocean ridge basalts (Hart, 1970).
2. Av. of 24 weathered ocean ridge basalts (Hart, 1970).
3. Av. of 225 'spilites' (Vallance, 1969).
X – Direction of model plagioclase variation and amount of albitization.
Y – Direction of alteration in terms of increasing H_2O content and therefore, inversely (see Fig.7), the relative amount of Ca that is combined with CO_2 and is removed from the system.
Z – The overal trend of basic rock alteration.

On the basis of Na and H_2O variation a chemical distinction between 'normal' basalt (affected by initial sea water – basalt chemical exchange) and 'spilitized' basalt* can be made although the division is somewhat arbitrary as demonstrated in Fig. 5 where Na is plotted against Ca in the rocks. The effect of modal plagioclase and the degree of alteration and albitization on the

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* Plagioclase is albite.

amount of soda enrichment is perhaps best illustrated in a plot of Na verses H_2O (Fig. 6). From Figs. 4, 5 and 6 rocks with high Na, H_2O and Fe^{3+}/Fe^{2+} ratio characterize a spilite association.

It should be pointed out here that in attempting to characterize the chemical nature of altered basaltic rocks an initial sampling problem exists (e.g. Hughes, 1972). Released Ca (from the plagioclase reaction) is able to combine with CO_2 to form calcite which may be removed from the system. Calcite is common as pods, veins and networks in the Hokkaido basic rocks. Consequently, rocks selected for analysis, often because of the paucity of calcite, would naturally lead to a biased representation of the basalt chemistry. Considering the mobility of a $CaCO_3$-rich fluid the choice of a representative sample becomes very difficult. Vallance (1965) has drawn attention to the selection of 'fresh' material in pillow lava cores with respect to the chorite (Na-poor) rims leading to a bias towards sodic enrichment. Further, the formation of areas rich in epidote with an increase in grade has the same effect as demonstrated by Smith (1968), Reed and Morgan (1971) and Graham (1975).

In the Hokkaido basic rocks there is an inverse relation between CO_2 and H_2O (Fig. 7) which largely reflects the modal relation between secondary hydrous minerals, e.g. chlorite – white mica and calcite. This feature indicates the dominant control of the relative mobilities and distribution of CO_2 (which

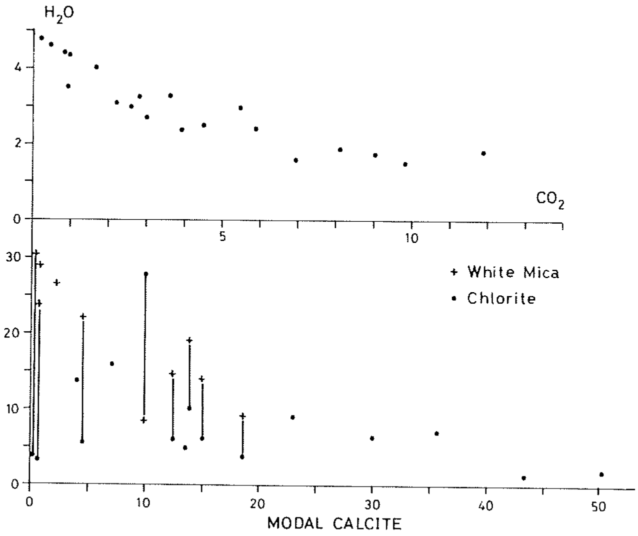

Fig. 7. Plots of H_2O verses CO_2 and modal white mica + chlorite verses modal calcite in the Hokkaido basic rocks. Modal chlorite includes small amounts of prehnite and/or pumpellyite.

combines with free Ca) and H_2O on spilite genesis. Fig. 8 shows major element variation with respect to that of Ca (less Ca combined with CO_2 to form calcite) which has a large variation from 0.7 – 15 wt%. Although a considerable scatter of points is to be expected from modal differences and the degree and duration of alteration of the rocks (e.g. Fig. 6) there is an increase in Si, Al, Fe^{3+}, Ti, K, P and H_2O (+ Na, as illustrated in Fig. 5), and a decrease in Mg and Fe^{2+} with decreasing Ca, i.e. increasing alteration. These trends

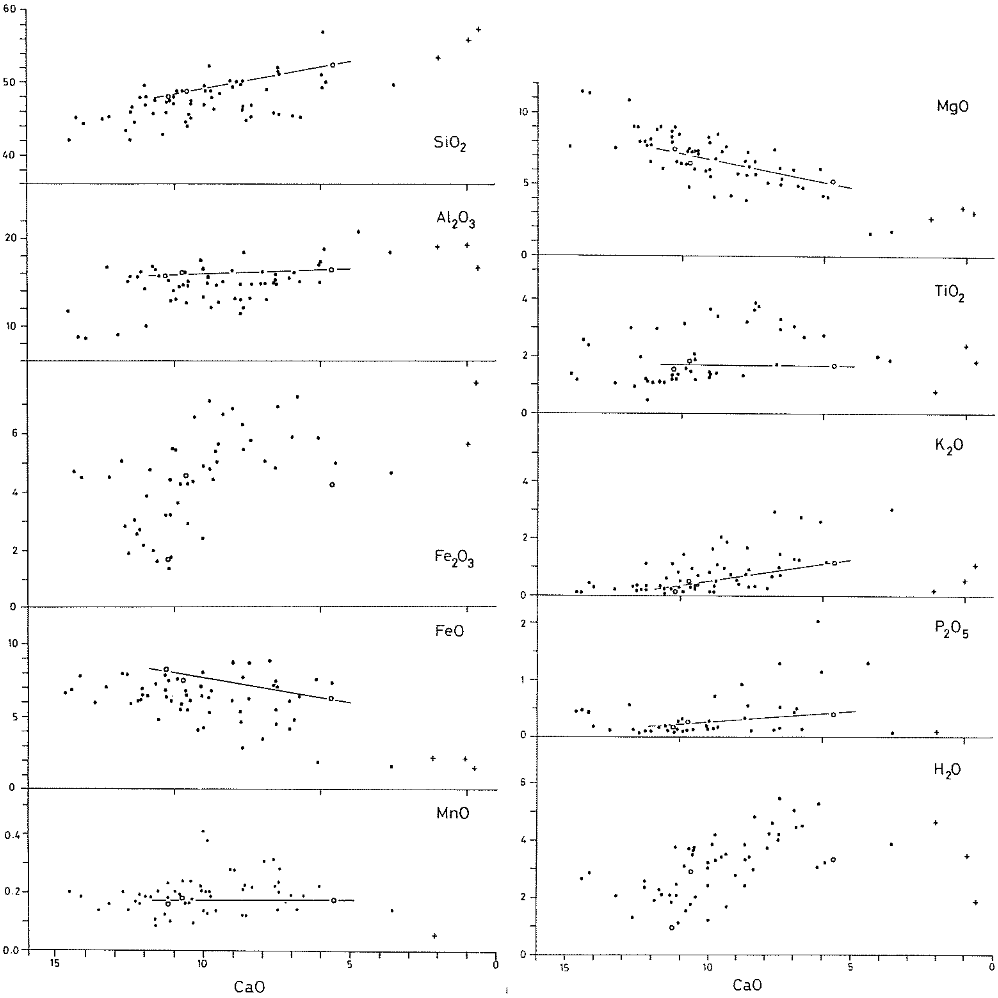

Fig. 8. Plots of major elements on CaO variation diagrams for the Hokkaido basic rocks. The low temperature alteration trend is given by the straight line as defined by average analyses of fresh ocean basalt, weathered oceanic ridge basalt and spilite in order of decreasing CaO content, respectively, as given in previous diagrams. Crosses represent keratophyric rocks.

indicate that the 'spilitization' process is merely an extension of the initial sea water – basalt reaction which is intensified during burial when calcic plagioclase becomes unstable and inverts to albite.

Metamorphic Grade

The presence of chlorite, calcite and albite, together with prehnite and/or pumpellyite in the metabasic rocks is typical of spilite associations (e.g. Vallance, 1969; Smith, 1968; Winkler, 1974) and indicates that burial conditions reached that of very low grade prehnite-pumpellyite metamorphism, i.e. conditions of 'normal' burial with a geothermal gradient of around 30°C/km (e.g. Winkler, 1974).

Consideration of the $CaO - Al_2O_3 - SiO_2 - H_2O - CO_2$ system (Ivanov and Gurevich, 1975) gives a lower limit of prehnite formation at about 260°C at $P_{total} = 1Kb$ with P_{CO_2} of around 23 bars. In the same system at a higher P_{total} of 2.5 Kb with $P_{CO_2} \sim 50-60$ bars the laumontite field is practically non-existent and the lower temperature stability of prehnite is near 285°C. These limits correspond to intermediate and unbuffered X_{CO_2} assemblages containing calcite as the main Ca-bearing phase with X_{CO_2} varying from 0.2 to 0.65 (Harte and Graham, 1975). As a result of the high CO_2 activity in the Hokkaido basic rocks during alteration Ca-zeolites, e.g. laumontite and heulandite, are essentially absent.

Primary Magma Type and Tectonic Environment

On the basis of $Na_2O + K_2O/SiO_2$ and AFM plots, Sawada and Kanmera (1973) and Bamba (1974) suggested that the primary magma of the Hokkaido basic rocks changed from tholeiitic to alkaline with time. However, no supporting mineralogical evidence, e.g. pyroxene compositions, is yet available to verify this trend. In view of the well known fact that low grade alteration of basalts leads to alkali enrichment, as discussed above, often to the extent that nepheline appears in the norm, such a conclusion should be treated with caution. The *least altered* basic rocks are all olivine – hypersthene or quartz normative tholeiites.

Successful attempts have been made in the application of the minor elements Ti, K, and P as a means of discriminating between oceanic and non-oceanic basalts and between ocean ridge tholeiites and alkaline basalts (e.g. Rhodes, 1973; Pearce et al. 1975). Bearing in mind the mobility of K (and P as demonstrated above) a $TiO_2 - P_2O_5 - K_2O$ plot given in Fig. 8 shows that unaltered as well as many of the more altered Hokkaido basic flows and diabases plot within the oceanic field, those rocks enriched in K relative to Ti and P reflecting more intensive burial metamorphism and alteration. Likewise a binary plot of Ti verses P indicates that the least altered rocks fall in the oceanic ridge basalt field. More altered basaltic rocks with higher Ti and, to a greater extent, P range into the alkaline basalt field and indicate that the use of these "alteration resistant discriminants" for characterizing petrogenetic environments cannot be applied to older basaltic rocks.

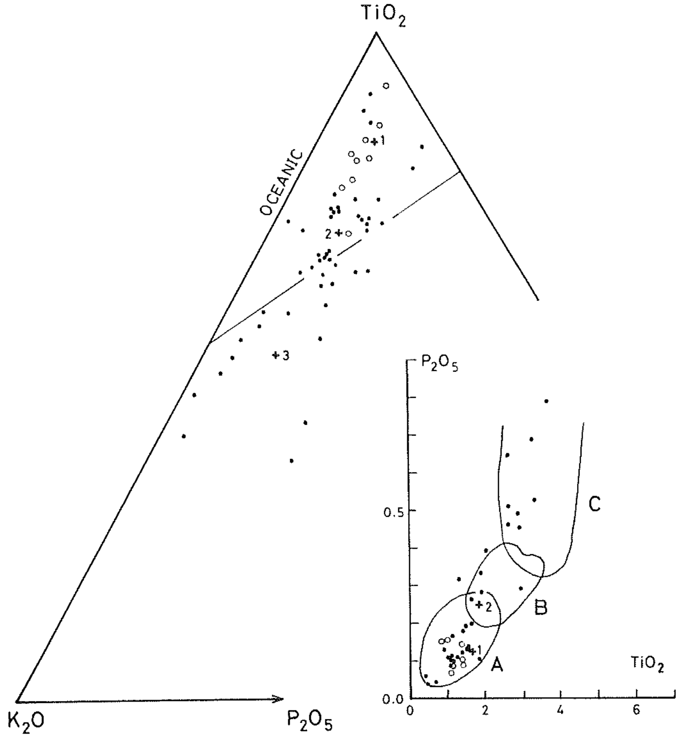

Fig. 9. Plots of Hokkaido basic rocks on $TiO_2 - K_2O - P_2O_5$ diagram (after Pearce et al. 1975) and a TiO_2 verses P_2O_5 diagram (after Rhodes, 1973).
Open circles represent unaltered basalts and dolerites.
1. Av. of 45 fresh ocean ridge basalts (Hart, 1970).
2. Av. of 24 weathered ocean ridge basalts (Hart, 1970).
3. Av. of 225 'spilites' (Vallance, 1969).
Field A – Oceanic tholeiites.
Field B – Oceanic island tholeiites.
Field C – Alkaline basalts.

The oceanic character of the basic rocks is supported by their field relations and tectonic environment. The basic rocks are associated with geosynclinal sediments deposited in a trough on the oceanic side of the contemporaneous

Sambosan Trough of Late Permian – Early Triassic age (Kimura, 1975). The association of some of the basic rocks with serpentinite and metagabbro is suggestive of an ophiolite-type suite which is now represented as obducted and disrupted oceanic crustal material (Grapes, in preparation).

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