GEOCHEMISTRY OF THE MIOCENE SANDSTONES OF BHUBAN FORMATION OF THE SURMA GROUP OCCURRING IN AND AROUND KATIGORAH, CACHAR DISTRICT AND ADJOINING AREAS OF BADARPUR, KARIMGANJ DISTRICT OF ASSAM, INDIA

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Abstract:
Geochemical composition (major oxides) of the Miocene Sandstones of Bhuban Formation of the Surma Group exposed in and around Katigorah, Cachar District and adjoining areas of Badarpur, Karimganj district of Assam, India was determined to reveal their palaeoweathering, chemical maturity, palaeoclimate, provenance and tectonic setting. Geochemical classifications show that the sandstones are sublithic to greywacke in composition. The positive correlation between the major elements (oxides) is observed and it suggests the influence of source rock lithology. From the study of palaeoweathering, it is seen that K-feldspar is abit more in Bhuban sandstones indicating moderate intensity of chemical weathering in the source area. Regarding chemical maturity, sandstones are slightly matured. The geochemical studies reveal that the climatic condition was arid during that time. It is also seen that the sandstones are derived mostly from metamorphic and igneous (mostly granite) sources which are present in the adjoining areas. Under tectonic setting, sandstones are of Active Margin type to Passive margin type and the sediments were deposited in Miocene foreland basin formed due to collision orogen.

Keywords: **Geochemistry, Miocene Bhuban Formation, Katigorah, Assam**

Introduction:

Fine grained sandstone samples were collected for chemical analysis. These samples were collected from Bhuban Sandstone Formation occurring in and around Katigorah, Cachar district and adjoining areas of Badarpur, Karimganj district of Assam, India.

The geochemical studies are mainly concerned with variations in major and trace elements in sedimentary rocks which reveal source and environment of deposition sediments because the distribution of the elements in sediments are controlled by various physical, chemical and biological factors. Besides these, geochemical studies help to understand the nature of redox changes in the course of the history of the earth.

The geochemistry of any particular clastic sediment is the cumulative result for four principal factors (Sawyer, 1986) like source rock composition in the provenance area, degree of chemical weathering experienced by the source rocks in the provenance area and during subsequent transportation, processes occurring during sediment transport and deposition, such as sorting and maturity and elements mobility during diagenesis. Factors controlling the geochemistry of a clastic sedimentary rock include: i. Composition of source terrain, ii. Chemical weathering, iii. Hydraulic sorting, iv. Diagenesis, v. Metamorphism and vi. Hydrothermal alteration (Fralick and Kronberg, 1997).

Thus the chemical composition of terrigenous sedimentary rocks is a function of various variables. It is controlled by composition of their source rocks, environmental parameters influencing the weathering of the source rocks, duration of weathering, transportation mechanisms of clastic materials from the source areas to the sites of deposition, depositional environments, post depositional processes and climatic variability (Nesbitt and Young, 1982: Cox *et. al*, 1995; Dey *et. al*, 2009; Ranjan and Banerjee, 2009). The addition or loss of elements due to relations with hydrothermal solutions or with organic rich solutions may also be recorded in sedimentary rock chemistry (Hayashi *et. al*, 1997). Tectonism has been advocated to have a primary control on sedimentary composition (Blatt *et. al*., 1980). The geochemical indices most frequently used in the interpretation of ancient depositional environment and the parameters that represent more reliable geochemical palaeo – oxygenation indices have been discussed by Jones and Manning (1994).

Many recent studies have shown a close relationship between geochemical composition of sedimentary rocks and their provenance characteristics. Tectonic environment and palaeoweathering conditions have been observed by many workers (Naqvi *et. al*., 1988; Condie, 1993; Sreenivas and Srinivasan, 1994; Raza *et. al*., 2002). Hence Geochemical data have been widely used as an indication of provenance of sedimentary formations and tectonic and climatic conditions in which they were deposited. Moreover, geochemical composition is a guide to the source rocks in the provenance and the tectonic setting of provenance and the depositional basin (Blatt *et. al*, 1980; Bhatia, 1983; Bhatia and Crook, 1986; Roser and Korsch, 1986 and Suttner and Dutta, 1986, Armstrong altrin *et. al*, 2004; Armstrong-Altrin and Verma, 2005; Sinha *et. al*, 2007; Das *et. al*, 2006; Nagaranjan *et. al*, 2007).

Geochemical studies are used to discriminate sediment composition into broadly defined tectonic or sedimentary environment (Argast and Donnelly, 1987). The major oxides variations and trace elements concentrations of the sandstones are utilized to reconstruct tectonic setting of depositional basin, provenance and reveal palaeo climatic conditions and palaeoweathering.

Geology of the area

The outcrops of Tertiary rocks are partially exposed in and around of Katigorah of Cachar district and adjoining areas of Badarpur, Karimganj district of Assam, India. The location map of the study area is shown in Fig. 1. Lithological Characters show that these are the Sandstones of Bhuban formation belonging to Surma Group. Lithologically, Sandstones are medium to fine grained, bluish grey to greenish and yellowish grey in colour. Moreover, sandstones are loose and hard, massive to bedded in nature. Intercalated shales are common mostly within the fine grained sandstones. Based on litholgical characters, Bhuban Formation may be divided into three divisions which are shown in the following stratigraphic succession (Table- 1) based on field observations.

Analytical Techniques

Some representative sandstone samples were analyzed for major and minor elements. The samples were analyzed in the Department of USIC, Gauhati University by WDXRES method. The instrument used in these analyses is PAN analytical AXIOS XRF spectrometer. The accuracy and precision of the instrument are within 1.5 percent of major.

Geochemical Analysis

The geochemical analysis of sandstones mainly includes major (oxides) elements studies. Ten representative sandstone samples of Bhuban Formation have been analyzed for major oxides.

Major Oxides:

Major oxide studies of sandstones are essential in order to identify the source rocks and tectonic settings in their composition. Major elements undergo some changes during sedimentary processes eg. in most basins SiO_2 is enriched and Na_2O and CaO are depleted in sandstones compared with the source rock composition. Thus the major element chemistry gives clues as to the provenance type as well as weathering conditions, both of which are controlled by the tectonic setting of the basin (Bhatia, 1983). Bhatia (1983) established a tectonic classification based on SiO_2 content and K_2O/Na_2O. Palaeoclimate can be done with the help of bulk composition (Suttner and Dutta, 1986). Chemical maturity and palaeoweathering etc. can also be studied with the help of major oxides.

The percentage of major oxides variation of Bhuban sandstones of Katigorah and adjoining areas of Badarpur are given in Table 2. These include SiO_2, Al_2O_2, Fe_2O_3, Na_2O, K_2O, CaO, MgO, TiO_2, MnO and P_2O_5.

In terms of mean element abundance the values reflect the mineralogical character of the sandstones. The variations in major element geochemistry are shown vide Harkar variation diagrams (Figs. 2-7). The variations are analyzed in relation to SiO_2 and variations between other radicals. The Bhuban sandstones have high SiO_2 concentration of 60.52 to 76.52 wt.%. Considering SiO_2 as a common factor, it is seen that Al_2O_3, Fe_2O_3, K_2O, Na_2O, MnO and MgO show positive correlation with it. It indicates dominance of free silica in the form of quartz. The correlation between alumina with oxides like K_2O and Fe_2O_3 shows negative nature. The Al_2O_3 content is high (Av. 11.50 wt. %) and it might be due to alteration of K-feldspar. It indicates an aluminous source. The correlation between Fe_2O_3 +MgO and Al_2O_3 implies that both Fe and Mg are controlled by clay minerals and /or mica. The TiO_2 concentration is low (0.29 to 0.91 wt. %) and might have come from metamorphosed argillaceous rocks or acid igneous rocks.

Palaeoweathering

Alteration of minerals due to chemical weathering mainly depends on the intensity and the duration of weathering. The dominant process during weathering of the upper crust is the degradation of feldspars and formation of clay minerals. The degradation of feldspar, which is very sensitive to chemical weathering, increases the mobility of many elements through clays (Taylor and McLennan, 1985). During weathering, calcium, sodium and potassium are removed from feldspar (Nesbitt *et. al*, 1980). The amount of these chemical elements surviving in the soil profiles and in the sediments derived from them is a sensitive index of the intensity of weathering (Nesbitt *et. al*, 1997). Thus, the abundance of easily removable elements serves as an index of chemical weathering. Elements such as calcium, sodium and potassium can be removed by solution during weathering. The influence of weathering on sedimentary rocks can be calculated by using Chemical Index of Alteration (CIA) (Nesbitt and Young, 1982). The CIA value indicates the degree of weathering of source rocks and is determined by the equation – CIA = [Al_2O_3 / (Al_2O_3 + CaO sil. + Na_2O + K_2O)] x 100 where CaO sil. represents CaO associated with silicate phases. The CIA gives a measure of the ratio of original/primary minerals and secondary products such as clay minerals. CIA values are nearly 50 to 100 for completely weathered rocks, consisting entirely of secondary minerals. Whereas low values (i.e. 50 or less) indicate unweathered source areas. The CIA is a good measure of palaeoweathering conditions and it essentially monitors the progressive weathering of feldspars to clay minerals (Fedo *et. al.*, 1995; Armstrong - Altrin *et. al.*, 2004). High CIA values indicate intensive chemical weathering in the source area.

K-metasomatic enrichment of potassium to sediments and sedimentary rocks produces mineralogical changes which alter the earlier composition (Nesbitt and Young, 1980; Condie, 1993). Potassium metasomatism is particularly common which involves conversion of kaolin to illite by reaction with potassium bearing pore-waters (Fedo *et. al*, 1995). In sandstones, K-metasomatism can take place in two different ways – i. Aluminous clay minerals (kaolinite) are converted to illite and / or ii. Conversion of plagioclase to K-feldspar (Fedo *et. al*, 1995). These processes produce K_2O enrichment in the sedimentary rocks and it may vary from the weathering trend. Both these processes may affect the composition of sedimentary rocks.

The calculated CIA values for the Bhuban sandstones vary from nearly 65.50 to 70% with an average of 68.50 indicating the sandstones are derived from the least to moderate weathered zones.

Chemical Maturity

Chemical Maturity Index (CMI) of clastic sediments can be expressed as SiO_2 percent and SiO_2 / Al_2O_3 ratio defining chemical maturity (Pettijohn *et. al*, 1972).

The sediments of Bhuban sandstones show comparatively higher SiO_2 / Al_2O_3 ratio. The plots of Al_2O_3 + K_2O + Na_2O against SiO_2 in the Suttner and Dutta (1986) binary diagram (Fig. 8) suggest slightly increasing chemical maturity.

Palaeoclimate

Major elemental data provides useful information regarding the climatic conditions which prevailed during the deposition of sedimentary rocks (Suttner and Dutta, 1986). The bivariate plots of Al_2O_3 + K_2O + Na_2O against SiO_2 differentiates the climatic conditions under which the sediments were deposited. The chemically immature sandstones with low silica content indicate a less intense weathering with arid or cold climate, whereas sub-mature and mature sandstones with high silica content indicates warm-humid climate (Suttner and Dutta, 1986).

The plots of Al_2O_3 + K_2O + Na_2O against SiO_2 in the Suttner and Dutta (1986) binary diagram (Fig. 7) suggest arid climatic condition for Bhuban sandstones.

Provenance

Source rock composition is commonly thought to be the dominant factor. However, secondary processes like weathering, diagenesis can have an effect on chemical composition (Cullers *et. al.*, 1988) and therefore, one best relies on elements that show little mobility under the geologic conditions.

Composition of clastic rocks like sandstone and shale is mainly influenced by the source rock characteristics, the nature of sedimentary processes within the depositional basin and the kind of dispersal paths that link provenance to the basin (Dickinson and Suczek, 1979). Nesbitt *et. al*, (1996) interpreted that the nature of a provenance area can be inferred from the study of the sedimentary rocks. According to them, the key relations between provenance and basin are governed by plate tectonics.

Geochemical characters of clastic rocks have been used to decipher the provenance of the source region. The geochemical composition of terrigenous sand is a function of complex interplay of provenance, weathering, transportation and diagenesis. Major element geochemistry gives clues to the provenance (Condie, 1993).

Condie (1967) has proposed a ternary diagram of Fe_2O_3 - MgO - TiO_2 for the determination of the source rocks of the sediments. The plots of Fe_2O_3, MgO and TiO_2 wt% of the Bhuban sandstone samples fall in the field 1 (Fig. 9) which indicates that the source rock of sandstone was probably granite and quartz monzonite.

The plots following Crook (1974) have suggested the sediments of the sandstones were derived from quartz rich provenance (Fig. 10).

The plots in the CaO-Na_2O-K_2O ternary diagram after Le Maitre (1976) show that the provenance is granitic (Fig. 11) for Bhuban sandstones.

Tectonic Setting

The tectonic setting of sedimentary rocks can be inferred from major elemental data (Crook, 1974). It predominantly exhibits the nature and proportion of their detrital components and the bulk composition reflects the tectonic setting of the basin (Siever, 1979; Roser and Korsch, 1986). Bhatia (1983) used the composition and the tectonic setting in which sediments were formed. Winchester and Max (1989) have successfully used elements as geochemical tectonic indicators in immature sediments.

For tectonic setting discrimination, plots suggested by Bhatia (1983) and Roser and Korsch (1986) were used. Moreover plots of Blatt *et al*, (1980) were also used for tectonic setting.

In a simplified plate tectonic classification of continental margins and oceanic basin based on the nature of the crust, Bhatia *et. al.* (1980) has recognized four types of settings namely Oceanic Island Arc (OIA), Continental Island Arc (CIA), Active Continental Margin (ACM) and Passive Margin (PM).

For sandstones of the area the plots in the Roser and Korsch (1986) binary diagram are falling mostly in the Active Continental to Passive Margin (Fig. 12).

The ratio of Al_2O_3 / SiO_2 gives an indication of the quartz enrichment in sandstones. The ratio of Al_2O_3 / (CaO + Na_2O) is a comparative measure of mobile to the immobile oxides. The plots of K_2O / for Bhuban sandstones are close to Active Continental Margin (ACM) to Passive Margin (PM) (Figs. 13 & 14). Similar views have also expressed by Rahman *et. al.* (2014) and Roy *et. al.* (2010).

The plot of Blatt *et. al*, (1980) for Bhuban sandstones fall mostly in Euogeosyncline of ferromagnesium sodium sandstone (Fig. 15) Blatt *et. al*, (1980) states that Euogeosynclinal sediments are deposited in near continental to Passive margin environments.

Sandstones of ACM types were derived mostly from uplifted areas (Roser and Korsch, 1986). Geochemical characters positively support its derivation from intracratonic highs of uplifted basement and deposited in a basin along a tectonic setting and sediments were derived from metamorphic and recycled older sediments.

Low Al_2O_3 / SiO_2 ratio of sandstones is further characteristic of quartz rich source deposited in a Continental Margin Setting (Phillips, 1991) and K_2O / Na_2O ratio and resultant overlap between passive and active continental margin settings reflect a complex provenance and history of recycling of these sediments (Bhatia, 1983).

Geochemical Classification of the Sandstones

Geochemical classification of terrigenous sedimentary rocks has been proposed by many authors based on major element composition (Pettijohn *et al*, 1972; Crook, 1974; Blatt *et al*, 1980; Herron, 1988).

Using the indices of SiO_2 / Al_2O_3 and Na_2O / K_2O ratios, Pettijohn *et al.*, (1972) proposed a classification for terrigenous sands based on a plot of log(Na_2O / K_2O) versus log(SiO_2/Al_2O_2).

Herron (1988) modified the diagram of Pettijohn *et. al*, (1972) using log (Fe_2O_2/K_2O) instead of (Na_2O / K_2O).

The plots after Pettijohn *et. al*, (1972) for sandstones fall mostly in the litharenite field to greywacke field (Fig. 16). The plots after Herron (1988) fall mostly in wacke field (Fig. 17).

Conclusion

From the above geochemical studies of major and minor elements of Bhuban sandstones of Surma Group of the study area the following conclusions can be made:

I. The positive correlation between the major elements (oxides) is observed and it suggests the influence of source rock lithology particularly granitoid and metamorphosed argillaceous source rocks.

II. From the study of palaeoweathering, it is seen that K-feldspar is more in Bhuban sandstones. Hence CIA values of Bhuban sandstones are high indicating moderate intensity of chemical weathering in the source area.

III. Regarding chemical maturity, sandstones are slightly matured.

IV. The geochemical studies suggest arid climatic condition.

V. From the geochemical studies, it is seen that the sandstones were derived mostly from metamorphic and igneous (mostly granite) sources which are present in the adjoining areas.

VI. Under tectonic setting, sandstones are of Active Margin type to Passive Margin and were derived mostly from uplifted areas which were characteristics of igneous, metamorphic and recycled sediments deposited in Miocene foreland basin formed due to collision orogen.

VII. The sandstones are classified as litharenite to wacke from geochemical classification.

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Table 1: Stratigraphic succession of the study area

Age	Group	Formation	Lithology
Miocene	Surma	Bhuban	Upper: Medium grained, grey to greenish grey, hard, bedded Sandstones. Middle: Medium to fine groined, grey to yellowish colour massive to bedded with shale. Lower: Fine ground hard and loose, bluish to grayish, bedded sandstones with intercalated shale.
------------------------------Unconformity-----------------------------			-----------------------
Oligocene	Barail	Undifferentiated	Sandstones, siltstone and shale.

Table – 2: Major oxide variations (in Wt. %) of the Bhuban Sandstones of the study area

Sp. No.	Lab No.	SiO_2	Al_2O_3	Fe_2O_3 (T)	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	CIA
1	4084	70.29	12.30	2.68	0.013	1.01	0.51	2.87	1.60	0.29	0.03	68.25
2	4085	71.75	10.56	2.74	0.011	1.05	0.60	2.80	1.76	0.38	0.07	69.20
3	4086	76.52	11.13	3.01	0.110	1.07	0.60	2.81	1.95	0.56	0.09	72.60

4	4087	70.14	10.50	4.51	0.100	1.08	0.58	2.79	1.80	0.78	0.24	66.50
5	4088	74.92	11.00	2.99	0.090	1.08	0.61	2.70	1.68	0.91	0.06	65.30
6	4089	60.52	10.98	5.05	0.155	1.56	2.88	2.70	2.40	0.86	0.11	75.10
7	4090	63.19	11.50	5.61	0.170	1.66	2.51	3.10	2.10	0.71	0.10	74.65
8	4091	73.30	12.82	2.80	0.01	1.08	0.50	2.81	2.22	0.70	0.08	69.25
9	4092	73.78	10.51	2.81	0.011	1.04	0.50	2.78	1.70	0.33	0.03	68.10
10	4093	72.08	11.25	4.82	0.012	1.48	0.48	2.80	3.01	0.68	0.22	66.20

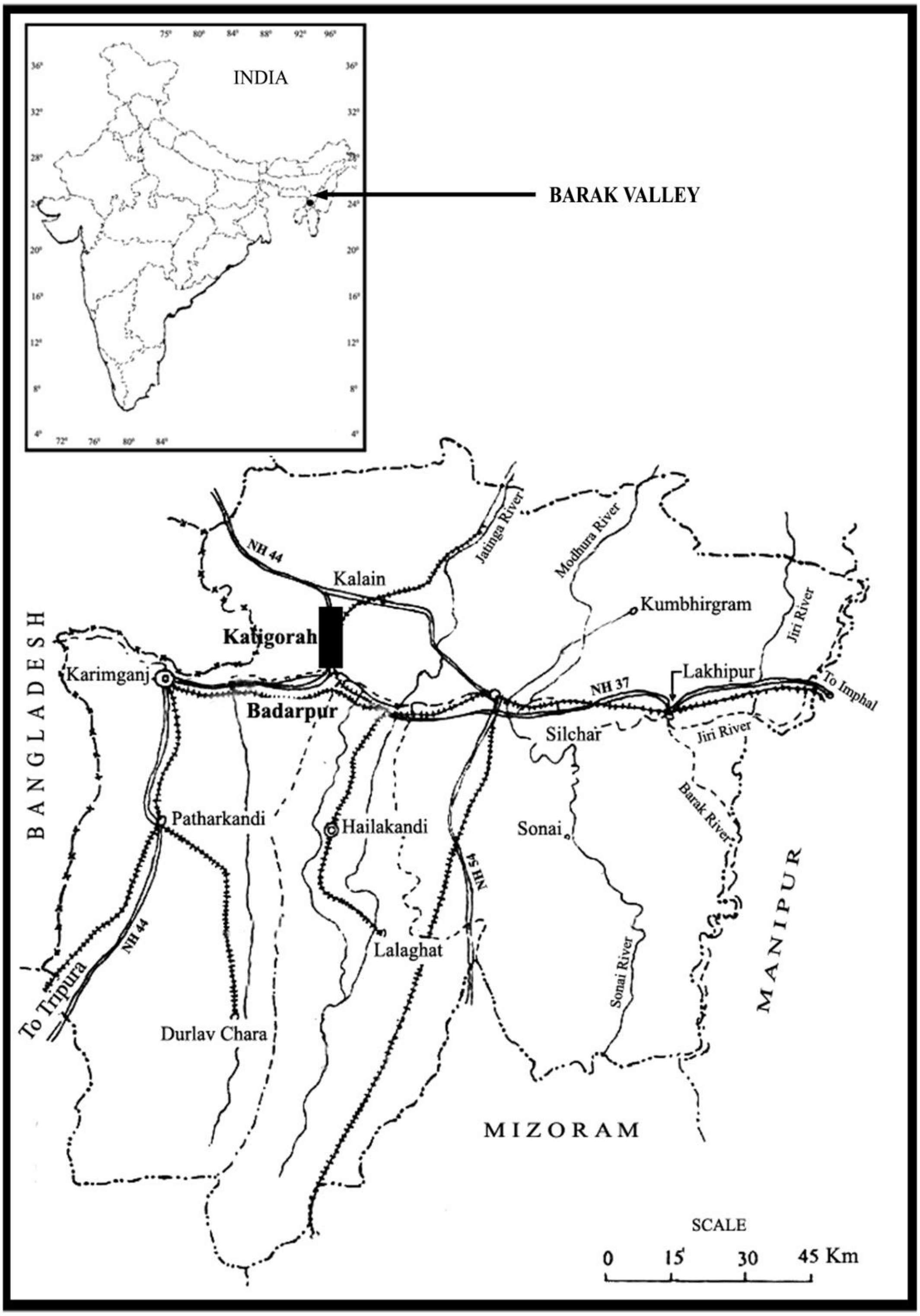

Fig. 1: Location Map of the Study Area.

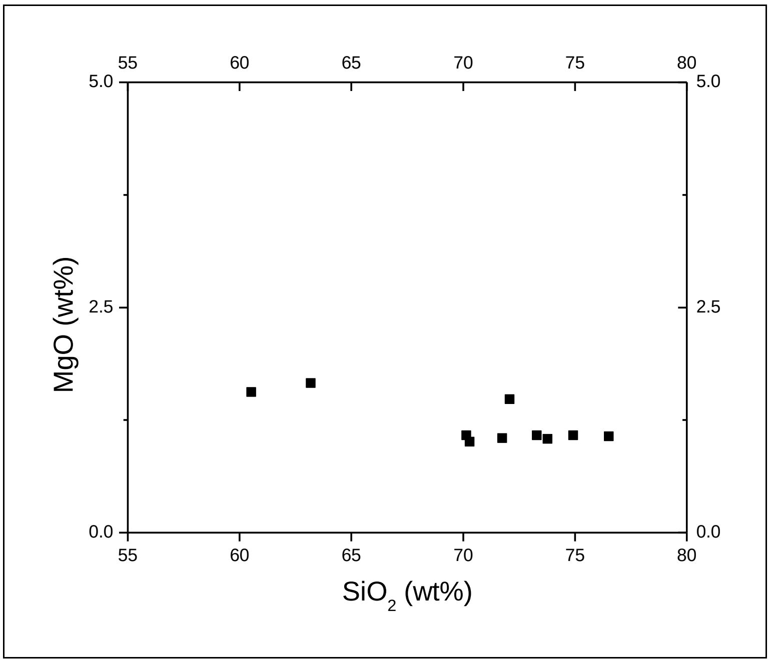

Fig. 2: Scatter plot of SiO_2 vs MgO of Bhuban Sandstones (Harkar Variation diagram).

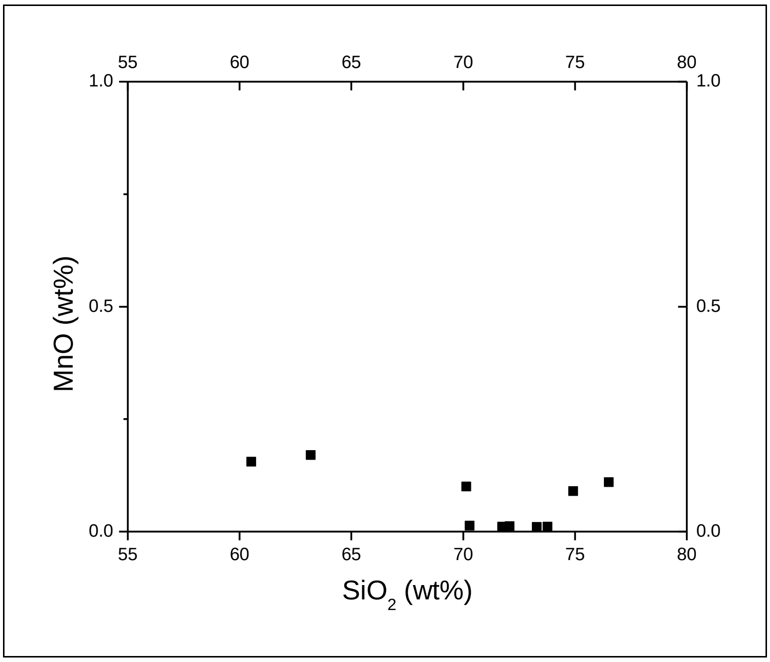

Fig. 3: Scatter plot of SiO_2 vs MnO of Bhuban Sandstones (Harkar Variation diagram).

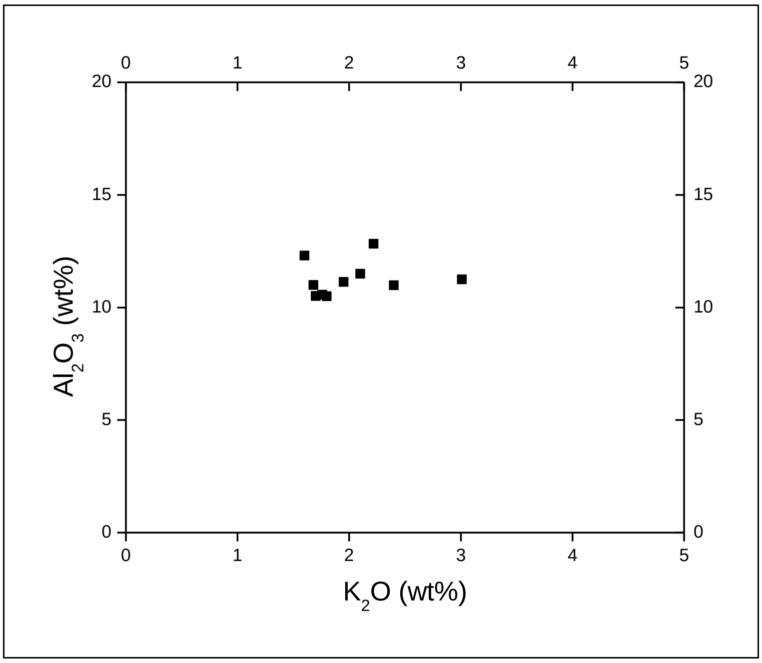

Fig. 4: Scatter plot of K_2O vs Al_2O_3 of Bhuban Sandstones (Harkar Variation diagram).

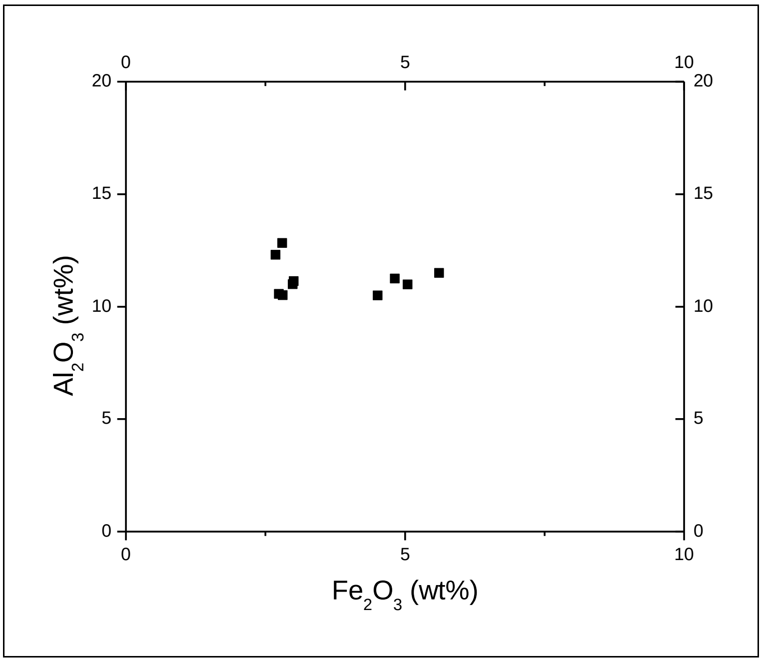

Fig. 5: Scatter plot of Fe_2O_3 vs Al_2O_3 of Bhuban Sandstones (Harkar Variation diagram).

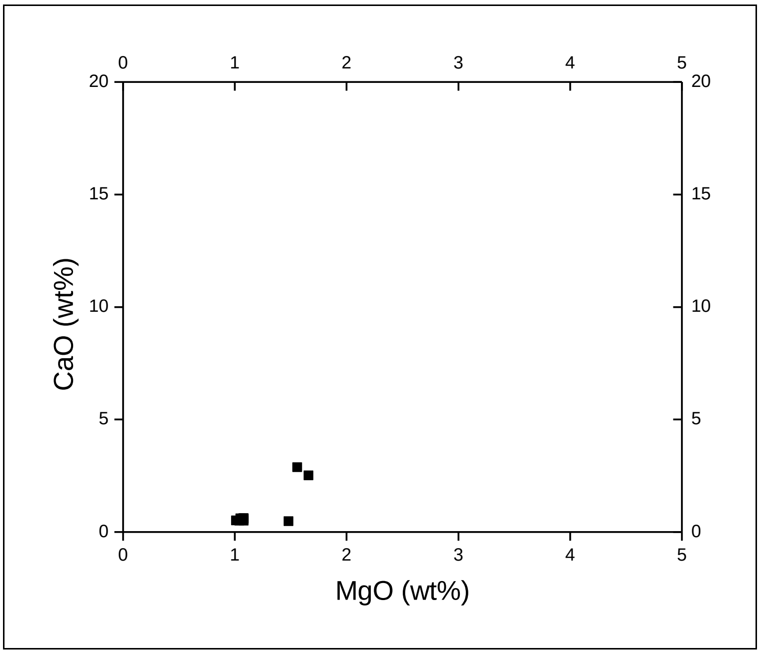

Fig. 6: Scatter plot of MgO vs CaO of Bhuban Sandstones (Harkar Variation diagram).

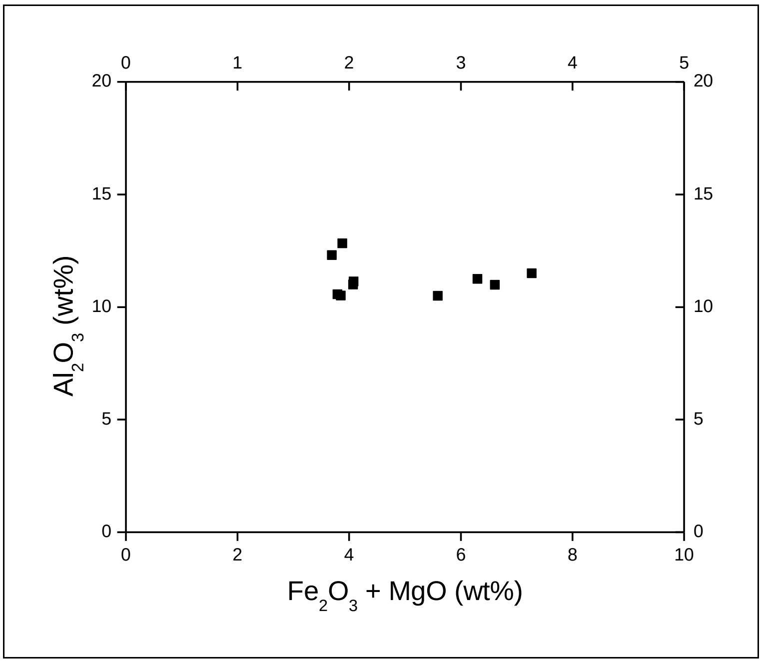

Fig. 7: Scatter plot of Fe_2O_3+MgO vs Al_2O_3 of Bhuban Sandstones (Harkar Variation diagram).

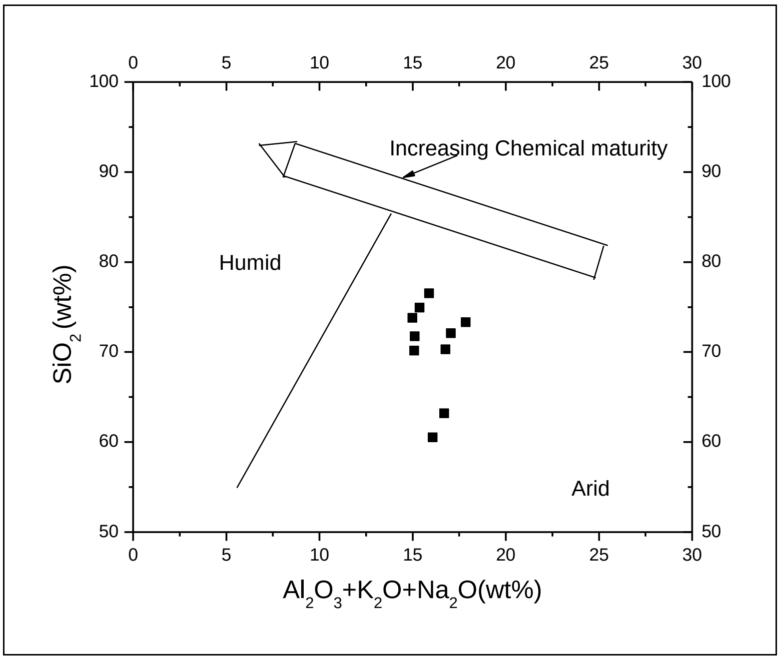

Fig. 8: Scatter plot of $Al_2O_3 + K_2O + Na_2O$ vs SiO_2 of sandstone (Suttner and Dutta 1986).

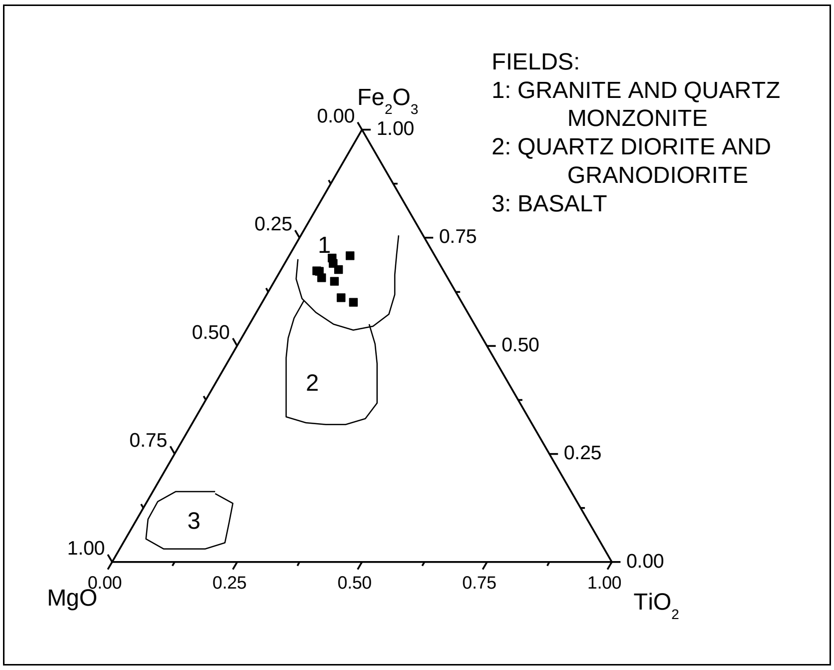

Fig. 9: Fe_2O_3 – TiO_2 – MgO compositional diagram (after Condie, 1967).

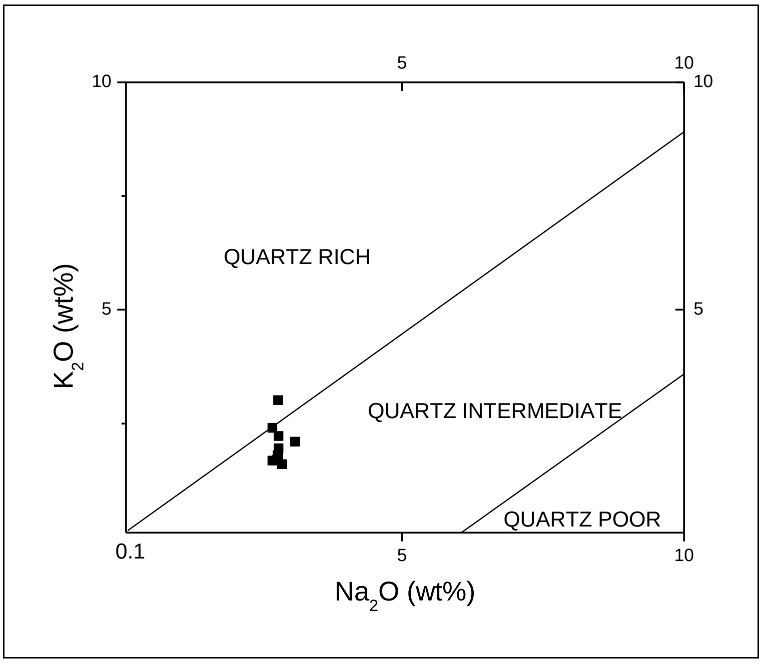

Fig. 10: K_2O vs Na_2O plot for analyses of sandstone (after Crook, 1974).

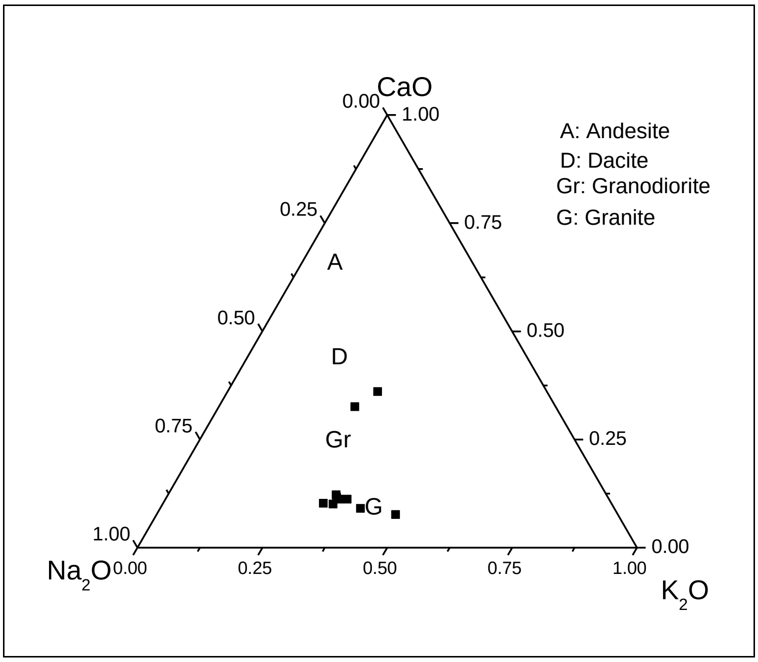

Fig. 11: CaO – Na_2O – K_2O plot (after LeMaitre, 1976).

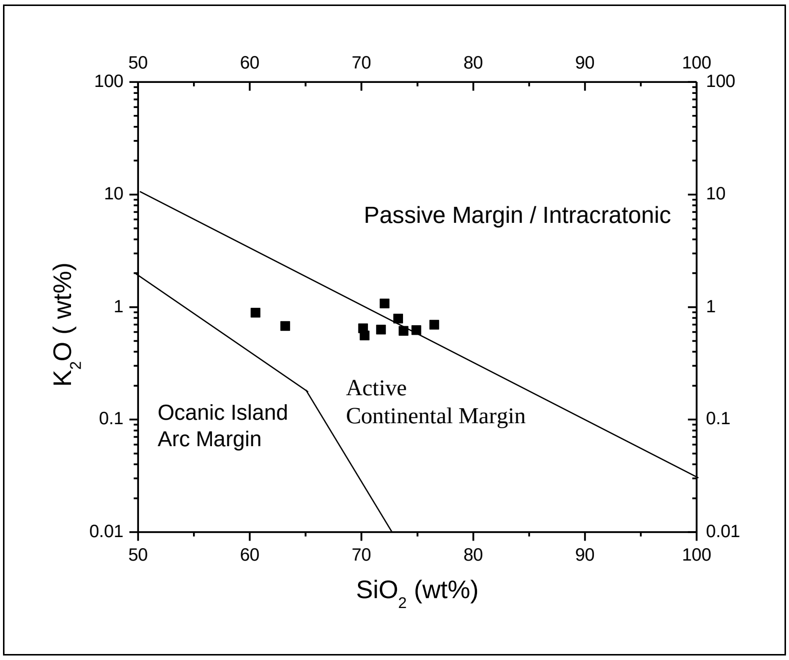

Fig. 12: Scatter plot of SiO_2 vs K_2O/Na_2O (after Roser and Korsch, 1986).

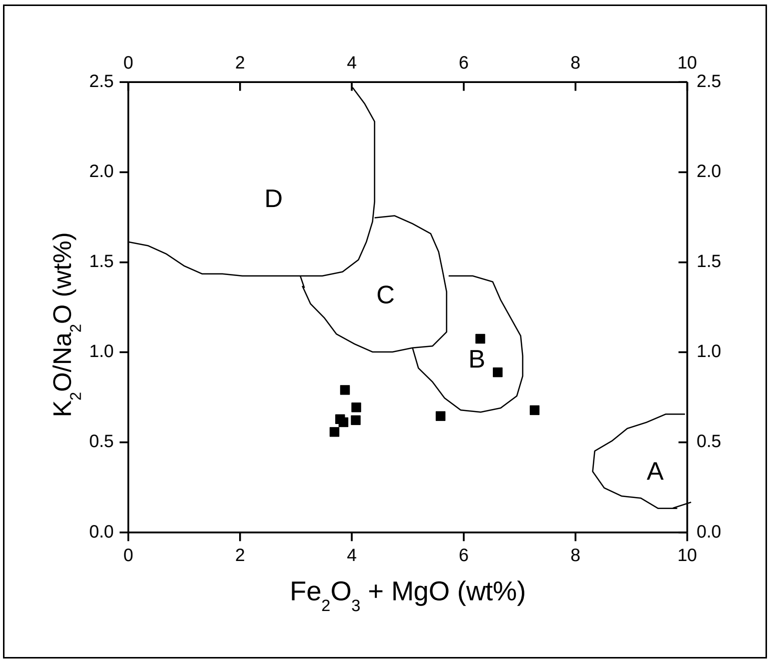

Fig. 13: Scatter plot of Fe_2O_3 + MgO vs K_2O/Na_2O (after Bhatia, 1983). [Tectonic Fields of Oceanic Island Arc (A), Continental Island Arc (B), Active Continental Margin (C) and Passive Margin (D)].

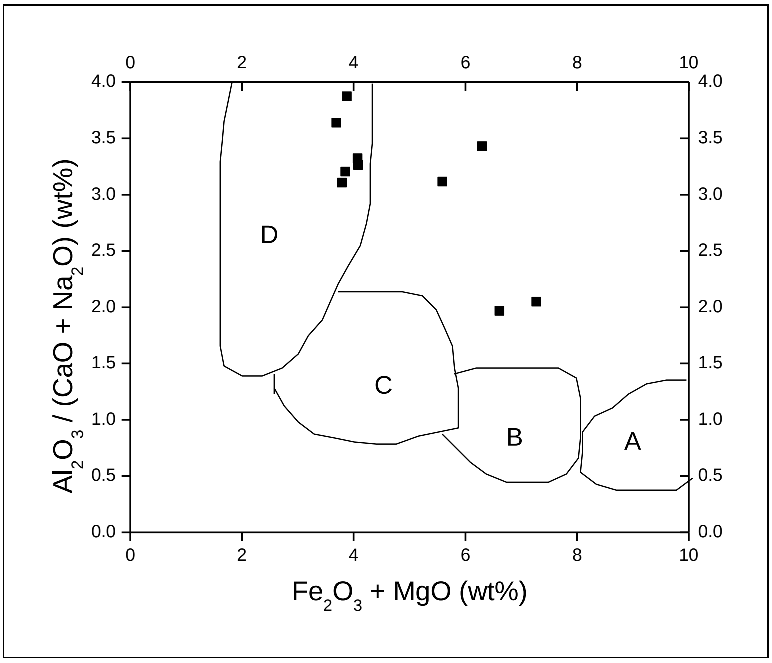

Fig. 14: Scatter plot of Fe_2O_3 + MgO vs Al_2O_3/ (CaO + Na_2O) (after Bhatia, 1983) [Tectonic Fields of Oceanic Island Arc (A), Continental Island Arc B), Active Continental Margin (C) and Passive Margin (D)].

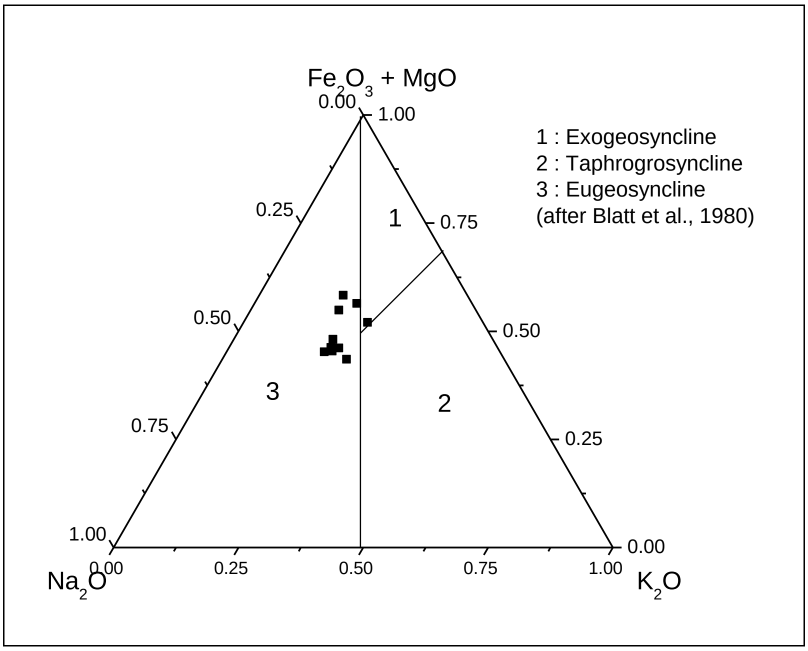

Fig. 15: (Fe_2O_3 + MgO) – K_2O – Na_2O plot (after Blatt et al., 1980).

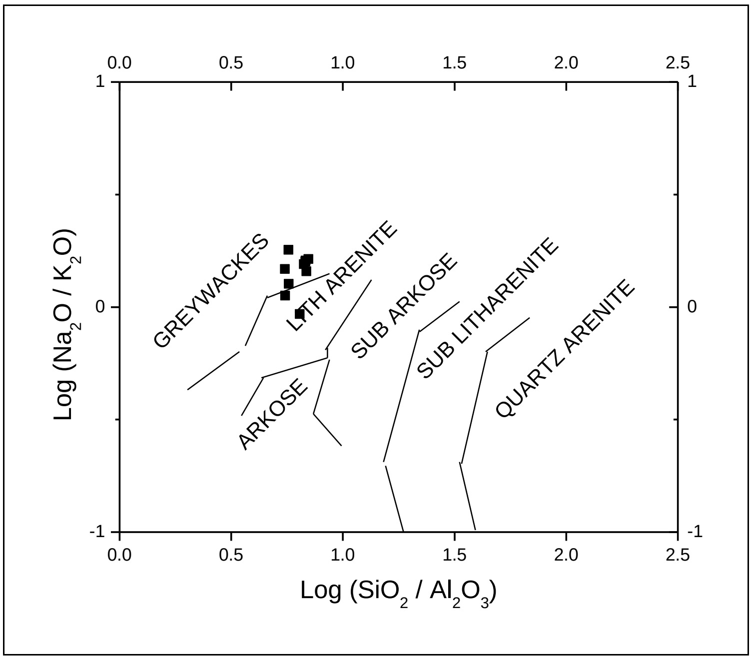

Fig. 16: Geochemical classification for Terrigenous sands (after Pettijhon et al., 1972).

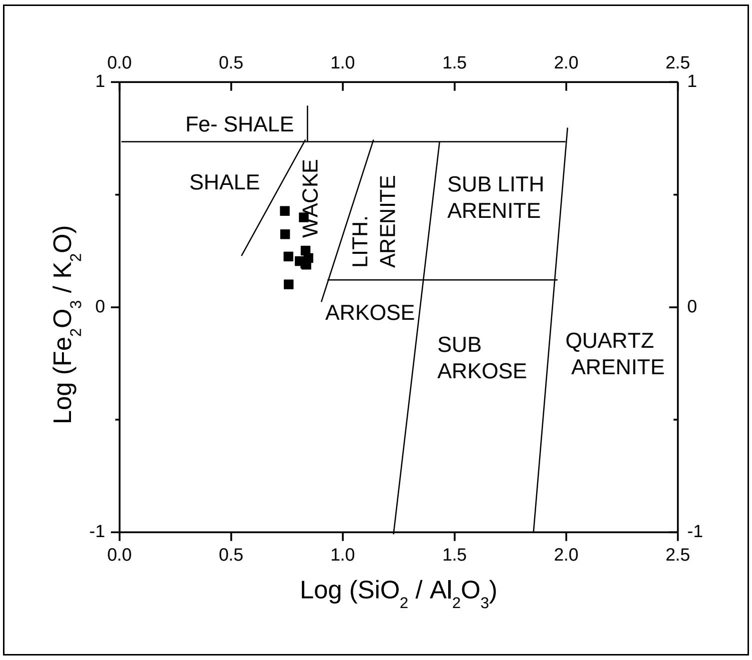

Fig. 17: Geochemical classification for Terrigenous sands (after Herron, 1988).