

Metallogeny of Tin: Magmatic Differentiation versus Geochemical Heritage

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Abstract

TiO₂-Sn and Rb/Sr-Sn variation diagrams for granitic fractionation series from various tin and non-tin provinces do not support the concept of geochemical heredity of tin, i.e., the assumption of a primary crustal Sn anomaly in tin provinces. The controlling factor for the generation of tin granites is a special bulk distribution coefficient of Sn, probably dependent on the oxygen fugacity of the melt.

Introduction

THE following basic observations on tin deposits must be taken into consideration for a discussion of the metallogeny of tin: (1) most primary tin deposits are closely related, in time and space, to granitic rocks; (2) these granitic rocks display a distinct geochemical specialization in Sn, as well as in other incompatible elements (e.g., F, Li, B, Cs, etc.); (3) on a worldwide scale, tin deposits are clustered in defined zones of restricted extent, the so-called tin provinces; (4) in the tin provinces itself, it is not unusual to find tin deposits of various ages close together or even superimposed. As the last observation may not be very widely known, examples of this "étagement temporel" (Routhier, 1967) are listed in Table 1.

There are essentially two basic theories by which the geochemical specialization of tin granites is explained: (1) enrichment in incompatible elements of an initially unspecialized melt during fractional crystallization (magmatic differentiation theory), and (2) the melt inherits its geochemical specialization during anatexis from source rocks, and magmatic evolution causes only an enhancement of the initial anomaly (geochemical heritage theory). The trace element pattern in igneous rock suites of tin provinces should allow one to decide which of these theories fits better the presently available data.

Variation Diagrams

A log-log plot of two trace elements *i* and *j* during Rayleigh fractionation (perfect fractional crystallization) will result in a straight line correlation with slope

$$m = \frac{\bar{D}_i^{x_i/liq} - 1}{\bar{D}_j^{x_j/liq} - 1} \quad (\text{Rayleigh, 1896}) \quad (1)$$

where $\bar{D}_{i,j}^{x_i/liq}$ = bulk crystal-liquid distribution coefficient for element *i* or *j*. This line will pass through the composition point of the initial, unfractionated melt x_i^0, x_j^0 . If we choose *i* to be a "neutral" indicator

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of magmatic fractionation (e.g., TiO₂, Rb/Sr) and *j* as the element tested for geochemical heritage (e.g., Sn), then the position of the correlation line, defined as

$$\log x_i = m \cdot \log x_j + b \quad (2)$$

should give information about the existence of a primary anomaly relative to other fractionation series, and the term *b* in (2) could be used as an indicator of the degree of geochemical heritage of element *j*.

The bulk distribution coefficient $\bar{D}_{i,j}^{x_i/liq}$ is treated here as a constant which of course is only a rough approximation. Nevertheless, this assumption seems acceptable because in the examples studied below the crystallizing phases and their proportions vary only in a very limited range within the granitic field (petrographic nomenclature according to Streckeisen, 1973).

Following this consideration, the two sets of variation diagrams in Figures 1 and 2 have been prepared showing the distribution of Sn in granitic whole-rock samples from various tin and non-tin provinces as a function of TiO₂ and Rb/Sr, respectively. TiO₂ and Rb/Sr have been chosen as two independent indicators of the degree of magmatic differentiation and are preferred to other possible fractionation indices because of the availability of these elements in many routine geochemical studies.

As far as petrographic-geochemical evidence goes, the granitic samples plotted in Figures 1 and 2 are essentially unaffected by alteration processes. Feldspars are fresh and unaltered; in some cases biotite may show the usual incipient chloritization. The trace element pattern of all sample suites is typical of magmatic differentiation series (log variation lines), clearly different from the scatter distribution and bimodal tin populations produced by hydrothermal alteration reactions (Groves and McCarthy, 1978; Hall, 1971; Lehmann, 1979, 1981).

The trace element variations of the examples studied do not fit the model of variable mixing of two end-member compositions (hypothesis of restite control) as observed in some granites by White and Chap-

TABLE 1. Persistence of Tin Mineralization in Some Tin Provinces

Tin province	Age of mineralization (genetically related rock type)	Prominent metals	Reference
Erzgebirge, East Germany and Czechoslovakia	Proterozoic (ca. 700 m.y.) (Spilitic metavolcanics)	Sn, Cu, Zn	Baumann et al. (1976)
	Cambrian-Ordovician (Quartzites, conglomerates)	Sn, Zr	Baumann and Tischendorf (1978)
	Upper Carboniferous- Permian (Granites)	Sn, W, Bi, Zn, Ag, Pb	Weinhold (1977)
Jos Plateau, Nigeria	Proterozoic-Cambrian (Granites, pegmatites)	Sn, Nb, Ta, Be	Wright (1970), Kloosterman (1969), Tugarinov (1968)
	Jurassic (Granites)	Sn, W, Nb	
Bolivia Pando, Rondônia Eastern Andes	Proterozoic (ca. 1,000 m.y.) (Granites)	Sn, W, Nb, Ta	Kloosterman (1967) Priem et al. (1971)
	Upper Triassic (210 m.y.) (Granites)	Sn, W, Zn, Bi, Pb	Clark and Farrar (1973), Grant et al. (1977)
	Tertiary (3-28 m.y.) (Granites, dacitic-latitic stocks)	Sn, W, Bi, Zn, Ag, Pb, Sb	
Hunan, China	Precambrian (?) (Sedimentary Sn-W anomalies in Lower Cambrian metasediments)	Sn, W (?)	Meng and Chang (1935)
	Caledonian (Granites)	Sn, W	
	Cretaceous (Granites)	Sn, W, Pb, Zn, Bi, Mo, Ag, Sb	Hsieh (1963)
Indonesia	Upper Triassic (ca. 215 m.y.) (Granites)	Sn (placers)	Priem et al. (1975)
	Upper Cretaceous (ca. 75 m.y.) (Granites)	Sn (placers)	Jones et al. (1977)
Thailand	Upper Permian-Lower Triassic (240-210 m.y.) (Granites)	Sn, W, Zn, Pb	Beckinsale (1979)
	Lower-Middle Cretaceous (130-90 m.y.) (Granites)	Sn, W, Zn, Pb	Beckinsale et al. (1979)
NE Queensland, Australia	Proterozoic (ca. 1,500 m.y.) (Granites)	Au, Sn (weak)	Sheraton and Black (1973),
	Carboniferous (ca. 320 m.y.) (Granites)	Sn, W, Pb, Cu	Blake and Smith (1970)
	Permian (ca. 260 m.y.) (Granites)	Sn (weak)	

pell (1977). Although their model may apply to large batholiths in general, the examples of high level intrusives treated in this paper fit better a model of fractional crystallization. The main argument is the log variation pattern of several indicative trace elements (Rb, Sr, Ba, Cs, etc.) and detailed discussions can be found in the original papers listed in Table

2 from which the Sn data of this study have been compiled.

It must be pointed out that Sn data are not too abundant in the literature. Outside of tin provinces, studies of systematic changes of Sn content of granitic rocks during fractional crystallization are rare. Exceptions are the investigations of the Cape Granite,

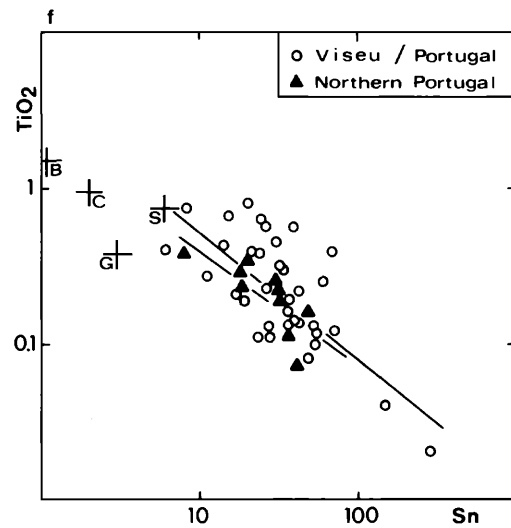
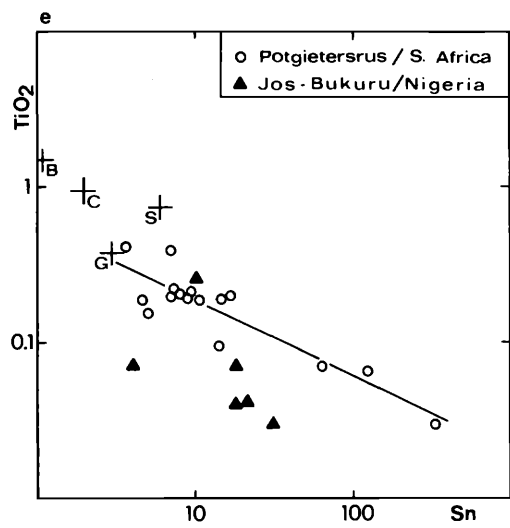
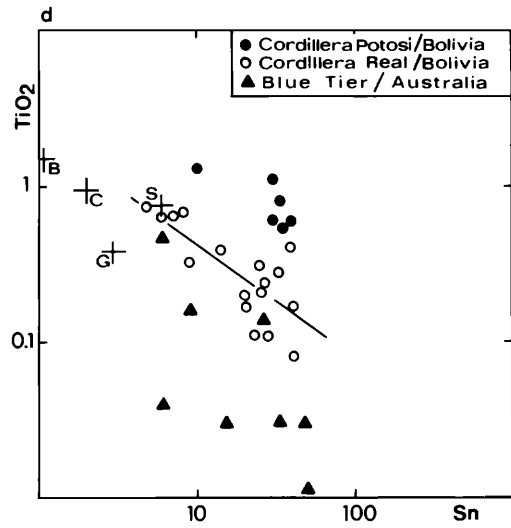
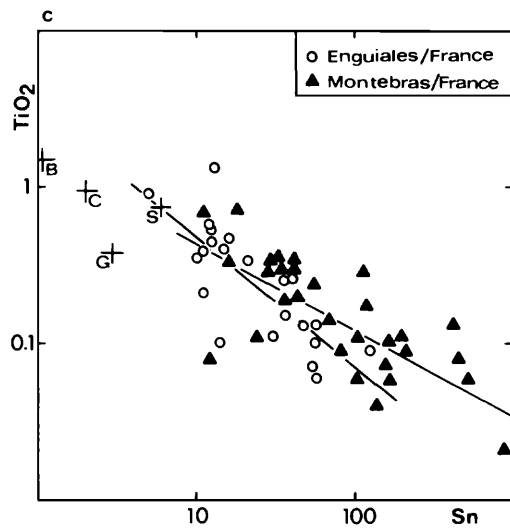
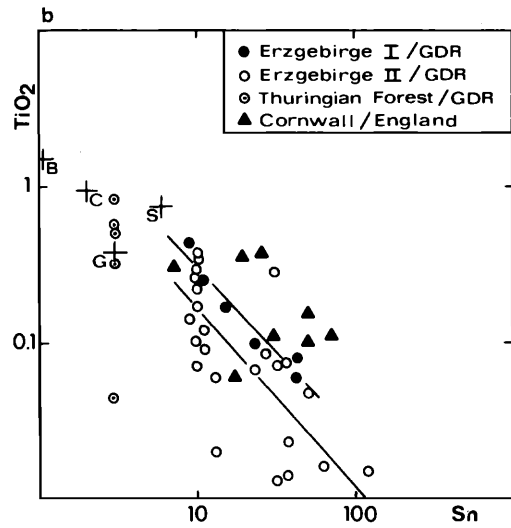
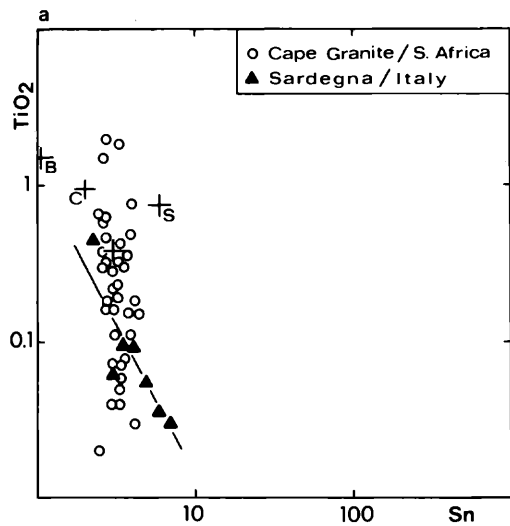


TABLE 2. Statistical Parameter and Data Sources to the Variation Diagrams of Figures 1 through 6 Magnetite series (M) or ilmenite series (I) affiliation in column 2 according to criteria by Ishihara (1977).

Sample location	Rock type	TiO ₂ -Sn diagrams			Rb/Sr-Sn diagrams			Reference
		Slope correlation line	Correlation coefficient	Number of samples	Slope correlation line	Correlation coefficient	Number of samples	
Cape Province, South Africa	Cambrian granites (M)			38			38	Kolbe (1966)
Southern Sardinia, Italy	Hercynian granites (M)	-1.90	-0.86	7 (means of 107)	2.20	0.92	6 (means of 107)	Biste (1979) (means recalculated)
Erzgebirge, East Germany (1)	Hercynian granites (I)	-1.08	-0.96	6 (means of 272)	2.20	0.92	6 (means of 272)	Lange et al. (1972) (only larger granitic outcrops, >10 km ²)
Erzgebirge, East Germany (2)	Hercynian granites (I)	-1.13	-0.78	26				Bräuer (1970)
Cornwall, England	Hercynian granites (I)			8			6	Hall (1971), Wilson (1972), Ghosh (1934), Alderton et al. (1980)
Montebras, Massif Central, France	Hercynian granites (I)	-0.55	-0.78	37	1.11	0.82	22	Boissavy-Vinau (1979), Burnol (1978)
Enguiales, Massif Central, France	Hercynian granites (I)	-0.83	-0.78	22	1.45	0.83	22	Boissavy-Vinau (1979), Burnol (1978)
Cordillera Real, Bolivia	Mesozoic granites (I)	-0.73	-0.76	17	1.44	0.75	18	Lehmann (1979)
Cordillera de Potosi, Bolivia	Tertiary granodiorite-latite (I)	-0.55	-0.76	6				Wolf (1975)
Blue Tier, Tasmania, Australia	Hercynian granites (?)	-0.92	-0.66	8 (means of 32)	2.37	0.90	7 (means of 30)	Groves and McCarthy (1978)
Potgietersrus, South Africa	Precambrian granites (M)	-0.73	-0.74	16 (means of 226)	0.87	0.98	8 (means of 226)	Lenthall and Hunter (1977)
Jos Plateau, Nigeria	Jurassic granites (I)			6			6	Imeokparia (1980)
Alijo-Sanfins, Portugal	Hercynian granites (I)	-0.49	-0.74	10 (means of 40)	1.35	0.77	16	Neiva (1975)
Viseu, Portugal	Hercynian granites (?)	-0.79	-0.71	35	0.80	0.62	35	Boissavy-Vinau (1979)
Thuringian Forest, East Germany	Hercynian granites (M)			5				Bräuer (1970)
Black Forest, West Germany	Hercynian granites (?)	TiO ₂ -F diagram: 13 samples (means of 400)						Emmermann (1977)

South Africa, by Kolbe (1966) and Kolbe and Taylor (1966a) and of the Snowy Mountains area, New South Wales, Australia, by Kolbe and Taylor (1966b). The Cape Granite data are plotted in Figures 1a and 2a; the Snowy Mountains data have been omitted but plot in the same field and show the same characteristics as the South African samples, i.e., no significant

change in Sn with a variation of TiO₂ and Rb/Sr of about two orders of magnitude.

A further example of a granite suite from a non-tin province is provided by the recent geochemical study of southwest Sardinia, Italy, by Biste (1979) (Figs. 1a and 2a). These granites are barren, but local occurrences of tin minerals of no economic signifi-

FIG. 1a-f. TiO₂-Sn variation diagrams for granite suites from various non-tin (a) and tin provinces (b-f). TiO₂ in weight percent, Sn in ppm. Data sources in Table 2.

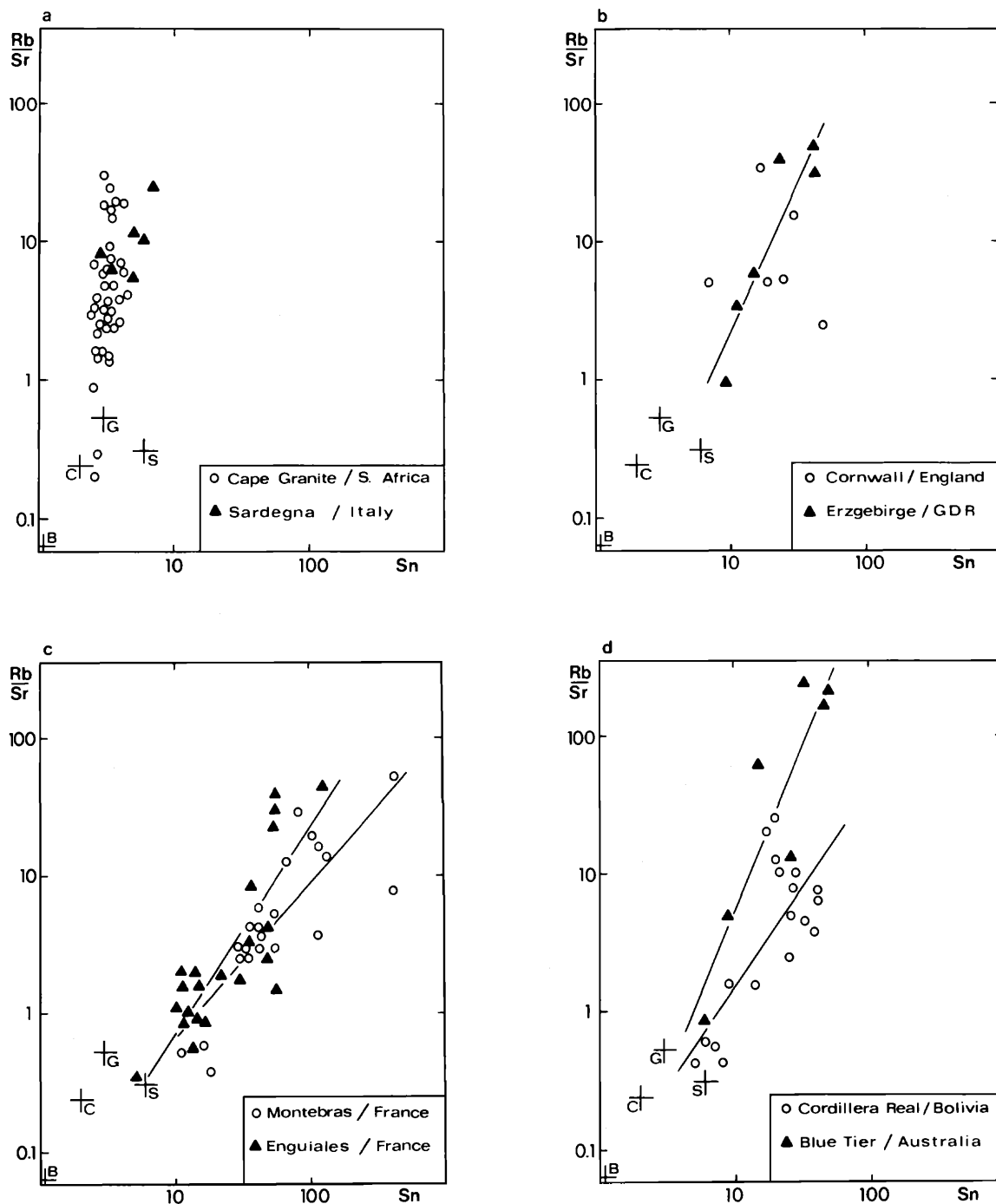


FIG. 2a-f. Rb/Sr-Sn variation diagrams for granite suites from various non-tin (a) and tin provinces (b-f). Sn in ppm. Data sources in Table 2.

cance are known. From the neighborhood of the Erzgebirge tin province, some analyses of the barren granites of the Thuringian Forest, East Germany, by Bräuer (1970) are plotted in Figure 1b.

Published Sn data for granitic series in tin provinces

have been found for the Erzgebirge and Cornwall (Figs. 1b and 2b), for the Massif Central, France (Figs. 1c and 2c), for Andean Bolivia and Tasmania (Figs. 1d and 2d), for the Bushveld and northern Nigeria (Figs. 1e and 2e), and Portugal (Figs. 1f and 2f). The

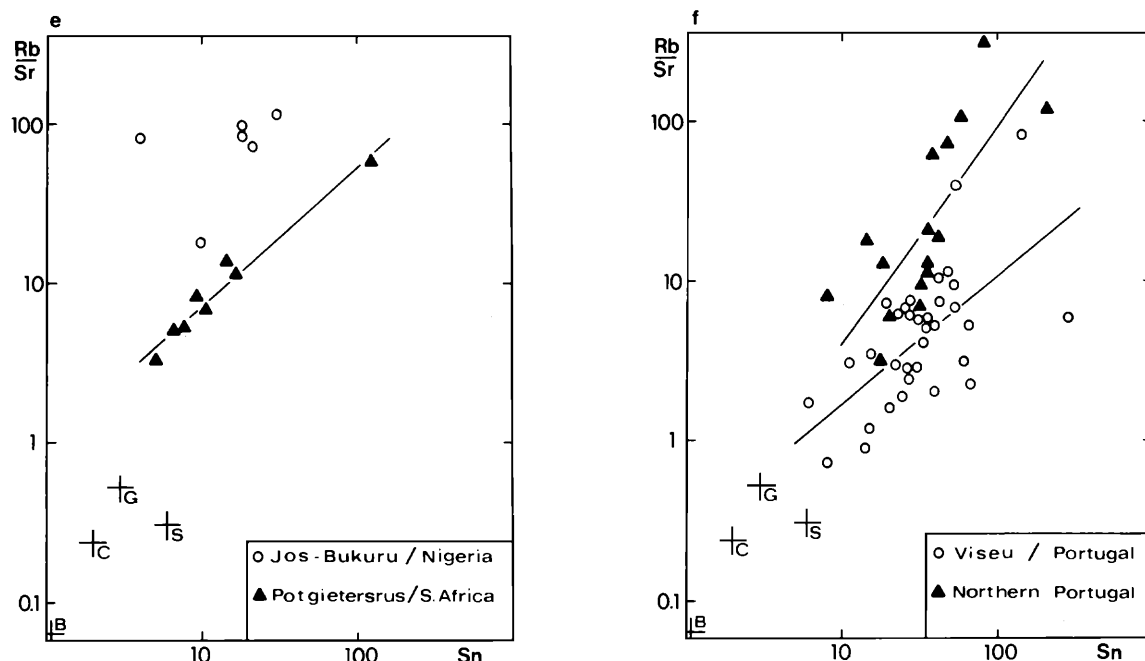


FIG. 2. (Continued).

lack of systematic geochemical studies of comagmatic series in the southeast Asian tin belt is lamentable.

As a consequence of the varying availability of geochemical data in different areas, data points refer to single sample analyses in some cases and to averages of numerous samples in other cases. Statistical parameter and data sources are listed in Table 2. Correlation lines have only been drawn in those cases where the correlation as a function of the number of data and the correlation coefficient can be considered valid at 95 percent probability or more. As reference points, the global averages for basalt (B), crust (C), granite (G), and shale (S) have been plotted in each diagram.

Discussion

The distribution trends of the granitic samples in Figures 1 and 2 are not against the interpretation as fractionation suites, as concluded by earlier workers from other trace element variations of the same sample suites. The scatter is probably related to problems of representative sampling, as tin is mostly concentrated in accessory minerals (see below). A synoptic view of the tin distribution as a function of the two differentiation indices TiO_2 and Rb/Sr is given in Figure 3a and b. It can be noticed that the fractionation suites are traceable back to rocks of average crustal composition. Hence, the assumption of a primary tin enrichment beyond average shale composition is not justified.

A possible exception might be the Tertiary tin subprovince in central and southern Bolivia (Cordillera de Potosi samples). Although the number of data points is very limited, a speculative explanation for the position of the samples in Figure 1d could be mixing of average andesitic rocks with highly fractionated material of Mesozoic tin granites. Upper Triassic tin granites and Tertiary tin porphyries are reported not only to occur close together (Cordillera Real) but also superimposed in the same area (Wolf, 1975; Wolf and Pilot, 1980).

A second observation arising from Figure 3a and b is the different slope of the fractionation series of different areas. The relative position of the individual correlation lines largely coincides in both diagrams. It appears that the bulk distribution coefficient of Sn is variable for granitic rocks and, given an effective fractionation mechanism, will critically control the potential for tin mineralization of a granitic intrusive. Modal composition cannot be the critical factor, as the mineralogy of tin and non-tin granites does not seem to be clearly different. A possible explanation is offered by the hypothesis of Ishihara (1977, 1978) that tin under magmatic conditions may be able to exist in the tetravalent or bivalent state corresponding to the oxygen fugacity of the melt.

The principal tin-bearing minerals in granitic rocks are sphene, ilmenite, magnetite, biotite, and hornblende (Hamaguchi and Kuroda, 1969; Tischendorf, 1970). The preferential entry of tin into these min-

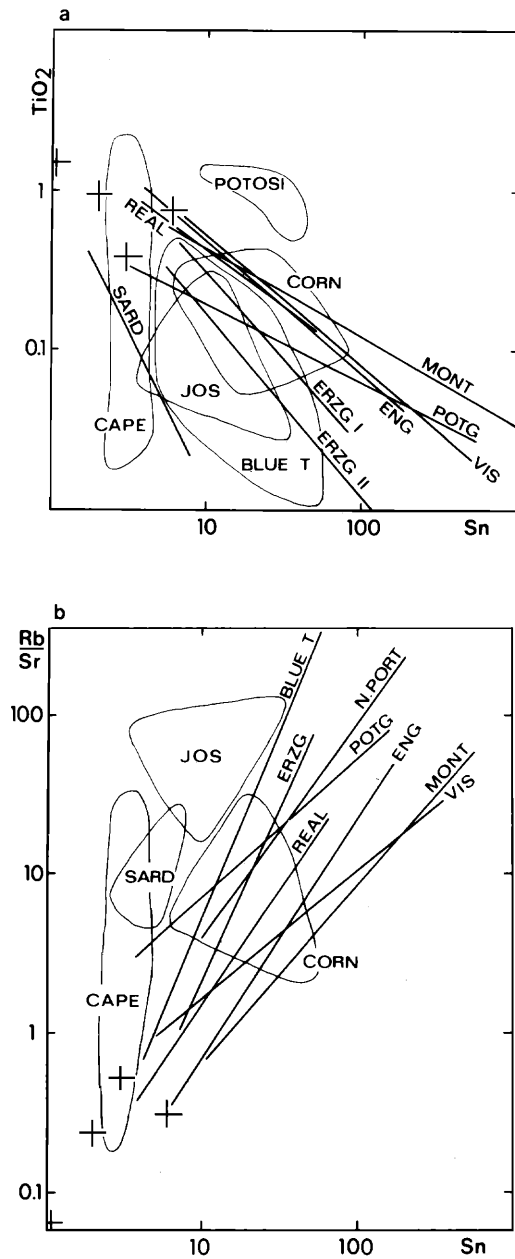


FIG. 3a and b. Synoptic variation diagrams, compiled from Figures 1 and 2.

erals is explained by the crystal-chemical affinity of Sn^{+4} to substitute Ti^{+4} and Fe^{+3} . Under conditions of high oxygen fugacity, i.e., a high $\text{Fe}^{+3}/\text{Fe}^{+2}$ ratio (magnetite series granitoids of Ishihara, 1977), tin may be present in the tetravalent state and therefore become incorporated in accessory minerals such as magnetite, sphene, and ilmenite during early stages of crystallization, thus giving rise to a relatively low tin concentration in later crystallization stages. In con-

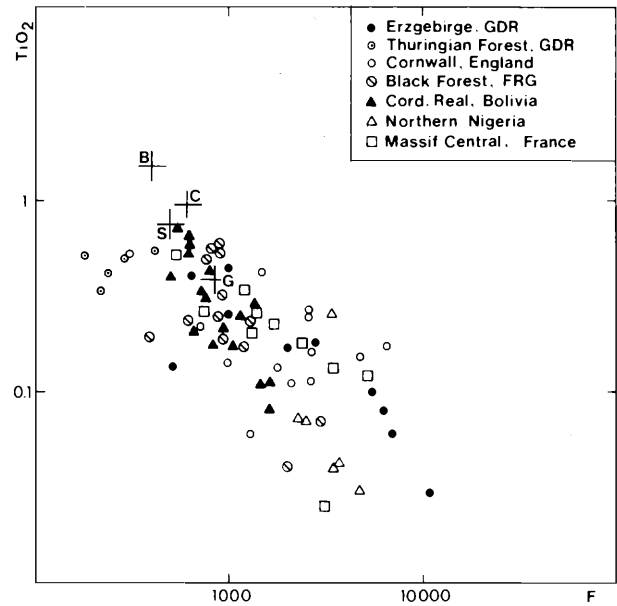


FIG. 4. TiO_2 -F variation diagram for granite suites from non-tin provinces (Thuringian Forest, Black Forest) and tin provinces (remainder).

trast, however, tin may be in a predominantly bivalent state in low f_{O_2} magmas (ilmenite series granitoids of Ishihara, 1977), thereby precluding its widespread entry into rock-forming minerals and favoring accumulation in the residual liquid.

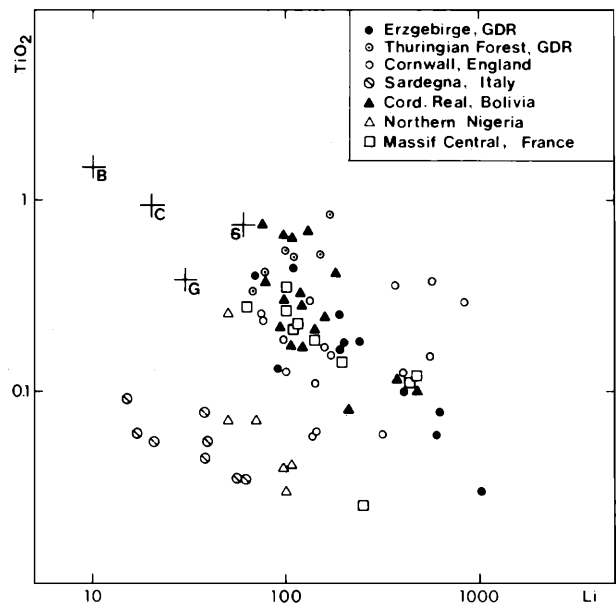


FIG. 5. TiO_2 -Li variation diagram for granite suites from non-tin provinces (Thuringian Forest, Sardinia) and tin provinces (remainder).

This model has been successfully tested in Japan and southeast Asia where Ishihara (1980) and Ishihara et al. (1980) could demonstrate the metallogenic relationship between tin mineralizations and ilmenite series granitic rocks. Nevertheless, its thermodynamic background is poorly understood because ionic equilibria of tin species at high temperatures are unknown. Calculations of Sn/SnO_2 and SnO/SnO_2 equilibria show that bivalent oxidic tin compounds are unstable at magmatic temperatures.

The low oxygen fugacity of the ilmenite series granitic magmas is probably a result of partial melting of carbonaceous metasediments which will adjust f_{O_2} of the melt near the equilibrium curve of the reaction $\text{CH}_4 + 2\text{O}_2 = \text{CO}_2 + 2\text{H}_2\text{O}$ (Ohmoto and Kerrick, 1977), below the quartz-magnetite-fayalite buffer.

In order to test the claim of extraordinary richness of tin granites in Li, B, and F, corresponding diagrams have been plotted in the above scheme. For reasons of space, these plots are only reproduced here for the TiO_2 fractionation index (Figs. 4–6). In Figures 4 and 5 it can be seen that the samples from tin provinces fall into the general fractionation trend given by the reference points of global averages. The TiO_2 -B plot in Figure 6 depicts significant differences in boron content between various tin provinces. Whereas the Erzgebirge province shows low boron values and no enrichment in boron during magmatic evolution, other tin provinces are extremely high in boron (Cornwall, Bolivia, Portugal). This may well reflect initial composition differences of the source rocks, corresponding to the distinct separation in boron content between average shale and average crustal rocks.

Conclusions

The presented geochemical trends do not support the view that tin provinces represent crustal anomalies in tin. Fractionation series of tin granites can be traced back to average granitic rocks. The variable bulk distribution coefficient of tin, as deduced from the TiO_2 -Sn and Rb/Sr-Sn plots of different granite series, controls the degree of tin enrichment during magmatic differentiation.

The variability of $\bar{D}_{\text{Sn}}^{\text{sl/liq}}$ seems to be related to the $\text{Sn}^{+2}/\text{Sn}^{+4}$ ratio in the melt, low f_{O_2} favoring enrichment of Sn in residual liquids (ilmenite series granitic rocks). Conditions of low oxygen fugacity can be expected during partial melting of thick metasedimentary sequences of black shales and graywackes, typical of tin provinces: e.g., the Erzgebirge (Weinhold, 1977), the Bolivian eastern Andes (Martinez, 1980), Portugal (Schermerhorn, 1981), or the southeast Asia tin belt (ESCAP/United Nations, 1978).

An ensialic character of tin provinces and a crustal origin of most tin granites can be deduced from geo-

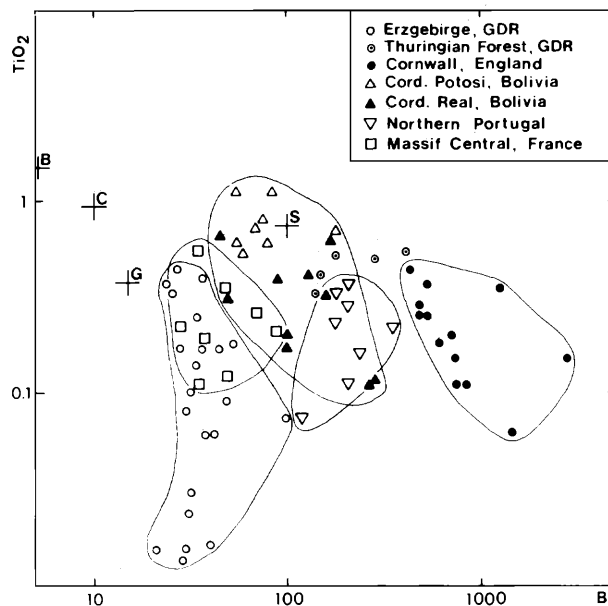


FIG. 6. TiO_2 -B variation diagram of granite suites from several tin provinces and from the Thuringian Forest. TiO_2 in weight percent, B in ppm.

tectonic observations, from the high initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, and other S-type features (White et al., 1977; Sillitoe, 1981). The persistence of tin in some tin provinces (étagement temporel) may be related to multiple melting of a carbon-rich metallotect which can provide the low f_{O_2} conditions necessary for effective tin concentration during magmatic differentiation.

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