## ORIGIN OF MAGNETITE- AND ILMENITE-SERIES GRANITIC ROCKS IN THE JAPAN ARC

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ABSTRACT. The exposure areas, radiometric ages, redox states, initial  ${}^{87}$ Sr/ ${}^{86}$ Sr and  ${}^{143}$ Nd/ ${}^{144}$ Nd isotope ratios, and  $\delta^{18}$ O values of granitic plutons in the Japan Arc have been compiled based on the results of previous studies of the Japanese granitic rocks. The exposure areas of the granitic rocks in a given period ( $\approx$  activity of granitic rocks) and initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios show positive correlations with the convergence rate of oceanic plates along the arc. In contrast, the redox states of the granitic rocks are negatively correlated with the oceanic plate convergence rate. The initial  ${}^{143}$ Nd/ ${}^{144}$ Nd isotope ratios and  $\delta^{18}$ O values combined with the initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios suggest that the temporal variation of initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios was mainly due to source contamination resulting from melting of subducted materials in the mantle source region. The granitic magmas contaminated with more than about 15 percent subducted materials became ilmenite-series granitic rocks.

These observations suggest that temporal variations in redox states in the Japanese granitic rocks are not due to the properties of the lower crust but rather to subducted materials. Sediment subduction was also increased with increasing convergence rate. Consequently, the oxidation states of granitic rocks declined and the initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios rose, probably due to the increase in the assimilation rates of subducted sediments into granitic magmas. The increase of sediment subduction may be attributable to the acceleration of subduction-related erosion and/or the changes in the Earth's surface conditions (the high CO<sub>2</sub> fugacity and temperature of the Cretaceous atmosphere, which promoted increased weathering and erosion of continental crust resulted in greater terrigenous sediment supply to subduction zones). The active deposition and subduction of carbon- and sulfide-rich sediments, caused by oceanic anoxic events during the Cretaceous, would also promote the extensive reduction of magma source regions to form ilmenite-series granitic rocks.

#### INTRODUCTION

The magnetite- and ilmenite-series granite classification is based on magnetite contents of granitic rocks, and the boundary of the contents is 0.1 modal percent (Ishihara, 1977). In practice, the two granite series are identified by their magnetic susceptibility values (Kanaya and Ishihara, 1973; Ishihara, 1979) because modal contents of magnetite in rocks are positively correlated with their magnetic susceptibility (for example, Balsley and Buddington, 1964); granites showing a value of  $\geq 100 \times$  $10^{-6}$  emu/g ( $\approx 3.0 \times 10^{-3}$  SI units) are classified as belonging to the magnetite-series (Ishihara, 1981). As the granite series reflects redox states of granitic magmas, the magnetite- and ilmenite-series correspond to oxidized- and reduced-type, respectively. The distributions of the two granitic series are closely linked to the metallogenic provinces in general; that is, sulfide deposits, such as Cu, Pb, Zn, and Mo, and oxide deposits, such as W and Sn, tend to be formed with magnetite- and ilmenite-series granitic rocks, respectively (Ishihara, 1971, 1977, 1981, 1984, 1998). Early studies indicated that the redox states are not intrinsic properties of granitic magmas but are acquired due to the physicochemical conditions of magma chambers (for example, Czamanske and others, 1981). Since the 1980's, there have been a number of geological and petrological studies of granite series in the circum-Pacific regions. These studies have indicated that the granite series is generally stable during a sequence of regional plutonism even though oxidation or reduction may occur locally

by degassing or assimilation of wall rocks *in situ*, and each granite series tends to form a terrain of more than 100 kilometers in length (for example, Ishihara and Ulriksen, 1980; Ague and Brimhall, 1988a, 1988b; Bateman and others, 1991; Gastil and others, 1990; Blevin and Chappell, 1995; Ishihara and Wang, 1999). Carmichael (1991) also established that the redox states of both silicic and basic magmas are inherited from their respective source regions based on the ferric/ferrous iron ratios of numerous glassy lavas in various geological settings. These results indicate that the redox states of granitic magmas are essentially intrinsic in nature; that is, they are a reflection of the redox states of magma source regions.

Takagi and Tsukimura (1997) showed thermodynamically that redox states of granitic magmas are linked to the  $SO_2$  concentration of the magmas, because  $SO_2$  gas acts as an oxidizing agent for silicate melts during late-magmatic to subsolidus stages. When a magma contains  $SO_2 > 250$  ppm, it becomes a magnetite-series granitic rock. In contrast, ilmenite-series magmas contain little  $SO_2$ , probably because most  $SO_2$  has been reduced to  $H_2S$  by reducing agents, such as  $CH_4$  and CO.

Sulfur isotope compositions of magnetite-series granitic rocks are generally heavier than those of ilmenite-series granitic rocks;  $\delta^{34}$ S values of the former range from -2 to +10, while those of the latter range from -10 to +2 (Sasaki and Ishihara, 1979; Ishihara and Sasaki, 1989, 2002). The difference in sulfur isotope compositions is attributable to differences in sulfur sources between the two series of magmas, because there is little isotopic fractionation of sulfur at magmatic temperatures (Ohmoto and Goldhaber, 1997); the heavier sulfur in magnetite-series granitic rocks is mainly derived from seawater as sulfate, whereas the lighter sulfur in ilmenite-series granitic rocks is mainly derived from sedimentary rocks as sulfide. In the Japan Arc, the most likely source of seawater sulfate for magnetite-series granitic rocks is subducted altered oceanic crust that interacted with seawater in/around ridges, whereas sedimentary sulfide for ilmenite-series granitic rocks is probably derived from subducted sediments on oceanic plates.

It is shown here that the amounts and properties of subducted materials control the redox states of granitic magmas (and therefore granite series) based on a compilation of data related to the Japanese granitic rocks, including exposure areas, initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  and  $^{143}\mathrm{Nd}/^{144}\mathrm{Nd},\delta^{18}\mathrm{O}$  values, and convergence rate of oceanic plates from previous studies. The close relationships between Earth's surface conditions and the nature of subducted sediments are also discussed.

### METHODS

## Spatiotemporal Distribution of Granitic Rocks

Activity of granite plutonism within a given period may be estimated from the total volume of plutons intruded within the period. However, the most practical way to estimate activity is the measurement of exposure areas of plutons intruded within a given period. For this measurement, geological information regarding individual plutons, such as radiometric ages and the shape of each intrusion, were attached to a 1/1,000,000 geological map of Japan (Geological Survey of Japan, 1992), and their exposure areas were measured. In the Japan Arc, there are a few Quaternary volcanoes in major granite fields, such as the Chugoku, southern Kinki-Chubu, and eastern Tohoku districts (fig. 1). The areas of granitic rocks covered by Quaternary volcanic strata, which cause errors in the expected activity, are estimated to be less than about 12 percent of the entire granite exposure based on the overlaps of granite terrains and Quaternary volcanic fields. Age estimates were made using Rb-Sr whole-rock isochron ages for most plutons. However, K-Ar or Rb-Sr mineral isochron ages for the plutons were used if no Rb-Sr whole-rock isochron ages were available. Differences between dating methods do not cause significant errors because K-Ar and Rb-Sr whole-rock



Fig. 1. Distribution of magnetite- and ilmenite-series granitic rocks in the Japan Arc after Ishihara and others (1992a, 1992b), and locations of the districts referred to in the text. MTL= median tectonic line (a large transform fault).

isochron ages of the Japanese granitic rocks generally agree within  $\pm$  5 million years (Shibata and Ishihara, 1979b). In the case of the plutons with no radiometric ages, the age of a neighboring pluton with similar rock facies was applied; the areas of these plutons were less than 5 percent of the entire granite exposure.

In part of the Chubu-Ryoke belt there are some plutons whose K-Ar (biotite) and Rb-Sr mineral isochron ages are more than 20 million years younger than those of Rb-Sr whole-rock isochron ages (for example, Yuhara and Kagami, 1996; Yuhara and others, 2000). Suzuki and Adachi (1998) reexamined the plutons geochronologically using CHIME (Chemical Th-U-total Pb Isochron Method), and obtained CHIME monazite ages that were consistent with intrusive relationships of the plutons, and were in near agreement with Rb-Sr whole-rock isochron and CHIME ages of the Setouchi-Ryoke belt plutons that have similar rock facies as those in the Chubu-Ryoke belt (Suzuki and others, 1996). The U-Pb zircon ages of some Ryoke belt plutons are essentially consistent with their CHIME and Rb-Sr whole-rock isochron ages (Herzig and others, 1998). Thus, the discrepancies among the age dating methods have been explained by differences not in the intrusive ages, but the more rapid denudation of the Setouchi-Ryoke belt as compared with the Chubu-Ryoke belt (Suzuki and Adachi, 1998; Yuhara and others, 2000). In this study, therefore, the Rb-Sr whole-rock isochron and CHIME ages for the plutons in the Chubu-Ryoke belt were adopted because intrusive ages are necessary to estimate the activity of granite plutonism.

### Redox States of the Japanese Granitic Rocks

To estimate the redox states of the granitic rocks, each pluton was distinguished as belonging to the magnetite- or ilmenite-series according to Ishihara and others (1992a, 1992b) and Tanaka (1992), and the areal ratios of the two granite series were calculated in units of 5 million years. The frequencies of magnetic susceptibility values of the Japanese granitic rocks clearly shows a bimodal distribution (Ishihara, 1990); the areal ratios of two-series granitic rocks may approximate the average of redox state of the Japanese granite plutons in a given period. Moreover, two rectangular fields with dense and continuous magnetic susceptibility measuring points were selected in the Chugoku district, and the correlation between redox states and initial <sup>87</sup>Sr/<sup>86</sup>Sr of granitic rocks was examined more closely.

Initial <sup>87</sup>Sr/<sup>86</sup>Sr, <sup>143</sup>Nd/<sup>144</sup>Nd, and Oxygen Isotope Ratios of the Japanese Granitic Rocks

The initial <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios of the granitic rocks yielded by the Rb-Sr whole-rock isochrons only were compiled. Initial <sup>143</sup>Nd/<sup>144</sup>Nd isotope ratios were calculated from the present ratios using the Rb-Sr whole-rock isochron ages of the same pluton. When a pluton yielded multiple <sup>143</sup>Nd/<sup>144</sup>Nd isotope ratios, the average and deviations of the ratios are indicated. The  $\delta^{18}$ O values (whole-rock  $\delta^{18}$ O values relative to Standard Mean Oceanic Water, ‰) of the Japanese granitic rocks were also compiled. Although isotopic fractionation of the oxygen isotopes with increasing SiO<sub>2</sub> contents of granitic rocks has been identified, raw values were used in this study because the differences between raw and corrected values are negligible for the purpose of this discussion.

## Convergence Rate of Oceanic Plates Beneath the Japan Arc

To estimate the convergence rate of subducted oceanic plates beneath the Japan Arc, relative plate motions between oceanic and continental plates were compiled based on the positions of hotspots (Engebretson and others, 1985). To calculate convergence rate, the direction of the trench along the Japan Arc was fixed at N45°E, and the linear velocity and azimuths of plate motions at latitude 40°N and longitude 140°E were used as representative of the Japan Arc (fig. 2).

### RESULTS

## Activity of Japanese Granite Plutonism

The Japanese granite plutonism was continuous from the Jurassic to the present. Figure 3B shows four prominent peaks of granite plutonism: (1) 120 to 110 Ma. This peak comprises the plutons in the Tohoku and northern Kyushu districts; (2) 90 to 85 Ma. This peak is the most prominent peak; plutonism occurred in extensive areas of southwest Japan. The activity declined abruptly at about 80 Ma; (3) 70 to 55 Ma. This peak is mainly composed of the magnetite-series plutons in the back-arc side of southwest Japan. (4) During 10 to 15 Ma, a small peak was recognized; Miocene plutonism characteristically includes S-type granitic rocks (in particular the southern Kyushu district), and their origin may be different from the older granitic rocks with I-type chemistry (Nakada and Takahashi, 1979; Ishihara and Matsuhisa, 1999).

Figure 3A also shows the variations of calculated convergence rates. Figure 4 shows the relationship between the convergence rate and the Japanese granite plutonism. The convergence rate was positively correlated with the activity of the Japanese granitic rocks, strongly suggesting a genetic relationship with a high correlation coefficient.

Figure 5 shows the active fields of granite plutonisms corresponding to the peak



**P**: Approximate location (lat. 40°N - long. 140°E) to calculate relative plate motions based on Engebretson and others (1985).

Fig. 2. Tectonic setting and distribution of Cretaceous granitic rocks in the Eurasian continental margin. Modified after Owada and others (1999).

granite activity periods. As shown in figure 5, extensive ilmenite-series granite activity occurred in areas from the Chugoku-Shikoku to Kinki-Chubu districts over a short period from 95 to 70 Ma.

### Redox States of the Japanese Granitic Rocks

Since the Jurassic, continuous magnetite-series granite activity occurred during all geological periods. The generation of ilmenite-series granitic rocks began at about 150 Ma, and gradually spread. During the period from 95 to 70 Ma, the percentage of ilmenite-series granitic rocks relative to all granitic rocks reached 85 to 98 percent (figs. 5 and 6). The plutons intruded during the dominant ilmenite-series period tended to form huge batholiths, and the exposure areas of the batholiths occupy about 60 percent of the total exposure area of the Japanese granitic rocks. The activity of ilmenite-series granitic rocks declined rapidly at about 70 Ma, and ceased at about 30 Ma with the exception of Miocene S-type granitic rocks. Figure 7 shows the relationship between the convergence rates of oceanic plates and percentage of ilmenite-series granitic rocks. The diagram shows a positive correlation, although the points are more scattered than those shown in figure 4. Thus, the redox states of the Japanese granitic rocks appear to be associated with the convergence rate.



Fig. 3. (A) Variation of convergence rate of oceanic plates at latitude 40°N – longitude 140°E in the unit of 5 million years using the data of Engebretson and others (1985). Upper bar shows the names of subducting oceanic plates at the time; (B) Variation of exposure area of magnetite- and ilmenite-series granitic rocks in the Japan Arc in units of 5 million years.

# Relationship Between Redox States and Initial <sup>87</sup>Sr/<sup>86</sup>Sr Isotope Ratios

Figure 8 shows the variations of magnetic susceptibility (Ishihara, 1979) and initial <sup>87</sup>Sr/<sup>86</sup>Sr (Kagami and others, 1992) along two cross-sections through these areas. Negative correlations were detected between the variation trend of magnetic susceptibility and that of initial <sup>87</sup>Sr/<sup>86</sup>Sr in both sections. In the area shown on figure 8A, a



Fig. 4. Relationship between the exposure area of the Japanese granitic rocks and convergence rate of oceanic plates at the Japan Arc in units of 10 million years.

similar negative correlation between magnetic susceptibility and  $\delta^{18}$ O of quartz has also been reported previously (Ishihara, 1981).

# Temporal Variations of Initial <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd Isotope Ratios of the Japanese Granitic Rocks

Figure 9A shows the relationship between Rb-Sr whole-rock isochron ages and initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ . The average initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  generally increases gradually from 0.704 to 0.712 from 150 to 70 Ma, subsequently falling to 0.705 during 70 to 60 Ma. Particularly from 150 to 50 Ma, initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  variations of ilmenite- and magnetite-series show two contrasting trends; both showed good fits to quadratic regression curves (fig. 9B). Although the reasons why the trends fit the quadratic curves are not clear, it is evident that the initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  ratios of two granite series vary systematically with time. Positive correlations were found between the initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  ratio is also correlated with the spatiotemporal pattern of granite plutonism. The relationship between Rb-Sr whole-rock isochron ages and initial  $\epsilon_{\rm Nd}$  value were examined in the same way as the initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  ratio (fig. 11). The relationship between Rb-Sr whole-rock isochron ages and initial  $\epsilon_{\rm Nd}$  values was also similar to that of the initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  ratio even though the plots were scattered to a greater extent.

# Initial <sup>87</sup>Sr/<sup>86</sup>Sr Versus $\delta^{18}O$ of the Japanese Granitic Rocks

Figure 12 shows initial  ${}^{87}$ Sr/ ${}^{86}$ Sr versus  $\delta^{18}$ O to determine whether the trends of the plots are due to source contamination or crustal contamination, as suggested by



Fig. 5. Active fields of granitic rocks corresponding to each peak period of the Japanese granitic plutonisms.



Fig. 6. Variation of the percentage of ilmenite-series granitic rock areas for the Japanese granitic rocks in units of 5 million years. The period from 75 to 110 Ma is the ilmenite-series dominant period in the Japanese granitic plutonism. OAE= oceanic anoxic events (Jenkyns, 1980).



Fig. 7. Relationship between the percentage of ilmenite-series granite areas for the Japanese granitic rocks and convergence rate of oceanic plates in units of 10 million years.







Fig. 9. (A) Relationship between initial <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios and ages of the Japanese granitic rocks yielded by Rb-Sr whole-rock isochrons. Error bars means the errors due to the dispersion of Rb-Sr isochron plots; (B) Correlative relationships between the initial <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios and ages during 150 to 50 Ma. X and Y are the quadratic regression curves of ilmenite- and magnetite-series granitic rocks, respectively. Raw data are listed in appendix A.

James (1981). Most of the plots display trends typical of source contamination. To draw mixing curves in figure 12, the Assimilation – Fractional Crystallization (AFC) model of DePaolo (1981) was used, and the  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  and  $\delta^{18}\mathrm{O}$  of the primitive magma were assumed have values of 0.7035 and +6.5, respectively, which are normal mantle rock compositions. The ratio of the mass assimilation rate to extent of fractional crystallization (r), and the partition coefficient of Sr between bulk solid and liquid (D<sub>Sr</sub>), were assumed to be 0.5 and 1.2, respectively. Even if the variables are changed to other possible values, the modified mixing curve should also yield similar results, because the fitting curves are strongly convex downward. Isotopic compositions of assimilated material, in particular  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  and  $\delta^{18}\mathrm{O}$  are 0.7085 and 20, respectively (curves A and B). However, some plots deviate to the higher  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  side, suggesting assimilants of



Fig. 10. Relationship between the averages of initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios of the two granite series in the Japan Arc yielded by Rb-Sr whole-rock isochrons and convergence rate of oceanic plates in units of 10 million years.



Fig. 11. (A) Relationship between initial <sup>143</sup>Nd/<sup>144</sup>Nd isotope ratios and ages of the Japanese granitic rocks yielded by Rb-Sr whole-rock isochrons. The plot and error bar mean the average and variation of the isotope ratios in a pluton, respectively. (B) Correlative relationships between the initial <sup>143</sup>Nd/<sup>144</sup>Nd isotope ratios and ages during 150 to 50 Ma. X and Y are the quadratic regression curves of ilmenite- and magnetite-series granitic rocks, respectively. Raw data are listed in appendix A.

higher  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ . Thus, the curve using the assimilant composition of  ${}^{87}\text{Sr}/{}^{86}\text{Sr} = 0.713$  and  $\delta^{18}\text{O} = +20$  is shown for reference (curve C). The shape of the curves is mostly dependent on the ratio of the Sr concentration between magma and assimilant  $(C_m/C_a)$ ; the ratio of curve B is 0.08. This low value indicates that the magma is strongly depleted relative to the assimilants, suggesting that the contamination resulted from melting of subducted materials in the mantle source region.

Some granitic rocks of the Kitakami and northern Kyushu districts show compositions that are plotted on/around the curve defined for  $C_m/C_a = 1.0$  (curve A). The granitic rocks of the Kitakami and northern Kyushu districts (fig. 1) commonly show Sr/Y ratios of > 25, and the granitic rocks have been partly classified into adakite (Tsuchiya and Kanisawa, 1994; Kamei, 2004). Therefore, the high initial Sr concentration of the magmas would be reflected in the plots on/around curve A.



Fig. 12. Relationship between initial  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios and  $\delta^{18}$ O values of the Japanese granitic rocks.  $C_m$  = concentration of the element in magma;  $C_a$  = concentration of the element in assimilant; Small numerals on mixing curves = mass of magma/initial mass of magma. Raw data of  $\delta^{18}$ O values are listed in appendix A.

In figure 12, the plots of magnetite-series and ilmenite-series are clearly separated by  $\delta^{18}$ O; the upper limit of the mass of assimilant in magnetite-series granitic magmas is about 15 percent with a few exceptions.

# Initial <sup>87</sup>Sr/<sup>86</sup>Sr Versus <sup>143</sup>Nd/<sup>144</sup>Nd Isotope Compositions of the Japanese Granitic Rocks

Figure 13 shows three mixing curves drawn by the AFC model using the same r and  $D_{Sr}$  as those in figure 12. The partition coefficient of Nd between bulk solid and liquid ( $D_{Nd}$ ) was assumed to be 0.05. To draw the fitting curve A, the  ${}^{87}Sr/{}^{86}Sr$  ratio of the primitive magma and assimilant and  $C_m/C_a$  (Sr) were set to the same values as used in figure 12, and appropriate  ${}^{143}Nd/{}^{144}Nd$  of the primitive magma and assimilants and  $C_m/C_a$  (Nd) were defined as 0.5131, 0.5122, and 0.03, respectively. Curve A suggests that the primitive magma was strongly depleted, similar to mantle materials, and thus the isotopic variations of the Japanese granite magmas were formed mainly through the process of source contamination.



Fig. 13. Relationship between initial <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios and initial <sup>143</sup>Nd/<sup>144</sup>Nd isotope ratios of the Japanese granitic rocks. The meaning of each symbol and numeral is the same as those in figure 12.

In figure 13, another mixing curve on the lower <sup>143</sup>Nd/<sup>144</sup>Nd side than curve A can be generated using values of 0.713 and 0.5120 for the <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd ratios of assimilants, respectively. This isotopic composition for assimilants is equivalent to those of the Ryoke metamorphic rocks at 85 Ma (Yuhara and Kagami, 1999). The Ryoke metamorphic rocks are high-temperature and low-pressure type metamorphic rocks constituting the upper to middle crust, and occur as the main host rock of the granitic rocks in the Ryoke belt (Nakajima, 1994; Brown, 1998). To draw the fitting curve (curve B), it is necessary to increase  $C_m/C_a$  (Sr) from 0.08 to 0.5 and  $C_m/C_a$  (Nd) from 0.03 to 0.1, because the curve is strongly convex downward relative to curve A; the increases mean a shift from source contamination toward crustal contamination. Similar to figure 12, the granitic rocks in the Kitakami district and a portion of the granitic rocks in the northern Kyushu district show isotopic compositions plotted on/near curve B is probably due to the intrinsic high Sr contents of the magmas.

The Miocene S-type granitic rocks show a distinctive trend in figure 13; another mixing curve could be generated. Nakada and Okamoto (1984) demonstrated that the AFC process between intermediate magmas of I-type chemistry and the pelite-rich continental crust may be applicable to the origin of the S-type granitic rocks. Thus, a primitive magma of <sup>87</sup>Sr/<sup>86</sup>Sr = 0.7045 and <sup>143</sup>Nd/<sup>144</sup>Nd = 0.51264 was tentatively assumed for the primitive magma of the S-type granitic rocks, which is the composition of magnetite-series granitic rocks on curve A. As the main host rock of the S-type granitic rocks is sedimentary rock of the Shimanto group, which is an Early Cretaceous to Paleogene accretionary complex, the average isotopic compositions of the Shimanto group at 14 Ma (<sup>87</sup>Sr/<sup>86</sup>Sr = 0.7125, <sup>143</sup>Nd/<sup>144</sup>Nd = 0.51235, Terakado and others, 1988) were applied for assimilants. At C<sub>m</sub>/C<sub>a</sub> (Sr) = 0.35 and C<sub>m</sub>/C<sub>a</sub> (Nd) = 0.1, the curve (curve C) shows a good fit to the plots of the S-type granitic rocks.

Consequently, the curves A, B, and C in figure 13 indicate that the granitic rocks dominated by source contamination are in the majority. However, there are also several granitic plutons dominated by crustal contamination in the Japan Arc.

In figure 13, the plots of magnetite-series and ilmenite-series are separated by the lines of  ${}^{87}\text{Sr}/{}^{86}\text{Sr} = 0.706$  and  ${}^{143}\text{Nd}/{}^{144}\text{Nd} = 0.5124$  with some exceptions. Similar to the results shown in figure 12, figure 13 also shows that the assimilation rates of subducted and crustal materials into primitive magmas determine whether a granitic magma becomes magnetite-series or ilmenite-series, at least in the Japan Arc.

### DISCUSSION

### Origin of the Late Cretaceous Pulse in Granite Plutonism

The data presented above suggest that there is a correlation between spatiotemporal variations in granite plutonism in the Japan Arc, in particular the pulse in plutonism in the Late Cretaceous, and the increase in convergence rate of subducted oceanic plates. Two genetic models may explain the increase in granite activity with the increase in convergence rate:

- (1) With increased convergence rate, the supply of arc-related basaltic magmas would increase as larger amounts of slab-derived aqueous fluid are added to the mantle wedge. This process in turn would increase the production of granitic magmas by the injection of voluminous basalt magmas into the lower crust (Huppert and Sparks, 1988).
- (2) With increased convergence rate, more fertile felsic materials from subducted materials would be added to the mantle wedge as melt or aqueous solution. This process would also increase the production of the subduction-related granitic magmas.

The above data suggest that model (2) is more likely than (1) for the following reasons: (a) The variations of  $\delta^{18}$ O and initial <sup>143</sup>Nd/<sup>144</sup>Nd combined with initial <sup>87</sup>Sr/<sup>86</sup>Sr (figs. 12 and 13) suggest source contamination resulting from melting of subducted materials in the mantle source region. This result supports model (2). (b) The main oxidizing agent required to form magnetite-series granitic magmas is SO<sub>2</sub> derived from the subducted oceanic crust (Takagi and Tsukimura, 1997). Even if granitic magmas are derived from the melting of the lower crust induced by injection of subductionrelated basaltic magmas containing SO<sub>2</sub>, the granitic magmas show little inheritance of  $SO_2$  from the basaltic magmas. This may be because the  $SO_2$  would be dispersed into the lower crust as a volatile component and/or fixed within the lower crust as magnetite and sulfides by reduction of ferrous ion in the lower crustal materials. Similarly, it would be difficult for granitic magmas to inherit reducing agents, such as  $CH_4$  and CO gases, from the basaltic magmas. To bring the gas phases from subducted materials to granitic magmas effectively, their direct ascent as melt solution without an inheritance process in the lower crust is more likely; these relationships also support model (2).

The positive correlation between the granite activities and the convergence rate (fig. 4) suggests the steady production of granitic magmas in proportion to the amount of subducted materials; it is inconsistent with granite genesis by injection of basaltic magmas into the lower crust or slab melting due to the episodic subduction of the Kula-Pacific ridge (for example, Nakajima and others, 1990; Maruyama, 1997; Kinoshita, 1999, 2002). Although it is likely that the subduction of the Kula-Pacific ridge beneath the Japan Arc took place, this ridge subduction may not have contributed to the formation of granitic magmas due to the cessation of the ridge spreading (for example, Byrne and DiTullio, 1992).

## The Increase in Sediment Subduction in the Late Cretaceous

The temporal variation of initial <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd in Japanese granitic rocks may be due to the variation of assimilation-rate of fertile sedimentary rocks by granitic magmas. In subduction zones, sedimentary rocks that can be assimilated into magmas include not only crustal sediments but also subducted sediments: for example, on the basis of the actual cross-section at Japan 40°N, Shreve and Cloos (1986) estimated that approximately 58 percent of the total sediment supply at the trench is subducted to the volcanic arc. The main sediments assimilated into granitic magmas in Japan are inferred to be subducted sediments for the following reasons; (1) Significant variations of initial  ${}^{87}$ Sr/ ${}^{86}$ Sr and  ${}^{143}$ Nd/ ${}^{144}$ Nd occurred simultaneously across the entire southwest Japan Arc during the period from 150 to 50 Ma. The phenomenon is difficult to interpret in terms of crustal assimilation. In contrast, assuming that the contaminant is subducted sediments, the variation of initial <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/ <sup>144</sup>Nd can be explained by regional tectonic movements and surfacial environmental conditions. (2) Figures 12 and 13 suggest that the Sr, Nd, and O isotopic variations of the Japanese granitic rocks are due mainly to source contamination processes. As mentioned above, the  ${}^{87}$ Sr/ ${}^{86}$ Sr,  ${}^{143}$ Nd/ ${}^{144}$ Nd, and  $\delta^{18}$ O of the assumed major assimilants at 85 Ma are 0.7085, 0.5122, and +20, respectively. The assimilants are not equivalent to the basaltic oceanic crust and the Ryoke metamorphic rocks in terms of the Sr and Nd isotopic compositions, but are similar to the present bulk sediment columns subducting at the Japan-Kurile trench (Plank and Langmuir, 1998): that is, their <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd at 85 Ma are 0.7088 and 0.51226, respectively. Although oxygen isotopic compositions of the sediment columns have not been reported, they should be higher than +10 because the columns are mostly composed of siliceous ooze, clay, and chert. Even though the subducted sediments attending granite plutonism may not have been the same as the present subducting sediments, both would be similar in chemistry because the nature of the bulk sediment columns would be a reflection of the mean crustal chemistry of the eastern Eurasian continents. Thus, the assumed assimilants in figures 12 and 13 are likely to be subducted sediments.

### Origin of the Systematic Variation of Granite Series

The rise of the ilmenite-series/magnetite-series ratios in the Late Cretaceous could be due to the lowering of oxidation states of magma source regions resulting from a substantial increase in sediment subduction. The ilmenite-series/magnetite-series ratios are in direct proportion to the convergence rate of oceanic plates (fig. 7), suggesting that rapid subduction was closely linked with the reduction of granitic magmas. As mentioned above, redox states of granitic magmas are generally dependent on the sources of magmatic sulfur. Therefore, if rapid subduction changes sulfur sources from the subducted altered oceanic crust to sediments, it is consistent with the increases in sediment subduction.

### The Causes of the Increase in Sediment Subduction

On the basis of the above evidence, the positive correlation between initial  $^{87}$ Sr/ $^{86}$ Sr of granitic rocks and convergence rate (fig. 10) indicates that the amount of subducted sediment increased progressively with increasing convergence rate. In southwest Japan, the thick Early Cretaceous to Tertiary accretionary complex, the Shimanto belt, was formed trench-ward with a width of more than 100 kilometers and length of 900 kilometers. The great mass of the complex may indicate a large supply of sediments to the trench during the period. Two factors may explain the linkage of sediment subduction and convergence rate: (1) the acceleration of subduction erosion, and (2) the increase of terrigenous sediment supply to subduction zones.

Subducting plates are hypothesized to erode the bottom and anterior margins of accretionary complexes, and draw sediments into subduction zones (Hilde, 1983; von Huene and Scholl, 1993). By this mechanism, even if the supply of terrigenous sediments to trenches does not increase, progressive sediment subduction should be promoted with the acceleration of plate subduction. The degree of subduction erosion would also be controlled by the supply of sediments and the geometry of subduction zones (Shreve and Cloos, 1986).

Chemical weathering rates of continental surfaces are significantly affected by rainwater acidity and air temperature (Berner, 1995), and the greenhouse Cretaceous atmospheric conditions likely enhanced weathering rates and increased terrigenous sediment supply to trenches and subduction zones. The concentration of atmospheric  $CO_2$  is closely linked to the productivity of oceanic crust at ridges, and is therefore also linked to the global convergence rate of oceanic plates. Thus, increased supply of terrigenous sediment to granite source regions may show a positive correlation with the local convergence rate.

The regional uplift of the Eurasian continental margin due to the accretion of an oceanic plateau and seamount chain in the Early Cretaceous would have increased the terrigenous sediment supply to the subduction zone (Kimura, 1997). Although the uplift occurred coincidentally with subduction during this time, it may have played an important role in enhancing the continental surface erosion rate, at least in the Japan Arc.

### Simultaneity of Oceanic Anoxic Events with the Major Ilmenite-series Granite Activity

Organic materials and sulfides in subducted sediments play an important role in the formation of ilmenite-series granitic rocks by controlling the redox state in the source regions. Oceanic anoxic events (OAE's) preserve organic materials and sulfides in marine sediments (Hartnett and others, 1998). OAE's occurred during the unusual global warming in the Late Cretaceous, and it is widely accepted that OAE's are the main cause of the widespread deposition of carbon-rich pelagic black shales at this time (Jenkyns, 1980).

As shown in figure 6, the timing of the OAE's agrees well with the peaks in ilmenite-series plutonism. This relationship suggests that the widespread oceanic anoxic conditions that prevailed along the margins of the Eurasian continents in the Late Cretaceous led to the deposition of carbon- and sulfide-rich sediments, and that the subduction of these sediments supplied the extensive amounts of reduced source rocks needed to form ilmenite-series magmas. The OAE's, combined with the increase in sediment subduction, probably produced the high-initial <sup>87</sup>Sr/<sup>86</sup>Sr and reduced granitic magmas in the Japan Arc.

Since the Paleogene, the global plate production has declined (Larson, 1991), resulting in the lowering of atmospheric  $CO_2$  and global cooling. In addition, this change brought about a slowdown of global plate subduction, the cessation of OAE, and the decrease in sediment subduction. Consequently, the initial  ${}^{87}$ Sr/ ${}^{86}$ Sr of the granitic rocks was also significantly lowered and magnetite-series granite plutonism became dominant by the effect of subducting oxidized oceanic crust.

### CONCLUSIONS

Examination of granite plutonism in the Japan Arc indicated the following features: (1) granite plutonism and initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  isotope ratios show positive correlations with the convergence rate of oceanic plates; (2) the oxidation states of the granitic rocks have a negative correlation with the granite plutonism, initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ , and the plate convergence rate; and (3) initial  $^{143}\mathrm{Nd}/^{144}\mathrm{Nd}$  isotope ratios and  $\delta^{18}\mathrm{O}$  values combined with the initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  isotope ratios indicate that the temporal variation of initial  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  isotope ratios was mainly due to source contamination

in the Japan Arc

resulting from melting of subducted materials in the mantle source region. These relationships suggest that the rise of initial <sup>87</sup>Sr/<sup>86</sup>Sr and the drop of oxidation states of Late Cretaceous granitic rocks were the result of increased sediment subduction into subduction zones. Reducing agents in the subducted sediments, including carbonaceous material and sulfides brought about the reduction of magma source regions, canceling out the oxidative effects of subduction zones due to the high erosion and the increase of sediment supply to subduction zones due to the high erosion rates of continental crusts under greenhouse Cretaceous atmospheric conditions. Simultaneity of oceanic anoxic events and the major ilmenite-series granite activity also suggests that the active subduction of carbon- and sulfide-rich sediments accumulating in anoxic marine environments promoted the reduction of granitic magmas. Consequently, at least in the Japan Arc, the variations of plate convergence rate and the Earth's surface conditions controlled the redox states of granitic magmas, generating the magnetite-and ilmenite-series.

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	Radiometric	: ages, Sr,	Nd, at	ıd Os isotop	APPEND e compositi	IX A ons of	Japanese g	ranitic	rocks	nsed	in thi	s study				
Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IbN	(=)+	ŀ	Z	Rf &	00 <sup>18</sup> * (%e)	(∓) +		z	Rf
Kyushu	district and South-West Islands															
IIm	Kunisaki	143.3	9.1	Rb-Sr(W)	0.70442	7					1					
IIm	Yamanaka	164.0	14.7	Rb-Sr(W)	0.70452	20									,	
Ilm	Watada	101.6	7.2	Rb-Sr(W)	0.70544	7					1					
IIm	Nioki	116.9	7.3	Rb-Sr(W)	0.70521	15					1					
IIm	Kusano	100.6	3.7	Rb-Sr(W)	0.70649	4					-					
IIm	Haki	118.0	11.3	Rb-Sr(W)	0.70557	20					1					
IIm	Kikuchi	121.3	8.4	Rb-Sr(W)	0.70542	13					1	12.0	0.8		7	a
IIm	Tamana	116.8	12.7	Rb-Sr(W)	0.70530	20					1					
llm	Okueyama (Upper)	14.6	0.3	Rb-Sr(W)	0.70630	7					70					
Ilm	Okueyama (Lower)	13.8	0.3	Rb-Sr(W)	0.70552	1					70	10.1	0.4	1.0	4	Ą
IIm	Kunisaki	142.9	1.6	Rb-Sr(W)	0.70441	7					71					
llm	Yonama	69.8	8.0	Rb-Sr(W)	0.70462	70					5					
IIm	Kanemi	61.0	6.7	Rb-Sr(W)	0.70639	38					2					
Ilm	Todoroki	69.3	2.6	Rb-Sr(W)	0.70987	12					5					
IIm	Chayama	40.7	14.5	Rb-Sr(W)	0.70586	36					5					
Mag	Higo	229.4	26.3	Rb-Sr(W)	0.70678	28					3					
Mag	Itoshima	116.0	17.0	Rb-Sr(W)	0.70506	12	0.51252				e				-	
Ilm	Sawara	114.0	11.0	Rb-Sr(W)	0.70516	23	0.51254			-	<i>ж</i>	10.4	0.0		7	es I
Mag	Fukuoka	108.0	16.0	Rb-Sr(W)	0.70517	25	0.51239	4	8	4	33					
Mag	Mitsuse	95.5	11.0	Rb-Sr(W)	0.70635	20	0.51232	9	10	4	e					
IIm	Saga	87.9	18.2	Rb-Sr(W)	0.70526	27	0.51254	5		5	<i>е</i>	10.7	0.6	0.4	5	а
Mag	Shiroishino	121.0	14.0	Rb-Sr(W)	0.70493	7					72					
IIm	Omoto	39.3	2.0	Rb-Sr(M)	0.70478	16					5					
Mag	Miyanohara	210.8	0.8	Nd-Sm(W)			0.51251	1			72	8.6				а
Mag	Fukae	115.0		Geol			0.51252	5		7	3				-	
llm	Okueyama	13.8		Geol	0.70537		0.51228			_	4					
IIm (S)	) Tanegashima	16.2		Geol	0.70869*		0.51232	-		6	4					
llm	Taishu	15.0		Geol	0.70643*		0.51244	11		7	28					
IIm (S)	) Yakushima	16.2	0.6	SHRIMP								12.0	0.2		7	٩
IIm	Takakumayama	16.0	2.0	K-Ar								11.6	0.3	0.2	3	q

Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IPN	(∓) +		z	Rf	$\delta O^{18*}$ (%)	(Ŧ) +	1	z	Rf
IIm (S)	) Ichifusayama	14.0	1.0	K-Ar								12.2	0.7	0.8	3	Ą
Ilm	Shibisan	15.0	4.0	K-Ar								10.2	0.1	0.2	e	q
IIm (S)	Kaseda	14.1	0.7	K-Ar								10.1			-	q
IIm (S)	Osuzu	13.0	2.0	K-Ar							_	13.1	0.5	0.7	3	þ
Chugok	u and Shikoku districts															
Ilm	Kurami-Nagisen	78.1	0.6	Rb-Sr(W)	0.70857	10	0.51232	S	4	4	73, 5					
IIm	Mochigase	79.2	0.4	Rb-Sr(W)	0.70650	б					73					
IIm	Chizu-Koshihata	80.7	0.6	Rb-Sr(W)	0.70596	2	0.51234	14		2	73,5					
IIm	Okayama	84.0	3.7	Rb-Sr(W)	0.70745	22	0.51229	ŝ	9	6	6,5	8.6	1.4	0.5	4	a B
Ilm	Kurashiki	90.5	I	Rb-Sr(W)	0.70770	I					7					
Ilm	Ukan	92.0	6.5	Rb-Sr(W)	0.70696	14					×					
IIm	Nishikawa-Kaito	84.4	6.3	Rb-Sr(W)	0.70568	ю					6					
IIm	Mikawachi	80.4	2.8	Rb-Sr(W)	0.70595	22					6					
IIm	Hiroshima (Eastern)	92.8	4.0	Rb-Sr(W)	0.70627	20					10					
Ilm	Hiroshima (Eastern)	90.5	5.1	Rb-Sr(W)	0.70631	15					10					
Ilm	Hiroshima (Eastern)	88.2	15.3	Rb-Sr(W)	0.70678	155					10					
IIm	Oasa	72.9	2.8	Rb-Sr(W)	0.70733	64					11					
Ilm	Obatake-Gamano	93.6	12.6	Rb-Sr(W)	0.70660	40					74					
Ilm	Yanai (Yonger)	88.6	1.5	Rb-Sr(W)	0.70749	-					74					
IIm	Hiroshima (Western)	92.7	2.5	Rb-Sr(W)	0.70662	20					12	9.4	1.4	2.1	9	a
llm	Yanai	95.1	4.6	Rb-Sr(W)	0.70740	80					12					
IIm	Murotsu	94.1	4.2	Rb-Sr(W)	0.70750	60					12					
IIm	Uda	92.9	4.4	Rb-Sr(W)	0.70510	150					13					
Ilm	Izuha	99.2	4.7	Rb-Sr(W)	0.70616	11					75					
IIm	Kibe (Hbl-Bt)	91.8	1.6	Rb-Sr(W)	0.70727	12					75					
IIm	Kibe (Bt)	91.6	4.0	Rb-Sr(W)	0.70734	112					75					
Ilm	Shodoshima	82.1	3.0	Rb-Sr(W)	0.70731	6	0.51230	б		7	6,5	10.2	0.5	1.0	e	в
IIm	Eastern Sanuki	93.4	28.1	Rb-Sr(W)	0.70745	LL	0.51231			1	6,5	10.5	2.0	1.9	9	es
IIm	Takanawa Peninsula	93.1	2.9	Rb-Sr(W)	0.70769	11					9	10.2	1.1	1.8	ŝ	ea
IIm	Shiratori	93.0	28.0	Rb-Sr(W)	0.70752	22					14					
IIm	Tsushigawa	83.2	1.7	Rb-Sr(W)	0.70803	10	0.51224	7	7	s	15					

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Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IbN	+(±)+	1	z	Rf	δO <sup>18</sup> * (‰) + (	( <del>+</del>		N N	÷.
llm	Nojima	95.0	15.0	Rb-Sr(W)	0.70741	33	0.51223	-	-	5	15					
IIm	Sumoto	0.06	11.0	Rb-Sr(W)	0.70734	17	0.51224	2	7	5	15					
Ilm	Tozanji	83.8	4.9	Rb-Sr(W)	0.70749	22	0.51225	2	г	4	15					
Ilm	Senzan	84.1	9.0	Rb-Sr(W)	0.70791	18	0.51223	2	-	S	15					
Ilm	Kagaribayama	75.5	5.8	Rb-Sr(W)	0.70794	34	0.51226	2	0	S	15					
Ilm	Iwaya	76.4	3.9	Rb-Sr(W)	0.70774	18	0.51227		1	S	15					
IIm (S)	) Okinoshima (Moshima)	16.0	2.0	Rb-Sr(W)	0.70740	18	0.51235	1	1	5	16	11.7			۹ ۱	<u> </u>
Ilm	Ishizuchi	14.0	i	Rb-Sr(W)	0.70676	I					76					
Ilm	Abu	85.0	3.1	Rb-Sr(W)	0.70526	23	0.51254	2	7	5	17					
Ilm	Omishima	84.3	3.0	Rb-Sr(W)	0.70583	47					- <i>LL</i>					
Ilm	Togouchi	85.3	3.6	Rb-Sr(W)	0.70635	10					78					
Ilm	Tateiwayama	77.3	2.6	Rb-Sr(W)	0.70653	13					79					
Mag	Ogamo (Coarse)	60.4	0.2	Rb-Sr(W)	0.70661	1					18	8.3 2.	9	6.	3	~
Mag	Ogamo (Medium)	61.0	0.3	Rb-Sr(W)	0.70558	7					18					
Mag	Ningyotoge	60.3	0.3	Rb-Sr(W)	0.70554	3					18	9.9 0.	2		2 a	
Mag	Neu	65.0	0.3	Rb-Sr(W)	0.70475	1	0.51246	1		2	19, 5	7.0 0.	1	•	2 a	-
Mag	Tottori	64.8	2.0	Rb-Sr(W)	0.70550	40	0.51258	-		5	20, 5	6.8 2.	0	0	3 9	Ŧ
Mag	Asagaridani	72.7	7.4	Rb-Sr(W)	0.70557	10					79, 5					
Mag	Asagaridani	74.5	5.2	Rb-Sr(W)	0.70553	11	0.51249	З		5	s					
Mag	Uchiidani	70.0	11.0	Rb-Sr(W)	0.70550	110	0.51246			1	S					
Mag	Hobutsusan	72.3	2.8	Rb-Sr(W)	0.70681	I	0.51244	1		7	21,5					
Mag	Kamochi	127.5	29.0	Rb-Sr(W)	0.70487	1					79					
Mag	Onbara	58.0	1.5	Rb-Sr(W)	0.70617	19					11					
Mag	Asuna	28.9	5.8	Rb-Sr(W)	0.70489	60					11					
Mag	Asuna	40.4	2.9	Rb-Sr(W)	0.70730	40					11					
Mag	Asuna	44.3	6.1	Rb-Sr(W)	0.70449	12					11	6.9			l a	_
Mag	Kawanobori	84.5	1.0	Rb-Sr(W)	0.70730	40					13					
Mag	Okutsu	69.1	0.3	Rb-Sr(W)	0.70574	1					18	9.3			la	
Mag	Namariyama	36.0	14.0	Rb-Sr(W)	0.70532	17					18					
Mag	Yubara South	85.2	1.7	Rb-Sr(W)	0.70535	1	0.51252			1	8, 29					
Mag	Yubarako	38.9	9.6	Rb-Sr(W)	0.70507	22	0.51263	2	5	3	5					

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Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IbN	(Ŧ) +		z	Rf	δO <sup>18</sup> * (%ο)	(Ŧ) +		z	Rf
Mag	Kayo	81.2	5.5	Rb-Sr(W)	0.70684	10					8					
Mag	Shikijiki	80.5	3.9	Rb-Sr(W)	0.70590	35					11					
Mag	Hobenzan	102.0	4.0	Rb-Sr(W)	0.70570	20					7	7.6			-	g
Mag	Okinoyama	81.5	11.8	Rb-Sr(W)	0.70620	40					7	8.0	0.5		6	t3
Mag	Kake (Rapakivi)	84.4	8.8	Rb-Sr(W)	0.70639	47					80					
Mag	Ishidogadake	86.5	5.9	Rb-Sr(W)	0.70572	35	0.51252	S	4	10	23					
Mag	Daito	60.8	4.0	Rb-Sr(W)	0.70499	5					84	7.1	1.6	1.0	S	es
Ilm	Aji	82.9	8.0	Rb-Sr(W)	0.70773	7	0.51233	4	ю	9	33					
IIm	Abu	T.9.T	5.3	Rb-Sr(M)	0.70521	17					17					
IIm	Omishima (tonalite)	82.9	1.5	Rb-Sr(M)	0.70581	4					<i>LL</i>					
IIm	Omishima (granite)	82.7	1.0	Rb-Sr(M)	0.70595	6					77					
Mag	Miyazu	61.9	0.9	Rb-Sr(M)	0.70725	З					25	8.4			-	a
Mag	Miyazu	60.4	0.9	Rb-Sr(M)	0.70769	4					25					
IIm	Yatagawa	62.6	1.6	Rb-Sr(M)	0.70793	e					25					
IIm	Rokko	72.1	1.5	Rb-Sr(M)	0.70740	10					25					
IIm	Rokko	71.9	1.5	Rb-Sr(M)	0.70840	20					25					
IIm	Rokko	71.2	1.5	Rb-Sr(M)	0.70760	10					25					
IIm	Awajishima	80.9	1.1	Rb-Sr(M)	0.70766	4					25					
Πm	Awajishima	80.4	2.0	Rb-Sr(M)	0.70796	ŝ					25					
IIm	Kitaobatake	6.66	4.0	Rb-Sr(WM)	0.70790	40					12	13.2			_	J
IIm	Gamano	89.4	4.0	Rb-Sr(WM)	0.70730	50					12	12.5			_	<del>ن</del>
IIm	Towa	93.5	4.3	Rb-Sr(WM)	0.70790	50					12	11.1			-	<u>с</u>
IIm	Habu	96.9	0.1	Rb-Sr(WM)	0.70607	1	0.51236	ę	ŝ	2	32	11.8	0.2		7	e
Mag	Mitsumori	80.4	3.3	Rb-Sr(WM)	0.70519	18					27					
IIm	Ikuridani	83.4	4.4	Rb-Sr(WM)	0.70775	31					27					
Mag	Okutsu	63.9	0.2	Rb-Sr(M)	0.70574	1					18					
IIm (S)	) Okinoshima (Tanijiri)	16.0	2.0	Geol	0.70913*		0.51232	æ	ŝ	9	16	8.9	0.1	0.3	m	٩
IIm (S)	) Kashiwajima	16.0	2.0	Geol							16	12.1	0.3		7	q
IIm (S)	) Takatsukiyama	13.9	0.2	K-Ar	0.70747*		0.51235	S	4	Ś	30	10.8	1.0	1.1	4	
Mag	Yamasa	54.1	2.7	K-Ar							31	7.0	0.1		7	a
IIm	Komaki	65.7	1.7	Rb-Sr(W)	0.70624	7					67	8.4	0.6	0.9		a

in the Japan Arc

191

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Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IbN	(Ŧ) +		z	Rf	δO <sup>18</sup> * (%o) +	+ (Ŧ)		z	Rf
Ilm	Shido	74.0	0.3	Rb-Sr(WM)	-						24					
IIm	Shirotori	87.1	0.6	Rb-Sr(WM)	ł						24					
IIm	Mannari (a part of Okayama)	84.0	3.7	Rb-Sr(W)	ı						6	8.3			1	c
Kinki al	nd Chubu districts															
IIm	Kyuanji	161.0	17.9	Rb-Sr(W)	0.70727	7					81					
Ilm	Fukihata	121.4	24.6	Rb-Sr(W)	0.70753	20					81					
llm	Katsuma	121.0	26.0	Rb-Sr(W)	0.70754	11	0.51219	б	-	10	37					
Ilm	Otagiri (North)	71.1	6.8	Rb-Sr(W)	0.71243	31	0.51186	ę	S	7	34					
Ilm	Otagiri (Central)	0.66	10.9	Rb-Sr(W)	0.71026	17	0.51201	9	9	10	34					
Ilm	Otagiri (South)	92.7	13.8	Rb-Sr(W)	0.70910	56	0.51213	ę	e	9	34					
Ilm	Ibaraki	81.5	0.7	Rb-Sr(W)	0.70769	б	0.51226	6	2	9	36					
IIm	Asakawazawa	148.8	11.8	Rb-Sr(W)	0.70605	20					38					
II	Aikawa	72.4	14.1	Rb-Sr(W)	0.70711	20					38					
IIm	Toki	72.3	3.9	Rb-Sr(W)	0.71060	10					7	9.5	0.3	0.3	ŝ	es
Ilm (S)	) Busetsu	82.5	3.9	Rb-Sr(W)	0.70960	20					7	11.5	1.0	1.0	S	ся
IIm	Otani	98.9	4.2	Rb-Sr(W)	0.71010	30					7	11.9	0.1	0.2	ŝ	a
IIm	Yagyu	74.6	10.9	Rb-Sr(W)	0.70938	16					39					
IIm	Takijiri	78.3	3.0	Rb-Sr(W)	0.70764	14					39					••••••
Ilm	Katsuragi	85.1	18.3	Rb-Sr(W)	0.70728	9					39					
IIm	Minamikouchi	72.3	2.4	Rb-Sr(W)	0.70895	19					39					
llm	Hira	78.2	3.9	Rb-Sr(W)	0.70914	38					40					
Ilm	Tanakami	79.5	0.6	Rb-Sr(W)	0.70951	37					40					
IIm	Suzuka (Older)	94.5	1.4	Rb-Sr(W)	0.70989	7					40					
llm	Suzuka (Younger)	7.9.7	3.5	Rb-Sr(W)	0.70984	42					40					
Ilm	Kaizukiyama	96.4	4.8	Rb-Sr(W)	0.70687	13					40	·				
Mag	Nyu	69.2	7.9	Rb-Sr(W)	0.70912	26					82					
Mag	Shirakawa	65.6	1.8	Rb-Sr(W)	0.71000	20					7	7.4	0.7	1.5	7	a
IIm	Kojaku	57.4	5.1	Rb-Sr(W)	0.71179	171					36					
IIm	Kabuto	79.2	10.2	Rb-Sr(W)	0.70831	21	0.51225	1	-	4	36					
IIm	Takato	85.2	6.6	Rb-Sr(W)	0.70774	17	0.51223	5	9	23	35					
Ilm	Inutagiri	67.3	1.8	Rb-Sr(WM)	0.71247	I					26					

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Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IPN	(∓) +	Z ,	Rf	δ0 <sup>18</sup> * (%ο)	( <del>=</del> ) +		z	Rf
Ilm	Otagiri	54.7	0.1	Rb-Sr(WM)	0.71052	2				26					
Ilm	Oshima	79.3	0.9	Rb-Sr(M)	0.70719	4				38					
Ilm	Asakawazawa	62.4	3.6	Rb-Sr(M)	0.70740	11				38					
Ilm	Aikawa	63.8	9.1	Rb-Sr(M)	0.70803	б				38					
IIm	Kaizukiyama	94.1	5.0	Rb-Sr(WM)	0.70740	24				40					• •,• •,
IIm	Nakatagiri	54.6	0.4	Rb-Sr(WM)	0.71073	×				26					
Πm	Katsuma	62.7	0.1	Rb-Sr(WM)	0.70778	7				37					
IIm	Ichida	58.9	0.5	Rb-Sr(WM)	I					24					
Πm	Takato	59.3	0.3	Rb-Sr(WM)	I					24					
Ilm	Hiji	59.1	0.1	Rb-Sr(WM)	I					24					
Ilm	Kisokoma	64.2	0.2	Rb-Sr(WM)	I					24					
llm	Kise	61.3	0.5	Rb-Sr(WM)	I					24					
IIm	Hissori	64.5	0.1	Rb-Sr(WM)	I					24				-	
Пm	Ogawa	67.5	0.4	Rb-Sr(WM)	I					24					
IIm	Ikuta	60.5	0.2	Rb-Sr(WM)	I					24					
Ilm	Kiyosaki	63.5	0.3	Rb-Sr(WM)	I					24					
Ilm	Mitsuhasi	59.3	0.3	Rb-Sr(WM)	I					24				•	
Ilm	Nagawa	69.1	3.5	K-Ar						99					
Ilm	Naegi	67.2	3.2	CHIME						41	7.8	0.3		6	сı
IIm	Inagawa	81.9	1.4	CHIME						41	10.6	1.5	1.4	18	a a
IIm	Kamihara	94.9	4.9	CHIME						41	9.1	0.0		7	сı
IIm	Mitsuhashi	83.8	1.3	CHIME						41	10.5	0.5	0.7	n	es
IIm	Minakata	94.9	4.9	CHIME						41	10.5			-	ა
Mag	Takidani	1.93	0.16	K-Ar						62					
Hida ar	nd Kanto districts														
IIm	Tsukuba	61.6	4.3	Rb-Sr(W)	0.71291	16	0.51203	31	17 9	42,4	5				
IIm	Yamanoo pegmatite	59.9	4.0	Rb-Sr(W)	0.71138	88				42					
Mag	Utsubo	183.2	9.8	Rb-Sr(W)	0.70529	5	0.51247	5	6 5	43, 4	6				
Mag	Kegachidake	185.8	7.5	Rb-Sr(W)	0.70487	14				69					
Mag	Komagatake	192.6	21.4	Rb-Sr(W)	0.70441	9	0.51280	e	7	46, 6	6				
Mag	Tsurugidake	69.3	5.1	Rb-Sr(W)	0.70800	31				44					

APPENDIX A

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Rf																						р									
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( <del>+</del> ) +												ļ										0.4									
· (%)																															
δ0 <sup>18</sup> *																															
Rf	68	68	42	42	42	45	46	68,46	68,46	68	29	63		47	48	48	49	83	50	50	51	51	51	51	51	51	51	85	85	52	53
z						7	ŝ	2	e		-			4																	
						11	27		4					7																	
(Ŧ) +						11	14	11	9					3																	
*IbV						51220	51216	51247	51233		51309			51250																	
						0.5	0.4	0.5	0.5		0.4			0.4																	
2σ	7	23	23	4	×	I		8	ŝ	1	ł			15	10	6	21	13	11	26	35	٢	15	17	6	Ś	16	24	6	16	9
SrI	0474	0485	6960	1320	1063	)817*		0499	0681	0617	0365			0518	0553	0645	0521	0493	0518	0489	0463	0392	0363	0415	0419	0445	0435	0420	0427	0390	0355
	0.7	0.7	0.7	0.7	0.7	0.7(		0.7	0.7	0.7	0.7			0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7
poq	r(W)	r(W)	r(M)	r(M)	r(M)	-Ar	Ar	r(M)	r(M)	r(M)	Ar	Ar		r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)	r(W)
Met	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Ar	K-	Rb-S	Rb-S	Rb-S	K-	K-		Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S	Rb-S
error	5.7	9.1	2.3	0.4	2.7	0.2	1	3.8	5.5	2.7	I	0.27		16.0	3.5	2.4	40.2	17.0	17.0	18.0	29.8	11.9	19.2	26.5	17.9	13.8	16.1	18.0	9.0	8.0	25.0
Age Ma)	01.2	84.1	58.3	50.2	59.5	92.0	66.0	82.6	73.4	10.9	7.0	3.91		0.00	06.4	11.6	14.7	11.7	20.0	19.0	07.4	28.2	56.7	56.8	23.6	06.8	31.6	42.0	38.0	18.0	35.0
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	himon	unatsu	amanc	sukub	sukub	shio (;	lodatsı	kumay	atsuo	layatsu	anzaw	anigav	strict	shikaw	<b>1</b> arum(	farum	abito	inomu	fiyamc	fiyamc	hura	fiyako	akaino	urihas	ono	enmay	abashi	urihas	urihas	tanabé	lashika
eries	Mag S	Mag F	Ilm Y	Ilm T	Ilm T	Ilm A	Mag H	Mag C	Mag Y	Mag H	Mag T	Mag T	hoku dis	llm Is	llm N	Ilm N	Ilm T	llm Jı	llm N	llm N	Mag C	Mag N	Mag S	Mag K	Mag T	Mag S	Mag T	Mag K	Mag K	Mag C	Mag H
Š	ſ.						<u> </u>	-	<u> </u>			-	To																		-

APPENDIX A

Series	Pluton	Age (Ma)	error	Method	SrI	2σ	*IPN	(∓) +		z	Rf	$\delta O^{18*}$ (%) + (±) -	z	Rf
Ilm	Iwafune	89.5	1.9	Rb-Sr(W)	0.70496	20					54			
IIm	Taiheizan	<i>T.</i> 66	11.9	Rb-Sr(W)	0.70541	16					55			
IIm	Gozu	93.6	3.8	Rb-Sr(W)	0.70537	15	0.51242	2	7	5	22, 56			
IIm	Sekikawa (Oguni)	96.8	21.5	Rb-Sr(W)	0.70630	81	0.51222	e	-	5	56			
IIm	Sekikawa (Yuzawa)	100.2	1.5	Rb-Sr(W)	0.70492	5	0.51231	2	S	5	56			
Ilm	Sekikawa (Onnagawa)	71.4	6.3	Rb-Sr(W)	0.70736	23	0.51230	9	10	12	56			
Ilm	Miomote dam	91.4	6.1	Rb-Sr(W)	0.70564	7	0.51239	13	9	×	56			
Ilm	Kogawa	73.8	2.0	Rb-Sr(W)	0.70779	45	0.51230	S	4	5	56			
Ilm	Takane	43.8	5.6	Rb-Sr(W)	0.70935	6	0.51231	7	٢	4	56			
Mag	Daiyama	23.8	4.4	Rb-Sr(W)	0.70528	48	0.51264	1	ŝ	12	57			
Ilm	Komuro	94.5	0.5	Rb-Sr(M)	0.70535	5	0.51254	2	ŝ	4	49			
Ilm	Myojinishi	90.4	0.6	Rb-Sr(M)	0.70528	9	0.51255	1		б	49			
Ilm	Iritabyuto	94.5	0.5	Geol	0.70521*	I	0.51249	ŝ	æ	ŝ	49			
Ilm	Samekawa	112.0	4.0	K-Ar							63	10.6	-	c
IIm	Yoshimagawa	118.0	5.0	K-Ar							63			
IIm	Ronden	103.0	5.0	K-Ar							63			
IIm	Hanawa	119.0	5.0	K-Ar							63			
IIm	Irishiken	98.9	3.1	K-Ar							63			
IIm	Kamikimita	109.0	4.0	K-Ar							63			
IIm	Miharu (Nagaya)	112.0	4.0	K-Ar							65			
IIm	Miharu (Hatsumori)	94.8	3.0	K-Ar							65			
Mag	Tanohata	128.2	11.9	Geol	0.70377	1	0.51261			-	29			
Mag	Taro	107.4	29.8	Geol	0.70398	I	0.51262			1	29			
Mag	Otomo	135.0	25.0	Geol								8.1		ပ
Mag	Nissho	10.0	I	Geol	0.70439*	ł	0.51286	4	0	×	57			

APPENDIX A (continued)

Series	Pluton	Age (Ma)	ептог	Method	SrI	2σ	*IbN	- (∓) +	N N	Rf	ôO <sup>18</sup> * (%o) + (±) -	z	Rf
Hokkait	to district												
Ilm	Hidaka (tonalite)	51.2	3.6	Rb-Sr(W)	0.70452	4				58			
Ilm	Imakane	129.7	23.0	Rb-Sr(W)	0.70487	45				59			
Ilm	Kudo	133.7	26.2	Rb-Sr(W)	0.70500	45				59			
IIm	Okushirito	104.0	16.0	Rb-Sr(W)	0.70536	33				60			
Ilm	Nisshotoge	17.3	ŀ	Rb-Sr(W)	0.70460	I				7			
Ab	breviations: SrI = Initial $^{87}$ Sr/ $^{86}$ Sr	r isotope 1	ratio, Nd	I = Initial <sup>143</sup>	Nd/ <sup>144</sup> Nd is	otope ra	tio, $N = 1$	Number of	analysis	, Rf = I	References, Geol = Age	estima	tted by

APPENDIX A (continued) geology, Ilm = Ilmenite-series, Mag = Magnetité-series, (S) = S-type granites, (W) = Whole-rock isochron age, (M) = Mineral isochron age, (WM) = Whole-rock mineral isochron age, + = Upper deviation (last digits for NdI values), - = Lower deviation (ditto), \* = average value, - = no description. When a pluton has multiple age data, the oldest age is listed above.

196

# Appendix B

Data sources for numbered references referred to in Appendix A

Radi	ometric ages and Sr and Nd isotopic compo	sitions	· · · · · · · · · · · · · · · · · · ·
1	Osanai and others (1993)	35	Yuhara and Kagami (1999)
2	Kawano and Kagami (1993)	36	Tainosho and others (1999)
3	Owada and others (1999)	37	Yuhara and Kagami (1995)
4	Terakado and others (1988)	38	Ohtomo and others (1993)
5	Kagami and others (1992)	39	Morioka and others (2000)
6	Kagami and others. (1988)	40	Sawada and others (1994)
7	Shibata and Ishihara (1979a)	41	Suzuki and Adachi (1998)
8	Takagi and Kagami (1995)	42	Arakawa and Takahashi (1988)
9	Matsumoto and others (1994)	43	Arakawa (1988)
10	Nakajima and Shirahase (1987)	44	Tanaka and Kagami (1987)
11	Rezanov and others (1994)	45	Kawano and others (1999)
12	Shigeno and Yamaguchi (1976)	46	Arakawa and Shinmura (1995)
13	Seki (1978)	47	Shibata and Tanaka (1987)
14	Kagami and others (1995)	48	Shibata (1987)
15	Yuhara and others (1998)	49	Tanaka and others (1999)
16	Dai and others (1993)	50	Fujimaki and others (1991)
17	Yuge and others (1998)	51	Maruyama and others (1993)
18	Sudo and others (1988)	52	Wang and others (1994)
19	Iizumi and others (1984)	53	Fujimaki and others (1992)
20	Hattori and Shibata (1974)	54	Kagashima (1999)
21	Iizumi and Kagami (1987)	55	Maruyama and Yamamoto (1994)
22	Rezanov and others (1996)	56	Rezanov and others (1999)
23	Ikawa and others (1999)	57	Shimakura and others (1999)
24	Yuhara and others (2000)	58	Owada and others (1997)
25	Terakado and Nohda (1993)	59	Kawano and Kagami (1999)
26	Yuhara and Kagami (1996)	60	Kawano and others (1993)
27	Takagi and others (1989)	62	Harayama (1992)
28	Ishizaka and Ishikawa (1990)	63	Kawano and others (1992)
29	Terakado and Nakamura (1984)	64	Shibata and Uchiumi (1983)
30	Ishikawa and Kagami (1992)	65	Tomizuka and others (1991)
31	Kano and others (1994)	66	Nakano and others (1995)
32	Yuhara and others (1999)	67	Takagi and others (2003)
33	Yuhara and others (2003)	68	Tanaka (1992)
34	Yuhara (1994)	69	Arakawa (1990)
Oxy	gen isotope compositions		- Ne hole a factor -
a	Ishihara and Matsuhisa (2002)	с	Matsuhisa and others (1972)
b	Ishihara and Matsuhisa (1999)	d	Ishihara and others (1985)
Writt	en communications (domestic journals or a	bstracts of r	neeting written in Japanese)
70	Okamoto and Honma (1983)	78	Ishioka and Iizumi (1998)
71	Masao and others (1990)	79	Iizumi and others (1982)
72	Kamei and others (1997)	80	Imaoka and others (1998)
73	Honma (1986)	81	Fujii (1997)
74	Okano and Honma (1983)	82	Nakajima and others (1990b)
75	Wakisaka (1982)	83	Goto and others (1990)
76	Nozawa and Tainosho (1990)	84	MITI (1988)
77	Imaoka and others. (1997)	85	Kanisawa and others (1993)

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