

Carbon and oxygen isotopic composition of the carbonates from the Jacupiranga and Catalao I carbonatite complexes, Brazil

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Abstract: Carbon and oxygen isotope compositions were measured for carbonates from the Jacupiranga and Catalao I carbonatite complexes in Brazil. The $\delta^{13}\text{C}$ values of the Jacupiranga carbonates are uniform, ranging from -6.4 to -5.6‰ with the average of -6.07‰. Except for one sample, the $\delta^{18}\text{O}$ values of the carbonates are between 7.1 and 8.1‰, and the average value is 7.6‰. The isotopic compositions of the Jacupiranga carbonates represent the value of primary igneous carbonatite. The $\delta^{13}\text{C}$ values of dolomites are about 0.5‰ higher than those of calcites.

The $\delta^{13}\text{C}$ values of carbonates from the Catalao I complex range from -6.8 to -5.2‰ with the average of -5.83‰. Those values are similar to the values of the Jacupiranga carbonates. However, oxygen isotopic compositions of the Catalao I carbonates show a wide range of 8.4 to 22.3‰. Carbonates with the lowest $\delta^{18}\text{O}$ values in the complex are considered to represent the igneous stage. Carbonates with extremely high $\delta^{18}\text{O}$ values of about 22‰ are considered to have precipitated from low-temperature hydrothermal fluids. The group of intermediate $\delta^{18}\text{O}$ values indicates a variable degree of contamination by the $\delta^{18}\text{O}$ -rich hydrothermal carbonates. The contribution of secondary stage hydrothermal carbonates seems to be significant in the Catalao I complex as compared with the Jacupiranga complex. The development of a network structure in the Catalao I complex may have enhanced the circulation of the later stage hydrothermal fluids.

1. Introduction

Carbonatite is intimately associated with alkaline rocks such as nephelinite and ijolite. Initial $^{87}\text{Sr}/^{86}\text{Sr}$ values of carbonatite are low and they are similar to those of alkali basalt. It is inferred that carbonatite magma is derived from the upper mantle. Carbon isotopic ratios of carbonatites together with those of kimberlites and diamonds can be utilized to estimate the isotopic composition of deep-seated carbon.

Carbon and oxygen isotopic ratios of carbonatites have been reported by many research-

chers (TAYLOR *et al.*, 1967; CONWAY and TAYLOR, 1969; SUWA *et al.*, 1969; DEINES, 1970; DEINES and GOLD, 1973; SUWA *et al.*, 1975). TAYLOR *et al.* (1967) postulated that primary igneous carbonatite has relatively limited $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ranges. However, there is a slight isotopic difference between carbonatites from different localities (DEINES and GOLD, 1973). Stable isotopic compositions of the Brazilian carbonatites have scarcely been reported. This study aims at elucidating the carbon and oxygen isotopic features of the Jacupiranga and Catalao I carbonatite complexes in Brazil.

2. Geology of the Jacupiranga and Catalao I carbonatite complexes

Jacupiranga

Jacupiranga alkaline carbonatite complex is

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located 230 km southwest of Sao Paulo and is composed of peridotite, clinopyroxenite, jacupirangite, ijolite, fenite, nepheline syenite and carbonatite. It is oval shaped, and 65 km² in area. It intruded into the Precambrian mica schist and quartz diorite in the Early Cretaceous period (130 ± 5 Ma, AMARAL, 1978). The carbonatites occur near the center of the complex as two plugs, the northern and southern bodies. The total exposure of the carbonatites is about 0.4 km².

According to Melcher (1966), the peridotitic body was first emplaced in the northern half of the complex and then surrounded by a ring intrusion of clinopyroxenite. After that, the circular clinopyroxenite intruded into the southern half of the complex and differentiated into the crescent-shaped ijolite body. Finally, carbonatite intruded as small plugs into the central part of the southern clinopyroxenite body (MELCHER, 1966; GASPER and WYLLIE, 1983a; HIRANO *et al.*, 1990a).

HIRANO *et al.* (1990a) have classified the carbonatites into four rock facies as follows:

Calcite carbonatite (CC), magnetite-rich calcite carbonatite (CCmt), apatite-rich calcite carbonatite (CCap) and dolomite carbonatite (DC).

Calcite carbonatite is the major facies of both the northern and southern plugs. It consists mainly of calcite, magnetite, apatite and dolomite. Minor constituents are olivine, phlogopite, pyrrhotite, chalcopyrite and valleriite. The amount of mafic minerals are usually less than 10%. JC-11 and -46 are from this facies. Calcite carbonatite containing more than 20% of modal magnetite is called magnetite-rich calcite carbonatite in this paper.

Apatite-rich calcite carbonatite consists of medium-grained apatite and olivine with calcite, phlogopite and magnetite. A small amount of dolomite is also contained. JC-18 and -26 are from this facies.

Dolomite carbonatite occurs as small dykes in both carbonatite plugs. It consists essentially of dolomite with small amounts of apatite, phlogopite, magnetite and sulfide minerals. JC-27 and -44 are from this facies. The detailed petrographic descriptions of each rock facies

are given in HIRANO *et al.* (1990a). The Jacupiranga samples analyzed in this study are listed in Table 1 and their localities are shown in Fig. 1.

Catalao I

Catalao I carbonatite complex is located in the southeastern part of the Goias state, and it intruded into the Proterozoic metamorphic rocks. The age of the intrusion is 82.9 ± 4.2 Ma (HASUI and CORDANI, 1968). The complex has a circular shape and is about 6 km in diameter. The rocks of the complex are as follows (HIRANO *et al.*, 1990b);

(1) Ultramafics and their metasomatized rocks (ultramafics, phlogopite rock, metaphoscolite);

(2) Phoscolite;

(3) Carbonatite and carbonate rocks (dolomite carbonatite, magnetite-bearing dolomite carbonatite, dolomitic sinter and others).

Since the complex has been deeply weathered, the relationship between these rock types can only be observed in drill core samples.

The sequence of events leading to the formation of the various rock types is as follows (HIRANO *et al.*, 1990b):

(1) Ultramafic rocks were crystallized from a silicate magma. Some parts of the ultramafics were serpentinized during the cooling of the intrusion.

(2) Carbonatitic magmas intruded into the ultramafic rocks, forming the network veins and dykes of phoscolite and dolomite carbonatite. Metaphoscolite was also formed locally along the conduits.

(3) Crystallization of carbonate minerals (dolomite and magnetite) from hydrothermal fluids. Carbonate veinlets of the hydrothermal stage sporadically cut the early stage igneous members (including phoscolite and dolomite carbonatite).

The Catalao I samples analyzed in this study are listed in Table 1. All the samples were taken from a drill core, the site of which is shown in Fig. 2. CT-36, -38, -39A, -351 and -352 are the rocks from stage (2) mentioned above. CT-40 and -41 are representatives of stage (3).

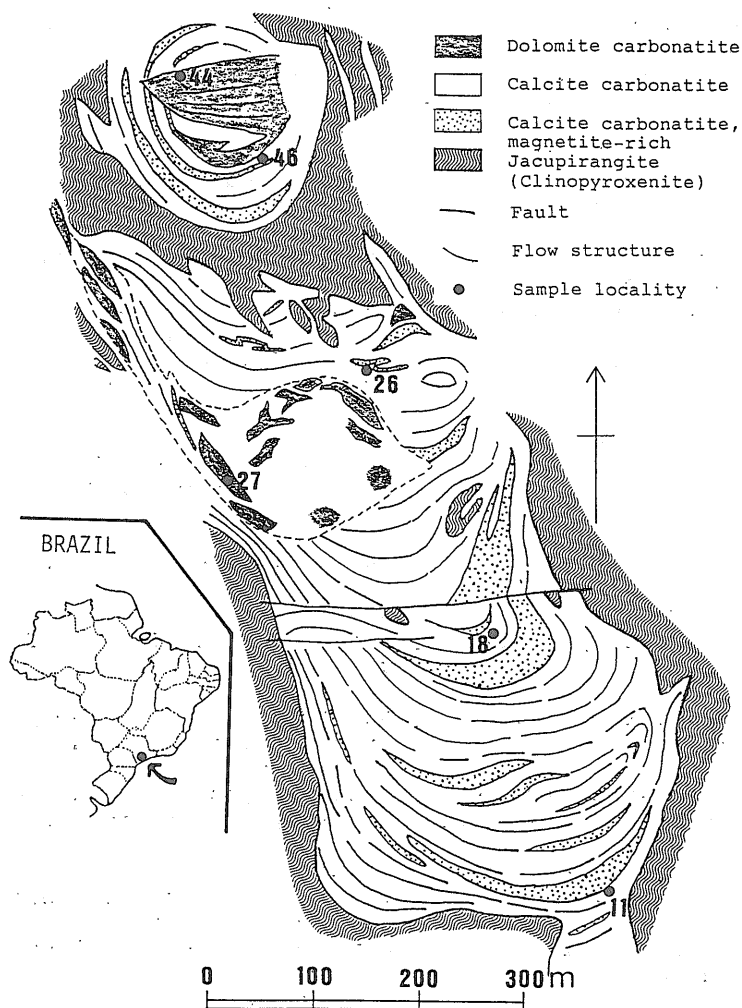


Fig. 1 Geological map (HIRANO *et al.*, 1990a) of the Jacupiranga carbonatite complex showing the sample localities.

3. Experiments

Carbon dioxide was liberated from carbonate by the reaction with 100% phosphoric acid at 25°C in a vacuum (MCCREA, 1950). Calcite-dolomite mixtures were analyzed using the double extraction technique developed by EPSTEIN *et al.* (1964). Carbon dioxide obtained was analyzed with a Varian Mat 250 mass spectrometer at Shinshu University. For oxygen, corrections were made for isotope fractionation between liberated carbon dioxide and original carbonate. Fractionation factors determined by

Sharma and Clayton (1965), $(\text{CO}_2\text{-cc}) = 1.01025$ and $(\text{CO}_2\text{-dol}) = 1.01109$, were used. The isotopic data are presented in terms of the δ notation relative to PDB for carbon, and SMOW for oxygen. The experimental errors are $\pm 0.1\%$ for both the ^{13}C and ^{18}O values.

Analytical results of the reference sample ($\text{CaCO}_3\text{-1}$, GSJ working standard) are presented in Table 2. The measured values agree well with the results by MORISHITA and MATSUHISA (1984).

Table 1 Petrographic descriptions of the samples studied from the Jacupiranga and Catalao I carbonatite complexes.

Rock type	Description
Jacupiranga	
JC-11 Olivine-bearing calcite carbonatite	76cc, 14ap, 8mt, 1ol, 1dol; medium grained, eaakly banded.
JC-18 Olivine-apatite-calcite carbonatite	37ol+clinohumite, 32ap, 21cc, 4mt, 4po, 2dol; coarse grained.
JC-26 Apatite-rich calcite carbonatite	82cc, 13dol, 4ap, 1mt; coarse grained.
JC-27 Dolomite carbonatite	69dol, 27ap, 2cc, 2phl; medium grained, weakly banded.
JC-44 Dolomite carbonatite	63dol. 35ap, 2mt: a very small amount of interstitial calcite occurs.
JC-46 Calcite carbonatite	81cc, 14dol, 3mt, 2sulfide; magnetite and sulfide concentration forms an irregular banding.
Catalao I	
CT-36 Nb-bearing phoscolite (277m)	40phl, 30mt, 20ap, 5dol, 5pyc; medium grained, dolomite occupies the interspaces of phl-mt-ap aggregates.
CT-38 Dolomite carbonatite (115m)	80dol, 10norsethite, 5py (fine grained), 3ap, 1phl, 1pyc; coarse grained.
CT-39A Dolomite carbonatite (402.5m)	90dol, 3mt, 1phl; medium grained, banded. Magnetite concentration forms a banding.
CT-40 Magnesite-bearing dolomite carbonatite (415m)	Magnesite, dol; fine-grained, possibly hydrothermal origin.
CT-41 Dolomitic sinter (Cocarde) (246.4m)	Dol, other mineals (not determined); aphanitic colloform texture develops.
CT-351 Metaphoscolite (292m)	80phl, clay mineral, ol (serpentinized), 10mt, 10dol; medium grained.
CT-352 Phoscolite (292m)	80ap, 6phi, 5dol, 4mt, 3pyc; medium grained.

Abbreviations: cc-calcite, dol-dolomite, mt-magnetite, ol-olivine, ap-apatite, po-pyrrhotite, py-pyrite, phl-phlogopite, pyc-pyrochlore. Numbers in description are volume percentages estimated petrographically. The nomenclature of carbonatites is after Streckeisen (1980). Numbers in parenthesis indicate the depth of the drill core samples (name of drillhole: 47E29N).

4. Analytical results and discussion

Six carbonate samples from the Jacupiranga complex and seven from the Catalao I complex were isotopically analyzed. The results are shown in Table 2 and Fig. 3.

Jacupiranga complex

The $\delta^{13}\text{C}$ values of the Jacupiranga carbonates are in the range of -6.4 to -5.6% with

the average of -6.07% ($\sigma_n=0.29$, $n=9$). The carbon isotopic compositions are very uniform. Except for one sample (JC-44 calcite), the $\delta^{18}\text{O}$ values of the carbonates are between 7.1 and 8.1% , and the average value is 7.6% ($\sigma_n=0.35$, $n=8$). JC-44 calcite seems to be exceptional and its $\delta^{18}\text{O}$ value is 2% higher than the average $\delta^{18}\text{O}$ value.

TAYLOR *et al.* (1967) postulated that the $\delta^{13}\text{C}$

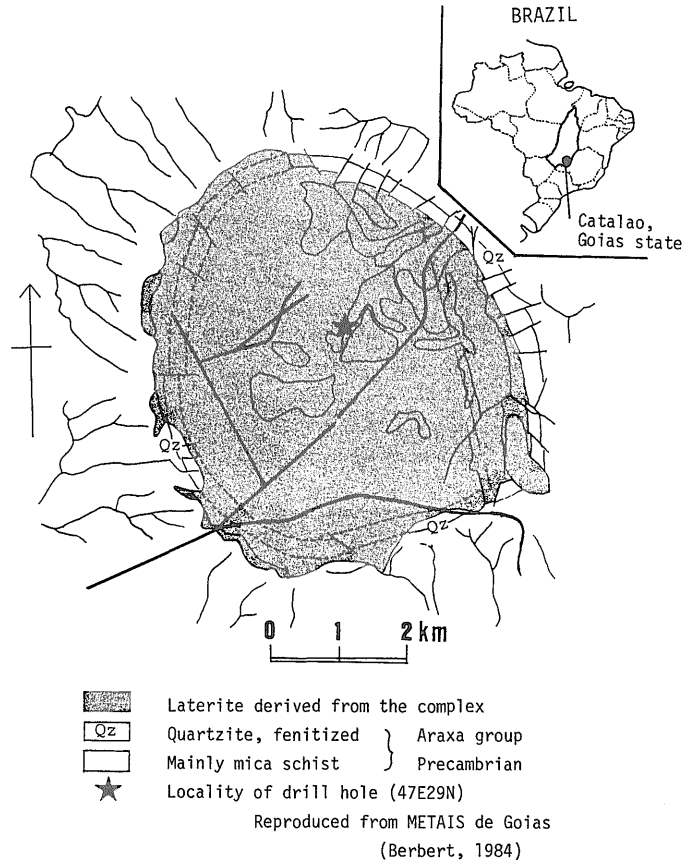


Fig. 2 Surface geological map of the Catalao I carbonatite complex showing the sample locality. Inner solid and dashed lines correspond to the outline of the complex.

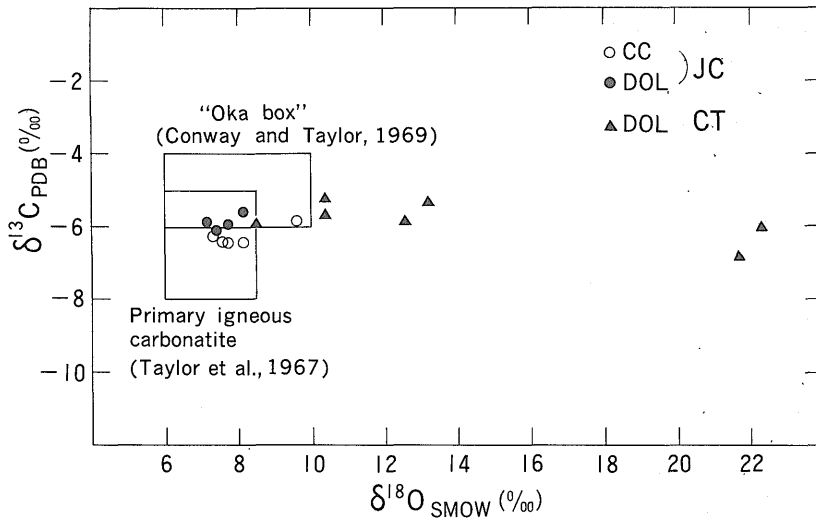


Fig. 3 Plot of $\delta^{18}\text{O}$ versus $\delta^{13}\text{C}$ for carbonates from the Jacupiranga and Catalao I carbonatite complexes.

Table 2 Isotopic composition of the carbonates from the Jacupiranga (JC-) and Catalao I (CT-) carbonatite complexes.

Sample	Calcite		Dolomite		Calcite: dolomite volume ratio
	$\delta^{13}\text{C}$	$\delta^{18}\text{O}$	$\delta^{13}\text{C}$	$\delta^{18}\text{O}$	
JC-11	-6.4	7.5			cc>dol
JC-18	-6.4	8.1			cc>dol
JC-26	-6.2	7.3	-5.8	7.1	cc>dol
JC-27			-5.9	7.7	dol
JC-44	-5.8	9.6	-6.1	7.3	dol>>cc
JC-46	-6.4	7.7	-5.6	8.1	cc>dol
CT-36			-5.3	13.2	dol
CT-38			-5.9	8.4	dol
CT-39A			-5.2	10.4	dol
CT-40			-6.0	22.3	dol
CT-41			-6.8	21.7	dol
CT-351			-5.8	12.6	dol
CT-352			-5.6	10.4	dol
CaCO ₃ -1	2.53	-15.00**			
	2.63	-14.97**			
CaCO ₃ -1*	2.59	-15.00**			

* MORISHITA and MATSUHISA (1984)

** $\delta^{18}\text{O}$ values are given in the PDB scale.

values of primary igneous carbonatite are in the range of -8.0 to -5.0‰ and the $\delta^{18}\text{O}$ values 6.0 to 8.5‰. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the Jacupiranga carbonates are remarkably uniform and the $\delta^{18}\text{O}$ values are the lowest among the carbonatites from all over the world (e.g. DEINES and GOLD, 1973). It is noteworthy that the isotopic data from the Jacupiranga complex are plotted in a small area within the primary igneous carbonatite field of TAYLOR *et al.* (1967). They are also plotted near the base of the "Oka box" which was defined by CONWAY and TAYLOR (1969). The Jacupiranga carbonatites occur as discordant igneous plugs in the clinopyroxenite as described before. The carbonatites are of the deep-seated plutonic type and the total amount of erosion to reach the present level has been estimated to be over 1500 m (MELCHER, 1966). It seems safe to say that there has been no effect or alteration from surface weathering. Therefore, it is considered that the isotopic compositions of the Jacupiranga carbonates represent the value for primary igneous carbonatites in southern Brazil.

The $\delta^{18}\text{O}$ value of JC-44 calcite is about 2‰

higher than that of the coexisting dolomite, whereas the $\delta^{13}\text{C}$ values are the same. The amount of calcite in the rock is very small and the calcites fill the grain boundaries of coarse-grained dolomites. Judging from the mode of occurrence, the enrichment of $\delta^{18}\text{O}$ in the calcite is interpreted as due to precipitation from the magmatic fluids of a later stage probably at lower temperature.

The $\delta^{13}\text{C}$ values of dolomite, on the whole, are higher than those of calcite. The average $\delta^{13}\text{C}$ value of dolomite is -5.9‰, whereas that of calcite (except for JC-44 calcite) is -6.4‰. The tendency is commonly recognized in carbonatites (DEINES, 1970), but the difference between calcite and dolomite observed in this study (0.5‰ on an average) is much smaller than the value described by DEINES (1970). As for a single rock sample which contains both calcite and dolomite, the dolomite has higher $\delta^{13}\text{C}$ value than the coexisting calcite, except for JC-44. If we assume that coexisting calcite and dolomite are isotopically in equilibrium, we can calculate isotopic equilibration temperatures using SHEPPARD and SCHWARZ's equation (1970). The results are as follows:

$$\text{JC-26: } \Delta^{13}\text{C}_{\text{dol-cc}} = 0.4 \quad T = 610^\circ\text{C.}$$

$$\text{JC-46: } \Delta^{13}\text{C}_{\text{dol-cc}} = 0.8 \quad T = 260^\circ\text{C.}$$

The former value is consistent approximately with equilibrium temperatures of 595-570°C estimated by GASPARD and WYLLIE (1983b) for coexisting ilmenite and magnetite from the reaction rock between jacupirangite and intruding carbonatite of the complex.

Catalao I complex

The $\delta^{13}\text{C}$ values of carbonates from the Catalao I complex range from -6.8 to -5.2‰ with the average of -5.80‰ ($\sigma_n = 0.49$, $n = 7$). Those values are almost the same as the $\delta^{13}\text{C}$ values of the Jacupiranga carbonatite. The $\delta^{18}\text{O}$ values of the Catalao I carbonates, however, show a considerable range of 8.4 to 22.3‰.

The oxygen isotope variation of the Catalao I carbonates seems to correspond to their diverse mode of occurrence. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the CT-38 dolomite, which exhibits an igneous texture, are close to the compositional range of the Jacupiranga carbonates. Therefore, it is

presumed that the CT-38 domomite represents a primary isotopic signature of the Catalao I carbonatites.

The samples CT-40 (magnesite-bearing dolomite carbonatite) and CT-41 (dolomitic sintér) occur as veinlets filling fissures of the ultramafics and are mainly composed of cryptocrystalline carbonate minerals. CT-41 shows a colloform texture. The mode of occurrence and textural characteristics suggest that they have precipitated from low-temperature hydrothermal fluids (HIRANO *et al.*, 1990b). The $\delta^{18}\text{O}$ values of CT-40 and -41 are remarkably high, while the $\delta^{13}\text{C}$ values are not different from the other samples from the Catalao I complex. From the above observations, we may deduce the following genetic process for these carbonatites. Meteoric water or hydrothermal fluid which was enriched in ^{18}O permeated into the dolomite carbonatite bodies, and consequently, a part of the dolomite was dissolved into the fluid. After that, the carbon in carbon-bearing fluid re-precipitated the secondary dolomite along conduits at low temperatures. The carbon isotopic ratio of the system was buffered in the predominating primary carbonates. Thus, veins of ^{18}O -enriched cryptocrystalline dolomite, whose $\delta^{13}\text{C}$ values are almost identical to the primary dolomite carbonatite, were formed.

The carbonates of CT-36, -39A, -351 and -352, which have intermediate values of $\delta^{18}\text{O}$, occur interstitially and as "amygdulés" in the phoscolite, and their amounts in the rocks are small. They lie on a tie line between the primary igneous carbonates and the hydrothermal carbonates in the $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ diagram. The increase in ^{18}O is ascribed to a variable degree of contamination by the ^{18}O -rich hydrothermal carbonates. The contribution of secondary stage hydrothermal carbonates seems to be significant in the Catalao I complex as compared with the Jacupiranga complex. The development of network structure in the Catalao I complex may have enhanced the circulation of the later stage hydrothermal fluids.

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ブラジル国ジャクピランガとカタロン I カーボナタイト岩体の
炭酸塩鉱物の炭素・酸素同位体組成

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要 旨

ブラジル南部の2つのカーボナタイト岩体、ジャクピランガとカタロン I のカルサイトとドロマイトの炭素・酸素同位体比を測定し、次の結果を得た。ジャクピランガの炭素と酸素の同位体比はともに一様で、平均値は $\delta^{13}C = -6.07\%$ 、 $\delta^{18}O = 7.6\%$ 。これらは初生火成岩カーボナタイトの値をしめす。カタロン I の炭素同位体比はほぼ一様で、平均値は $\delta^{13}C = -5.83\%$ である。酸素同位体比は、試料の産状のちがいに、大きな変動幅をしめすが、それは火成活動後の熱水作用に起因するとして説明できる。

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