

Oxygen isotopic study of vein quartz in Neogene-Quaternary overprinting hydrothermal systems in the Toyoha-Muine area, Hokkaido, Japan

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Abstract: An oxygen isotopic study of vein quartz in the Toyoha-Muine area was conducted using a CO₂ laser microprobe technique, in order to determine origin of hydrothermal fluids. The oxygen isotopic values of the hydrothermal fluids resulting in the vein formations were calculated to be between -10.6 and 1.0 ‰, using the obtained oxygen isotopic values of quartz and previous data on the formation temperature of quartz from fluid inclusion studies. Combined with existing chronological data on the hydrothermal activities related to the vein formations, the isotopic values are classified into two different chronological ranges: -6.3 to 1.0 ‰ in Middle to Late Miocene and -10.6 to -6.7 ‰ in Pliocene-Pleistocene. Based on the previous geological reconstruction at the area, the values for the Middle to Late Miocene event indicate mixtures of fluids with variable ratios of meteoric water, seawater and magmatic water, whereas the values for Pliocene-Pleistocene result from fluids with high ratios of meteoric water to magmatic water.

Keywords: oxygen isotope, quartz, hydrothermal fluids, hydrothermal systems, Toyoha, Muine

1. Introduction

The Toyoha-Muine area (hereafter referred to as TMA) is located in southwest Hokkaido. The area has been a part of an active volcanic zone in southwest Hokkaido since Neogene, and Mt. Muine, an andesitic volcano, is located immediately in the south of the research area (Fig. 1). Toyoha, the largest polymetallic (Zn-Pb-Ag-Cu-Sn-In) vein-type deposit in Japan, is situated in the north of the research area (Fig. 1).

The TMA has been investigated by a number of researchers in various fields, including geology, geochronology, geotectonics, geophysics, geochemistry, and mineralogy (e.g., Akome and Haraguchi, 1963; Sawai *et al.*, 1989; Watanabe, 1990; Takakura and Matsushima, 2003; Ohta, 1989, 1995; Shimizu and Aoki, 2011). These studies revealed that the TMA has undergone complex overprinting magmatic-hydrothermal activities, with metallic mineralization since early Miocene. Prior to the present study, two oxygen isotopic studies have been conducted on vein quartz in the TMA (Matsuhisa *et al.*, 1986; Shimizu and Aoki, 2001). These previous studies, however, are restricted to the Pliocene-Pleistocene hydrothermal system.

Combined with recent studies on geochronology, hydrothermal alteration, and fluid inclusions (Aoki *et al.*, 1997; Shimizu and Aoki, 2000; Shimizu *et al.*, 2003;

Shimizu and Aoki, 2011), this paper presents new oxygen isotopic data of hydrothermal fluids for vein quartz in the Middle to Late Miocene, and Pliocene-Pleistocene hydrothermal systems.

2. Outline of geology, hydrothermal alteration, and ore deposit

The geology of the TMA comprises three stratigraphic units: the pre-Tertiary basement consisting of sedimentary rocks (Usubetsu Formation), Miocene volcanic and sedimentary successions (Shiramizugawa, Shiraigawa and Takinosawa Formations, and Asari Group), and the Pliocene Muineyama Lava of andesitic composition (Doi, 1953; Geological Survey of Japan, 1980; New Energy and Industrial Technology Development Organization, here abbreviated to NEDO, 1988). The pre-Tertiary basement and Miocene successions are intruded by numerous dikes (e.g., granodiorite, NEDO, 1988).

Shimizu and Aoki (2011) showed that hydrothermal alteration was extensive in the Miocene successions, and that it could be divided into acid and neutral alterations using data on the pH and temperature stabilities of the alteration mineral assemblages (Utada, 1980; Reyes, 1990). The acid alteration mainly produces quartz, pyrophyllite, dickite, kaolinite, and alunite, whereas neutral alteration forms quartz, sericite, chlorite, pyrite, and calcite. The

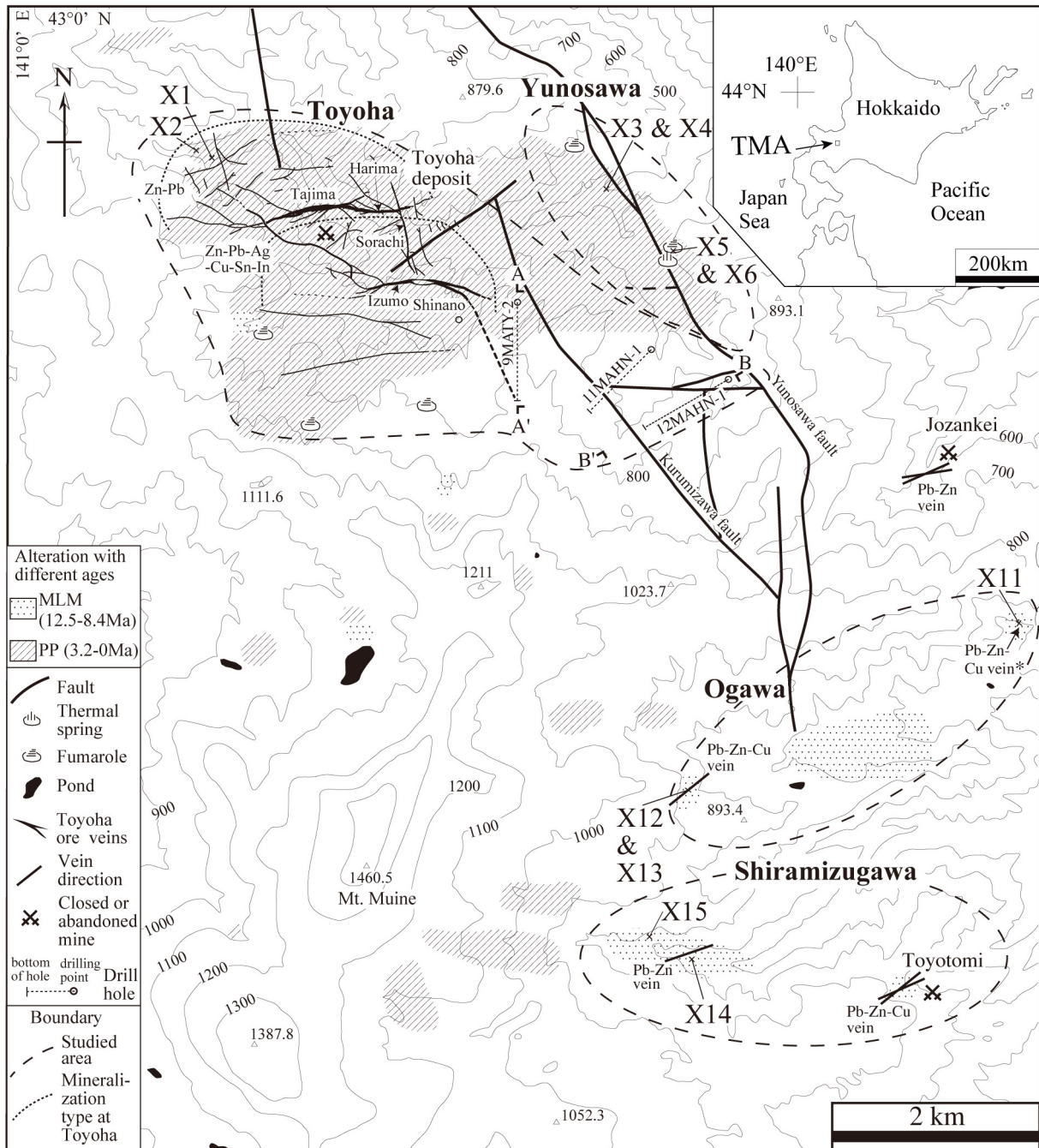


Fig. 1 Sampling locations for oxygen isotope analysis (X1 to X6 and X11 to X15) on a topographical map. Distribution of hydrothermal alteration of different ages and ore veins in the studied sub-areas of Toyoha, Yunosawa, Ogawa, and Shiramizugawa are depicted, after Shimizu and Aoki (2011). Chronological ranges: MLM and PP correspond to Middle to Late Miocene and Pliocene-Pleistocene. The ore veins at the Toyoha deposit are projected to the surface. A-A' and B-B' at Toyoha correspond to the locations of vertical sections in Figs. 2a and 2b, respectively. The short, thick, dashed line between the southeastern end of the Shinano vein and the southern end of 9MATY-2 drilling indicates a NNW-SSE fracture with ore veins intersected by the 9MATY-2 drilling at depth (Shimizu, 2012). *: There was no outcrop of ore veins at the surface; however, ore samples were collected from the dump in a previous prospect to the east of Ogawa.

latter alteration commonly accompanies hydrothermal veins. A number of K-Ar ages were determined for alunite, sericite, and interstratified sericite/smectite to constrain the timing of hydrothermal activities. Most of the ages

fall within three periods: 24.6 Ma, 12.5-8.4 Ma, 3.2-0 Ma (Shimizu and Aoki, 2011). The surface and subsurface distribution of chronologically different alteration phases is displayed in sub-areas of Toyoha, Yunosawa, Ogawa,

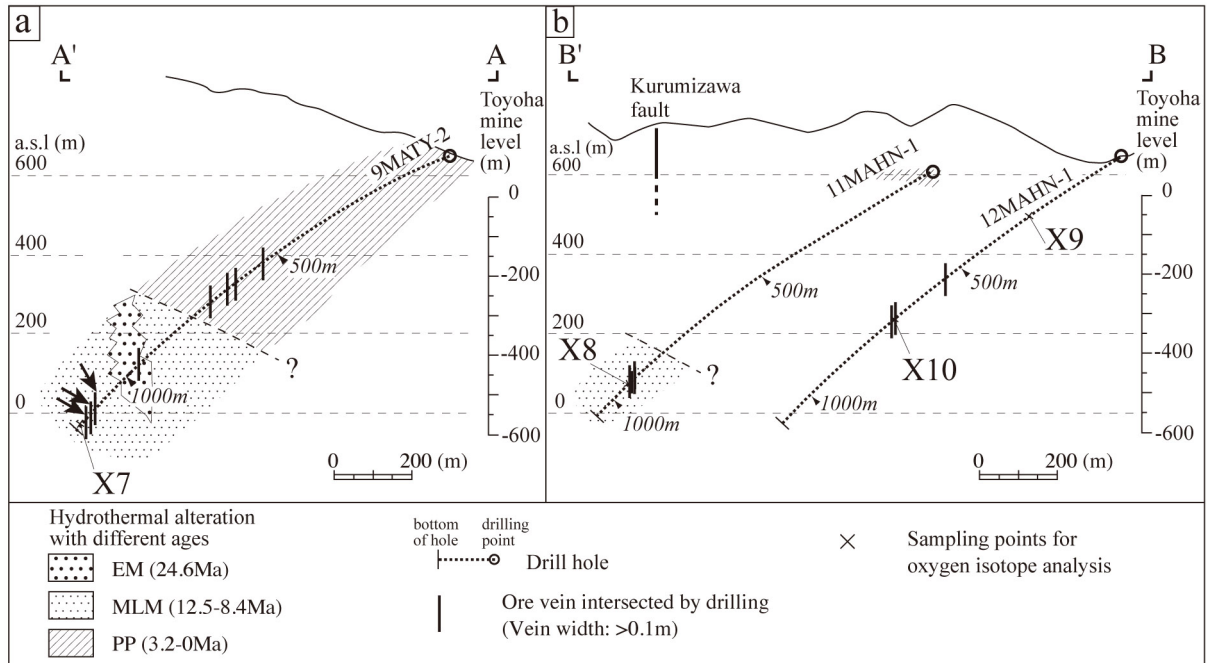


Fig. 2 Cross sections showing sample locations (X7, X8, X9 and X10) for oxygen isotope analysis. The cross sections are modified after the Ministry of International Trade and Industry (2001) and Shimizu and Aoki (2011). The sections A-A' (Fig. 2a) and B-B' (Fig. 2b) correspond to those in Fig. 1. Drill hole 11MAHN-1 is projected to B-B'. The chronological ranges of hydrothermal alteration correspond to those in Shimizu and Aoki (2011). EM: Early Miocene; MLM and PP: the same in Fig. 1. Meter in italic is the distance (m) along each drill hole from the drilling point. Three thick arrows at the depth of 9MATY-2 drilling in Fig. 2a indicate the ore veins that occupy the same NNW-SSE fracture as the Shinano footwall vein in Fig. 1 (Shimizu, 2012).

and Shiramizugawa (Figs. 1 and 2, Shimizu and Aoki, 2011). The Toyoha sub-area is slightly extended to the southeast to include a drill hole: 12MAHN-1 in the present study. The chronological range of the alteration along drill hole 12MAHN-1 is not determined in the present study.

Hydrothermal veins are mainly hosted by the Miocene successions with neutral alteration. The Toyoha deposit is at the north part of the TMA, comprising earlier and later veins characterized by Zn-Pb mineralization to the northwest and Zn-Pb-Ag-Cu-Sn-In mineralization to the southeast, respectively (e.g., Yajima and Ohta, 1979, Fig. 1). Quartz is commonly associated with ore minerals in the earlier veins whereas it tends to occur before and after the polymetallic mineralization in the later veins (Narui *et al.*, 1988).

Numerous quartz and calcite veins occur in the Toyoha, Yunosawa, Ogawa, and Shiramizugawa sub-areas. Most of the veins are barren, with a thickness between 1 and 40 cm. Some quartz veins, however, contain sulfide minerals (e.g., pyrite, sphalerite, galena, and chalcopyrite) and have a variable thickness between 2.5 cm and 50 cm or more (e.g., Ohta, 1997; Shimizu *et al.*, 2003; Shimizu, 2012).

3. Quartz samples

The samples were collected from surface outcrops and

from underground at various localities in the Toyoha, Yunosawa, Ogawa, and Shiramizugawa sub-areas (Figs. 1 and 2). The analyzed samples at Toyoha were collected from the peripheral parts: X1 and X2 to the northwest (Fig. 1) and X7 to X10 to the southeast (Fig. 2). The characteristics of quartz samples as described via oxygen isotopic studies are summarized in Table 1. Samples were obtained from quartz veinlets, except for X2, X5, and X14. Some of the samples (X10, X11, X12, X13, and X14) were intimately associated with sulfide minerals of sphalerite, galena, chalcopyrite, and pyrite.

4. Analytical procedure

Oxygen isotopic study was carried out for the thin sections that were also used for fluid inclusion microthermometry (Fig. 3). Quartz chips were extracted from each section for isotopic analysis. Some of the chips (X1, X3, X4, and X14) that were partly disseminated with goethite were treated with 5% oxalic acid to dissolve the mineral. Quartz aliquots between 0.94 and 2.17 mg were prepared for each isotopic analysis (Table 2).

Oxygen was liberated from the quartz using a CO₂ laser (SYNRAD Model 48-2-115 25W, $\lambda = 10.6 \mu\text{m}$) and a bromine pentafluoride method essentially the same as that described by Sharp (1990). The optical system that

Table. 1 Characteristics of quartz samples for oxygen isotope analysis

Sample name	Sampling location No.	Stratigraphic group	Strike/ dip	Depth above sea level	Vein width (cm)	Vein minerals	Comments
1998-07-03-01	X1	SI ^d	N50°E, 78°N ^b	605 ^b	<4 ^b	Qz ^b	Qz network vein. Sampled at nearby Reibun raise of the Toyoha mine. ^d
1998-07-03-02	X2	SI ^d	N10°E, 70°N ^b	660 ^b	20 ^b	Qz ^b	Qz vein. ^d
MU98102711-5	X3	SM ^c	n.d.	480 ^b	<3 ^b	Qz ^b	Qz veinlet. ^d
MU98102711-6	X4	ditto	ditto	ditto	ditto	ditto	ditto
1998070604D	X5	SM ^c	n.d.	550 ^b	40 ^b	Qz ^b	Qz veinlet nearby fumarole. ^d
1998070604D No.2	X6	ditto	ditto	ditto	ditto	ditto	ditto
Drill core sample							
9MATY-2 1184.1 m	X7	SM ^b	n.d.	-39 ^b	1.5 ^b	Qz-py-sp-gn -chl ^b	Qz veinlet. Oz occurs during and after sp and gn depositions. ^c
11MAHN-1 925.1 m	X8	SM ^c	n.d.	82 ^c	1 ^c	Py-sp-gn-chl -qz ^c	Qz veinlet. Black-umber reniform sp and euhedral gn form in quartz vug. ^c
12MAHN-1 280.2 m	X9	SM ^c	n.d.	500 ^c	5 ^c	Qz-ka ^c	Qz veinlet. Ka occurs in euhedral qz vug. ^c
12MAHN-1 712.9 m	X10	SM ^c	n.d.	225 ^c	≥1.5 ^c	Py-asp-sp-cp -qz ^c	Qz veinlet. A qz-py vein is crosscut by a qz-sp vein. Qz associated with sp was used for fluid inclusion and oxygen isotopic studies. ^c
961030-01	X11	SM ^f	n.d.	590 ^f	n.d.	Qz-cp-py-sp -gn ^f	Cp, py, sp and gn interstitially occur in a qz vein. The sample for fluid inclusion and oxygen isotopic studies was a float. ^f
MA-960919-1-3 No.1 (961030-2 No.1)	X12	SM ^f	n.d.	850 ^a	2.5 ^b	Qz-sp-gn-cp -py-he ^{b, f}	Qz network vein associated with small amounts of ore minerals. The vein was situated below a sp-gn-cp-py-qz vein with thickness, strike and dip of 50 cm, N45°E and 35°N, respectively. ^b
MA-960919-1-3 No.2 (961030-2 No.2)	X13	ditto	ditto	ditto	ditto	ditto	ditto
MU98102804	X14	SM ^f	N65°E, 90° ^b	650 ^b	≥50 ^b	Qz-sp-gn-py -goe ^b	Sp, gn and py sporadically occur in the quartz vein. Goethite occurs along cracks of the vein. ^f
98102901'	X15	SM ^f	N45°W, 90° ^b	790 ^b	1 ^b	Qz ^b	Qz veinlet. ^b

Sampling location Nos. X1 to X19 correspond to those in Figures 1 and 2. Stratigraphic group: SI = Shirai-gawa Formation, SM = Shiramizugawa Formation. n.d. = not determined. Abbreviations: asp = arsenopyrite, chl = chlorite, cp = chalcopyrite, goe = goethite, he = hematite, ka = kaolinite, gn = galena, py = pyrite, qz = quartz, sp = sphalerite. References: a. Ohta (1997); b. Shimizu and Aoki (2000); c. Ministry of International Trade and Industry (2001); d. Shimizu and Aoki (2001); e. Shimizu et al. (2003); f. This study.

introduces a laser beam into the stainless steel reaction chamber through a barium difluoride window is co-axial with a UV-CCD camera. The diameter of the focused laser beam is ~100 μm. The extracted oxygen was converted to CO₂ with a diamond furnace at 1000 °C. A triple-collector mass spectrometer (Finnigan MAT 251) was used for the isotopic analysis, the result of which is expressed in δ¹⁸O

(VSMOW: Vienna Standard Mean Ocean Water) notation. The overall experimental reproducibility in the routine analyses was ±0.1 ‰ (1σ).

The oxygen isotopic values of quartz samples (δ¹⁸O_{quartz} values) were determined, using a δ¹⁸O value of 9.34 of an international silicate standard material (NBS-28) on the SMOW scale (Matsuhisa and Aoki, 1994), resulting

Table. 2 Oxygen isotopic values of quartz and hydrothermal fluids

Sample name	Sampling location No.	Sample weight (mg)	$\delta^{18}\text{O}_{\text{quartz}}$ (‰)	Formation temperature (°C) based on the fluid inclusion study of quartz	$\delta^{18}\text{O}_{\text{fluid}}$ (‰)	Chronological range of hydrothermal alteration ^e
1998-07-03-01	X1	1.90	2.2	192 ^a	-9.9	PP
1998-07-03-02	X2	1.41	4.2	157 ^a	-10.6	PP
MU98102711-5	X3	1.08	2.9	173 ^a	-10.6	PP
MU98102711-6	X4	1.10	3.3	173 ^a	-10.2	PP
1998070604D	X5	2.17	2.2	241 ^a	-7.1	PP
1998070604D No.2	X6	0.94	2.6	241 ^a	-6.7	PP
Drill core sample						
9MATY-2 1184.1 m	X7	1.55	5.6	241 ^b	-3.7	MLM
11MAHN-1 925.1 m	X8	1.04	2.5	253 ^b	-6.3	MLM
12MAHN-1 280.2 m	X9	1.73	2.2	193 ^b	-9.9	n.d.
12MAHN-1 712.9 m	X10	1.39	2.9	213 ^b	-7.9	n.d.
961030-01	X11	1.42	9.9	223 ^c	-0.3	MLM
MA-960919-1-3 No.1	X12	1.16	8.4	284 ^c	1.0	MLM
MA-960919-1-3 No.2	X13	1.57	8.5	284 ^c	1.0	MLM
MU98102804	X14	1.17	4.2	232 ^d	-5.6	MLM
98102901'	X15	1.40	5.1	221 ^d	-5.3	MLM

Sampling locations for X1 to X19 correspond to those in Figure 1. Chronological ranges: MLM and PP are the same as those in Fig. 1. Stratigraphic group: SI = Shiraigawa Formation, SM = Shiramizugawa Formation. A chronological range for samples X9 and X10 is not determined in the present study. n.d. = not determined. References: a. Shimizu and Aoki (2001); b. Shimizu *et al.* (2003), c. Aoki *et al.* (1997), d. Shimizu and Aoki (2000), e. Shimizu (2012).

in 2.5‰, 4.3‰, 3.0‰, 3.4‰, 2.4‰ and 2.8‰ for X1, X2, X3, X4, X5 and X6, respectively (Shimizu and Aoki, 2001). To avoid any potential uncertainty involved in the previous $\delta^{18}\text{O}$ value of the international standard (e.g., an uncertainty caused by a variability of the equilibrium fractionation factor $\alpha_{\text{CO}_2\text{-H}_2\text{O}}$), however, the use of a $\delta^{18}\text{O}$ value of 9.18 on the VSMOW-SLAP (Standard Light Antarctic Precipitation) scale was deemed preferable because it was obtained through the measurement of oxygen gas directly derived from VSMOW and SLAP (Kusakabe and Matsuhisa, 2008). Thus, the present study uses the $\delta^{18}\text{O}$ value of 9.18 for the NBS-28 for the determination of $\delta^{18}\text{O}_{\text{quartz}}$ values of the whole samples (X1 to X15).

5. Results

The $\delta^{18}\text{O}_{\text{quartz}}$ values are listed in Table 2, in addition to the formation temperature data from fluid inclusion studies. The oxygen isotopic values for fluids ($\delta^{18}\text{O}_{\text{fluid}}$

values) were calculated from the $\delta^{18}\text{O}_{\text{quartz}}$ values and the formation temperatures, using an equation of quartz-water isotopic equilibrium ($1000\ln\alpha = 3.34 \times 10^6/T^2 - 3.31$; Matsuhisa *et al.*, 1979, Table 2).

The $\delta^{18}\text{O}_{\text{fluid}}$ values and a previous dataset (Matsuhisa *et al.*, 1986) are plotted in Fig. 4 for the different research sub-areas in the TMA. All the $\delta^{18}\text{O}_{\text{fluid}}$ values range between the values of meteoric water (nearby spring water, -12.6 to -12.5 ‰, Matsubaya *et al.*, 1978) and magmatic water (6 to 10 ‰, Giggenbach, 1992). Some $\delta^{18}\text{O}_{\text{fluid}}$ values (-0.3 to 1.0 ‰) are close to the value of seawater (0 ‰, SMOW).

6. Discussion

The common association of the quartz veins with neutral-pH alteration in the Miocene successions (Shimizu and Aoki, 2011; Shimizu, 2012) indicates that the quartz veins and the neutral-pH alteration formed contemporaneously due to hydrothermal activities. The author suggests that the analyzed quartz samples, except for X9 and X10, are

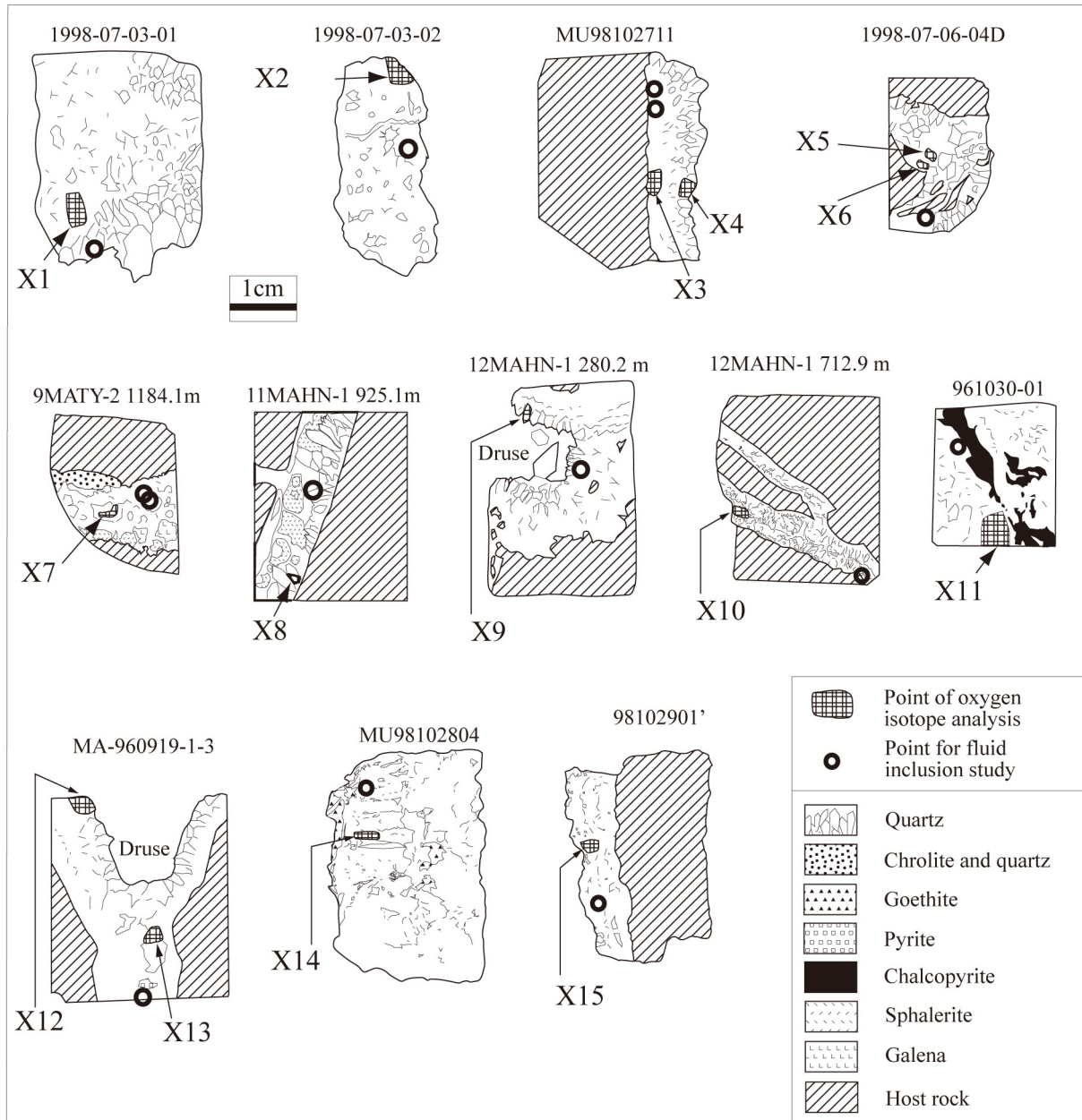


Fig. 3 Schematic map of locations for $\delta^{18}\text{O}$ laser microprobe analysis and fluid inclusion microthermometry for quartz in thin sections with a thickness of 0.2 to 0.3 mm.

genetically related to either Middle to Late Miocene (12.5-8.4 Ma) or Pliocene-Pleistocene (3.2-0 Ma) hydrothermal activities (Table 2).

Based on geological reconstructions in Middle to Late Miocene, the TMA was thought to have been located under a submarine environment during a large part of the period, but in terrestrial conditions between 10.5 and 8.5 Ma, except for the north part of the TMA during submarine volcanic activity at 9.5 Ma (Watanabe and Iwata, 1986; Watanabe and Ohta, 1999). A variety of magmatic activities, forming basalt, hyaloclastitic andesite, and intrusive granodiorite, led to many centers of hydrothermal activities in the north

and east parts of the TMA including Toyoha, Ogawa, and Shiramizugawa sub-areas (Shimizu and Aoki, 2011). The present study suggests that the $\delta^{18}\text{O}_{\text{fluid}}$ values of -6.3 to 1.0 ‰ from the peripheral parts at the Toyoha, Ogawa, and Shiramizugawa sub-areas (Fig. 4) resulted from mixing of magmatic water, seawater, and meteoric water in variable ratios. The intimate association of sulfide minerals with quartz in some samples (X10, X11, X12, X13, and X14, Table 1) from the Middle to Late Miocene alteration indicates that the hydrothermal fluids of magmatic origin carried sulfur from magma chambers. This is consistent with the sulfur isotopic study of sulfides concluding that

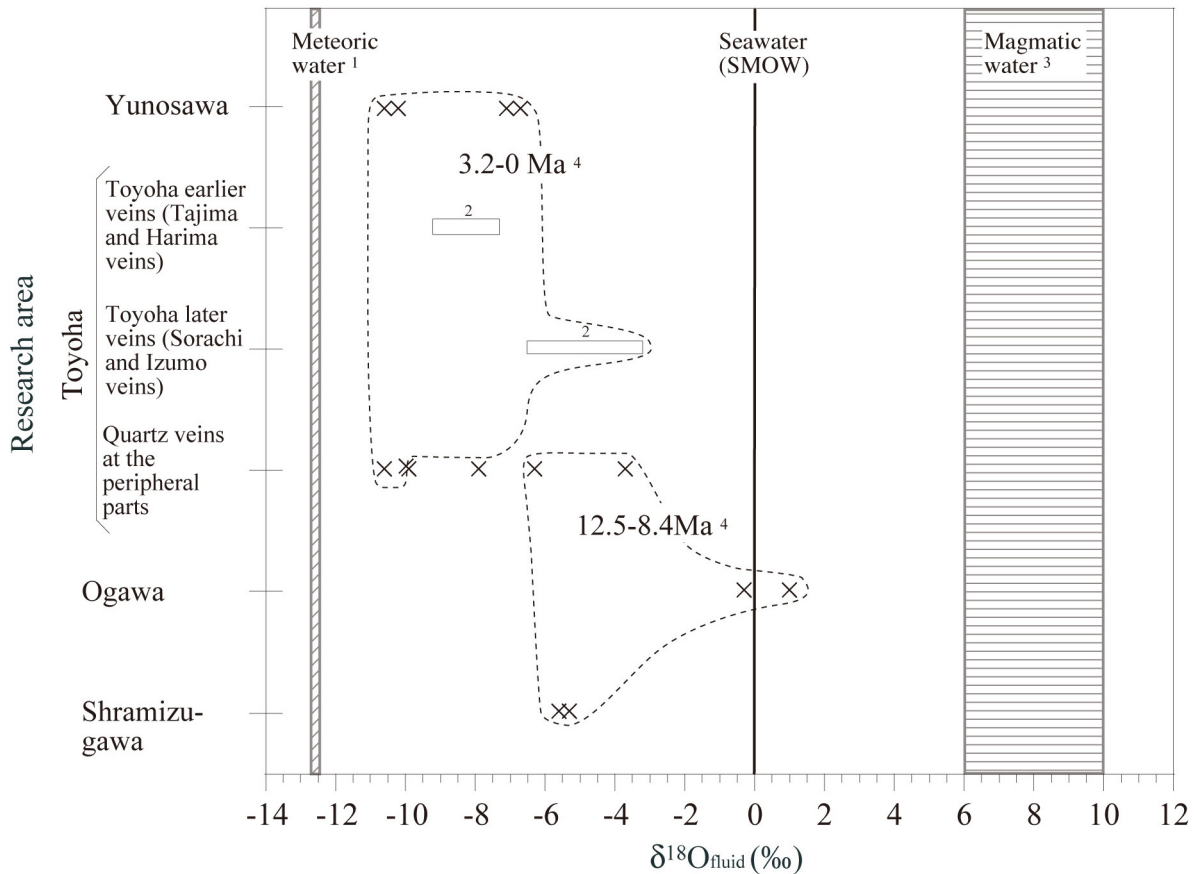


Fig. 4 Oxygen isotopic values of hydrothermal fluids for vein formations in the Toyoha-Muine area. Research areas: Toyoha, Yunosawa, Ogawa, and Shiramizugawa correspond to those in Figs. 1 and 2. References: 1, Matsubaya *et al.* (1978); 2, Matsuhisa *et al.* (1986); 3, Giggenbach (1992); 4, Shimizu and Aoki (2011). The ranges of isotopic values in the Toyoha earlier and later veins are shown as a bar.

the sulfide sulfur was sourced from arc magmatism in the Toyoha-Muine area (Shimizu, 2012).

In Pliocene-Pleistocene, the TMA was situated in a terrestrial environment with active magmatic-hydrothermal systems as a result of active uplift in western Hokkaido from Late Miocene (Yahata, 2002). The $\delta^{18}\text{O}_{\text{fluid}}$ values for the earlier (-9.2 to -3.2 ‰ in Fig. 4) and later veins of the Toyoha deposit (-6.5 to -3.2 ‰ in Fig. 4) imply that the hydrothermal water for the vein formations were generated as a result of mixing between shallow meteoric water and deep water, possibly magmatic in origin (Matsuhisa *et al.*, 1986). The relatively low $\delta^{18}\text{O}_{\text{fluid}}$ values for the earlier vein formation (-9.2 to -7.3 ‰) attributed to lower ratios of deep water to shallow water (Matsuhisa *et al.*, 1986). The similar range of $\delta^{18}\text{O}_{\text{fluid}}$ values from the Yunosawa sub-area and peripheral parts at Toyoha (-10.6 to -6.7 ‰ in Fig. 4) suggests that origin of hydrothermal water for the vein formations in the sub-area and parts is similar to that of the earlier veins at the Toyoha deposit. If the quartz samples (X9 and X10 in Table 2) in the peripheral parts were genetically related to the Pliocene-Pleistocene hydrothermal activity, the $\delta^{18}\text{O}_{\text{fluid}}$ values (-9.9 for X9 and

-7.9 ‰ for X10 in Fig. 4) could be interpreted as mixing of high ratios of meteoric water with magmatic water. The interpretation of the $\delta^{18}\text{O}_{\text{fluid}}$ values in Pliocene-Pleistocene in the TMA is similar to that for the nearby hydrothermal veins at the Koryu and Chitose Au-Ag deposits formed in Pliocene-Pleistocene (Hattori and Sakai, 1979; Sawai *et al.*, 1992; Shimizu *et al.*, 1998).

7. Conclusions

Combined with previous geological and geochemical studies in the TMA, the $\delta^{18}\text{O}_{\text{fluid}}$ values for quartz vein formations are interpreted according to two different chronological ranges of hydrothermal activities, in Middle to Late Miocene and Pliocene-Pleistocene. The relatively high $\delta^{18}\text{O}_{\text{fluid}}$ values of Middle to Late Miocene (-6.3 to 1.0 ‰) resulted from mixing of magmatic water, seawater, and meteoric water with variable ratios, whereas the relatively low $\delta^{18}\text{O}_{\text{fluid}}$ values of Pliocene-Pleistocene (-10.6 to -6.7 ‰) are a result of mixing magmatic water and meteoric water, but with a dominant contribution of the latter water source.

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北海道豊羽 - 無意根地域における
新第三紀 - 第四紀重複熱水系 脈石英の酸素同位体の研究

清水 徹

要 旨

熱水の起源を明らかにするため、CO₂レーザーマイクロプローブ法を用いて、豊羽-無意根地域の脈石英の酸素同位体の研究を行った。得られた石英の酸素同位体比データと既存の流体包有物研究によって得られた形成温度データを用いて、脈形成期の熱水の酸素同位体比は、-10~1.0‰と計算された。脈形成に関与した熱水系の年代値データと併せると、その酸素同位体比は、-6.3~1.0‰（中期~後期中新世）及び-10.6~-6.7‰（鮮新世~更新世）という二つの形成時期に分類された。既存の地質環境復元データに基づくと、中期~後期中新世の熱水の酸素同位体比は、天水、海水及びマグマ水が様々な割合で混合して熱水が形成されたことを示す。一方、鮮新世~更新世の熱水の酸素同位体比は、天水とマグマ水が混合して熱水が形成されたことを示すが、天水の割合が高かったことを示す。