Geology, Age and Style of the Advanced Argillic Alteration in the Kobui Area, Southwestern Hokkaido, Japan

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Abstract: Advanced argillic alteration in the Kobui area, southwestern Hokkaido, is distributed over 5.3 km² within the Pliocene Kobui andesite lavas, and overlain by Late Pleistocene Maruyama andesite lavas. The advanced argillic alteration consists of acid-leached andesite (including vuggy silica), hydrothermal breccia and massively silicified rocks, and is surrounded by argillic alteration characterized by smectite and pyrite.

The alteration is divided into western Kobui (KAA) and eastern Tokiwamatsu-Furube advanced argillic alteration areas (TFAA) in distribution. KAA formed during the period from 2.9 to 1.8 Ma and TFAA from 1.3 to 0.2 Ma, based on K-Ar ages on alunites. KAA is composed of a relatively large mass with several small exposures, which includes a hydrothermal breccia with strikes of N50°W and N70°E. TFAA consists of four isolated sub-areas aligning in a N10°E direction, with hydrothermal breccia subparallel to the alignment (N5°E, N20°E). In both areas, advanced argillic alteration commonly yield alunite- and kaolinite-group minerals, and KAA is characterized by the presence of pyrophyllite, diaspor, topaz and zinnite, whereas TFAA lacks these minerals and contains tridymite and cristobalite. The mineral assemblage and sequence, and sulfur isotopic temperatures suggest that the temperature of the hydrothermal fluids during the main alteration in KAA was higher (200°C-320°C) than that in TFAA (~200°C). Crater-forming explosions followed the advanced argillic alteration at both KAA and TFAA, and lacustrine sediments accumulated within the craters. Successive hydrothermal activity occurred after the crater formation is characterized by H₂S-dominant, more reduced fluids. This activity disseminated sulfur, arsenic, antimony and mercury in the lacustrine sediments. The loci of the activity has shifted to the east since 2.9 Ma.

1. Introduction

Lithocaps are potentially voluminous zones of advanced argillic and associated argillic alteration located above the subvolcanic intrusive environment, and they constitute exploration objectives for epithermal and porphyry-type deposits (SILLITOE, 1995). The Northeast Japan arc includes several lithocaps of Pliocene and Pleistocene age, which are situated mostly along its volcanic front (Fig. 1). These lithocaps are Muine (MITI, 1977), Koryu (FUJIKAWA et al., 1995), Orofure (YAHATA, 1989), Kobui (this study), Osorezan (AOKI, 1992), Eboshidake (MITI, 1996), Joboji, Inaniwa, Kuzumarukawa (YAMADA, 1995) and Ohtoge (ZENG et al., 1996). Among these lithocaps, high-grade gold precipitation was discovered in Osorezan (AOKI, 1992) and Ohtoge (SHIJO, 1992), and hence the Metal Mining Agency of Japan (MMAJ) started exploration on these lithocaps (MITI, 1995, 1996, 1997). In 1994, MMAJ discovered extensive advanced argillic and associated argillic alteration (wider than 5 km²) developed in the Kobui area (Fig. 1), southwestern Hokkaido (MITI, 1995). This paper presents the geologic framework, occurrence, age and structure of the alteration, and discusses the style of hydrothermal activity which formed the advanced argillic alteration.

2. Geology

2.1 Geologic setting

The Kobui area is situated in the Kameda peninsula, where epithermal base and precious metal deposits cluster in association with the Neogene igneous activity (Fig. 2). In the Kameda peninsula, Mesozoic sedimentary rocks (Toi Formation) underlie Middle Miocene submarine andesitic volcanic rocks (Kunnui Formation) and Middle to Late Miocene submarine sedimentary rocks (Shiodomarigawa Formation) (HATA and TSUSHIMA, 1971; HATA and UEMURA, 1983). These Miocene rocks are intruded by NNW-SSE trending basaltic dikes of Middle Miocene age, and WNW-ESE trending dactitic intrusions of Late Miocene age (HATA and TSUSHIMA, 1971; HATA and UEMURA, 1983). Andesitic rocks of Pliocene and Quaternary age cover these older rocks, forming a NW-SE trending volcanic chain (WATANABE...
Fig. 1 Distribution of Plio-Pleistocene lithocaps along the Northeast Japan arc. The ages of the lithocaps are referred to as follows: Koryu (FUJIKAWA et al., 1995), Kobui (This study), Osorezan (AOKI, 1991) and Ohtoge (ZENG et al., 1996).


et al., 1996). Major Quaternary volcanoes in the chain include Komagadake, Yokotsudake, Maruyama and Esan from northwest to southwest. The Kobui area is located near to the southeastern end of the volcanic chain (Fig. 2).

Epithermal deposits in the peninsula are base metal deposits within the Mesozoic or Miocene sedimentary rocks (Zenigamezawa, Tsuchiya-Ishizaki, Kamae and Kamaya), and gold deposits within the volcanic rocks of Miocene or younger age (Yamato, Hokko and Taisei). Exhalative-sedimentary sulfur deposits (Hakodate, Kobui, Furube, Takinosawa, Tokiwamatsu, Oshino and Esan), an arsenic deposit (Kinoashi), an iron sulfide deposit (Shojingawa) and a sedimentary limonite deposit (Amemasugawa), are located near to the Quaternary volcanoes (MITI, 1992; WATANABE et al., 1996).

2. 2  Geology of the Kobui area

The Kobui area, approximately 8 km by 6 km, includes the Quaternary Maruyama volcano (Figs. 2 and 3, Plate 1a). Mesozoic sedimentary rocks are not exposed at the surface in the mapped area, whereas several isolated exposures of mudstone, chert and limestone of the Toi Formation are distributed to the southwest of the area (SHOYA and TAKAHASHI, 1967). The Neogene and Quaternary strata are divided into the Miocene Shiodomarigawa Formation, Pliocene Kobui Lavas, and Pleistocene Maruyama and Futago Lavas in ascending order (Appendix 1). A dacitic intrusive body of Late Miocene age occurs within the Shiodomarigawa Formation. Pleistocene lacustrine sedimentary rocks are locally distributed and overlie the Kobui Lavas (Fig. 3; Appendix 1).

The Shiodomarigawa Formation is distributed in the north and southwest of the area (Fig. 3). The formation is composed of alternating beds of siliceous mudstone, tuffaceous sandstone, siltstone and andesitic hyaloclastite, partly intercalated with andesite lavas. The general strike of the formation is WNW-ESE and gently dips southward. Marine diatoms of the Denticulopsis
The Shiodomarigawa Formation is intruded by dacitic porphyries. The porphyries have a WNW-ESE trend, and are distributed upstream of the Yagi, Kinaoshi, Ponkinaoshi and Kobui Rivers (Fig. 3). They are composed of multiple intrusive rocks, attaining an approximate total thickness of 3 km. The central part of the intrusive rocks include coarser-grained porphyries than those at the margins. They are holocrystalline and contain phenocrysts of plagioclase, clinopyroxene, orthopyroxene, opaque minerals in a matrix of granular quartz and plagioclase, and partly affected by propylitic alteration. The intrusive rocks extend to the Minamikayabe area, where one of the quartz porphyries is dated at 6.2 Ma by the fission-track technique (NEDO, 1988).

Terrestrial andesite lavas (Pliocene Kobui Lavas, and Pleistocene Maruyama and Futago Lavas) unconformably overlie the Shiodomarigawa Formation and the dacitic porphyries (Fig. 3). In spite of indistinct shape of the "Kobui volcano or volcanoes" due to erosion of its upper portion, eruptive centers of the Kobui Lavas appear to have situated near to the center of the mapped area, because the lavas flowed northward and westward in the western and central parts of the area and eastward in the eastern part. The base of the Kobui lavas is marked by tuff breccia and volcanic breccia. The maximum thickness of the lava is 150 to 200 m. The lavas consist of pyroxene andesite, which includes phenocrysts of plagioclase, clinopyroxene, orthopyroxene and opaque minerals in an intersertal matrix of plagioclase, pyroxene and glass. The phenocrysts of pyroxenes and plagioclase are occasionally up to 4 mm in size. The andesite contains rare fragments of diorite and corroded quartz grains.

There are several explosion craters within the Kobui Lavas such as Yagi, Kobui, Okaji, Tokiwamatsu, Takinosawa and Furube. They are 1 km or less in diameter, based on circular topography. These craters include sulfur-disseminated lacustrine sedimentary rocks inside except for the Yagi craters (Fig. 3). The Yagi crater is
accompanied by sulfur-disseminated acid-leached andesite near to the center of the crater as well as sandstone without sulfur dissemination at its northwestern margin. A similar topographic feature is recognized at the “Aidomari crater”, which is covered by landslide debris and sedimentary rocks are not exposed. The lacustrine sedimentary rocks abut the Kobui Lavas, and are covered by the Pleistocene Maruyama Lavas in the Kobui and Okaji craters (Fig. 3). These sedimentary rocks consist of alternating thin beds of siltstone and mudstone, and commonly include plant fragments. The sedimentary rocks in the Takinosawa and Furube craters are intercalated with felsic welded pyroclastic flow deposits. The pyroclastic flow deposits are commonly silicified and disseminated by sulfur and orpiment. Massive fine pyrite is included within the sedimentary rocks in the Takinosawa crater. The age of the sedimentary rocks is inferred to be Pleistocene, based on the stratigraphic relationships (Appendix 1).

The Maruyama Lavas cover the Kobui Lavas and the lacustrine sedimentary rocks, and are divided into two topographic units. The lower unit is composed of pyroxene andesite lavas, which change into pyroclastic flow deposits toward the distal parts away from the eruptive center. The upper unit forms a lava dome approximately 700-800 m in diameter at Mt. Maruyama and an explosion crater lies on the northwest of the dome (Fig. 3, Plate 1a). The eruptive center of these units may lie near to Mt. Maruyama (Fig. 3). The andesite lavas both of the upper and lower units contain phenocrysts of plagioclase, clinopyroxene, orthopyroxene and opaque minerals. The lower andesite exhibits an intersertal texture, whereas the upper andesite has a glassy or cryptocrystalline texture with distinct flow banding.

The Futago Lavas are distributed in the western part of the survey area, overlying the Kobui Lavas (Fig. 3). The lavas form two domes consisting of pyroxene andesite. The lavas can be correlated to the Maruyama Lavas, because they are not hydrothermally altered and cover the advanced argillic alteration, although no age data is available for the Futago Lavas.

The area includes several exhalative-sedimentary sulfur deposits, which used to be one of the largest sulfur-producing areas in Japan (Takabatake, 1952). They are Kobui, Oshino, Okaji, Tokiwamatsu, Takinosawa and Furube sulfur deposits and Kinaoshi arsenic deposit (Fig. 3). The amounts of sulfur produced are 169,323 t (Kobui), 8,443 t (Furube), 6,575 t (Tokiwamatsu), 2,270 t (Takinosawa) and 596 t (Okaji) (Saito et al., 1967). Slightly silicified non-economic limonite beds are commonly found downstream of these sulfur deposits. Present-day hot spring activity precipitates arsenic and sulfur near to the Kobui River in the south of the area (Fig. 3). Around the spring, black or yellow-colored metal-rich clay less than a meter thick is distributed.

3. Advanced Argillic and Associated Argillic Alteration

Advanced argillic alteration characterized by the presence of pyrophyllite, alunite-group minerals (minamiite, alunite and natroalunite) and kaolinite-group minerals (kaolinite, dickite and nacrite) occur in the Kobui Lavas (Fig. 4). The advanced argillic rocks form topographic highs and are surrounded by argillic rocks characterized by the abundant presence of smectite and pyrite. The advanced argillic rocks converge downwards to compact dike-like hydrothermal breccias, exhibiting a funnel shape in a cross-section (Fig. 5). The advanced argillic alteration exposed at the surface is divided into the western and eastern parts, located at both sides of the Maruyama Lavas (Fig. 4). In this paper, the western and eastern parts are named the Kobui and Tokiwamatsu-Furube advanced argillic alteration areas, respectively.

3.1 Kobui advanced argillic alteration area (KAA)

KAA is exposed over an area approximately 4.1 km², and the largest exposure lies between the Maruyama and Futago Mountains. KAA includes the Yagi crater and a part of the Kobui crater, and is overlain by non-altered Maruyama and Futago Lavas (Fig. 4). Several small exposures of advanced argillic rocks are found as nodes within the Maruyama Lavas to the northeast of the main exposure, where the advanced argillic rocks of the Kobui Lavas are in direct contact with non-altered andesite lavas or pyroclastic flow deposits of the Maruyama Lavas. Small exposures are also found to the southeast of the main exposure in the argillic alteration (Fig. 4). KAA includes massively altered andesite lavas (Plate 1c), hydrothermal breccia (Plates 1d), vuggy silica (Plate 1e), and massively silicified rocks (Plate 1f). Massively altered andesite retains original volcanic textures, although phenocrysts of the andesite are completely replaced by pyrophyllite, alunite- and kaolinite-group minerals. Hydrothermal breccias consist of acid-leached and/or silicified breccias in a matrix of acid-leached volcanic fragments with alunite, pyrophyllite and limonite. The hydrothermal breccia contains rarely late-stage quartz veinlets. Vuggy silica occurs mainly in the Yagi crater and south of the Kinaoshi deposit. It consists of completely acid-leached massive andesite lava and hydrothermal breccia, which contain quartz and a subordinate amount of rutile or anatase. Massively silicified rocks, which form nodes within the advanced argillic alteration, consist of homogeneous, anhedral fine-grained compact quartz (Plate 2d). The largest body (700 m by 500 m in plan view) is situated to the north of the Yagi crater and a small body lies on the edge of the crater (Fig. 4). Pseudomorphs of hydrothermal
Fig. 4 Map showing the distribution of the advanced argillic and associated argillic alteration in the Kobui area. Orientation of the dike-like hydrothermal breccia, circular structures of the explosion craters and locations of sulfur deposits are also indicated.

Fig. 5 Cross-section of KAA. The location of section A-B is shown in Fig. 3.

breccias cemented by quartz occurs at the center of this largest body. The advanced argillic alteration in KAA converges downwards to vertical dike-like hydrothermal breccias which are approximately 10 meter wide, with strikes of N50°W and N70°E (Fig. 4).

3. 2 Tokiwamatsu-Furube advanced argillic alteration area (TFAA)
TFAA is composed of four isolated sub-areas, which align in a N10°E direction (Fig. 4). These subareas are separated by argillic alteration. The total surface exposure of TFAA amounts to 1.2 km². Individual sub-areas are located at the margin of the explosion craters of Furube, Takinosawa and Tokiwamatsu, except the southernmost sub-area, where no explosion crater has been recognized. TFAA consists mainly of hydrothermal breccia, massively altered andesite and vuggy silica. The Tokiwamatsu sub-area has the widest exposure, half of which is occupied by vuggy silica. Dike-like hydrothermal breccias with N5°E and N20°E strikes are present in the Tokiwamatsu and Furube sub-areas, respectively (Fig. 4; Plate 1b).

3. 3 Alteration mineral assemblages
The advanced argillic and associated argillic alteration in the Kobui area is divided into six main alteration zones, based on the mineral assemblage detected by XRD. The alteration zones from the inner to the
Fig. 6 Alteration mineral assemblage in each alteration zone. Thick and thin lines indicate approximate amounts (abundant and rare, respectively) of the altered minerals.

* Smectite: chlorite-smectite and illite-smectite mixed layer clays are included.

outer zone are as follows; pyrophyllite-diaspore-alunite, alunite-minamiite, alunite, dickite-kaolinite and smectite zones (Figs. 4 and 6).

Pyrophyllite-diaspore-alunite zone: The pyrophyllite-diaspore-alunite zone is characterized by the presence of pyrophyllite, diaspore, alunite-group minerals, kaolinite-group minerals, pyrite, topaz and zunyite (Fig. 6). Andalusite is rarely present. This assemblage is not found in TFAA, but widely in KAA. The rocks in the zone are mostly dominant in either pyrophyllite or alunite-group minerals. The relationship between the topaz and zunyite is the same. Among the 18 samples that include both pyrophyllite and kaolinite-group minerals in KAA, pyrophyllite coexists with nacrite and/or dickite in 16 samples, and with kaolinite in two samples. These two samples are both taken from the margin of the pyrophyllite-diaspore-alunite zone. Diaspore generally occurs with pyrophyllite, with or without dickite, whereas it is rare to find coexisting with the alunite-group minerals. Topaz and zunyite occur mainly within the Yagi and Kobui craters with pyrophyllite or alunite-group minerals in KAA (Fig. 4).

Alunite-minamiite zone: The alunite-minamiite zone is distinguished from the pyrophyllite-diaspore-alunite zone by the absence of pyrophyllite, diaspore, zunyite and topaz, and is characterized by the presence of minamiite (Fig. 6). Alunite-group minerals commonly coexist. This zone is distributed in two areas; south of the Kinaoshi deposit in KAA, and the Tokiwamatsu subarea in TFAA (Fig. 5). These areas are both accompanied by vuggy silica bodies. This mineral assemblage also occurs at the dike-like root zones in KAA.

Alunite zone: The alunite zone is distinguished from the alunite-minamiite zone by the absence of minamiite. The main constituent minerals of this zone are alunite, natroalunite, kaolinite-group minerals, pyrite and quartz (Fig. 6). This zone is located near to the Furube and Takinosawa craters in TFAA (Fig. 4).

Dickite-kaolinite zone: This zone is characterized by kaolinite-group minerals, pyrite and quartz with or without a small amount of pyrophyllite and smectite (Fig. 6). This zone fringes the pyrophyllite-diaspore-alunite zone, although the zone is, in general, not wide enough to be mapped in KAA. The gradational change from the alunite-minamiite or alunite zones to the dickite-kaolinite zone is only mappable in the southernmost sub-area of TFAA.

Kaolinite zone: This zone is distinguished from the dickite-kaolinite zone by the absence of dickite and nacrite. The main constituent minerals are kaolinite, quartz, cristobalite, tridymite, sulfur and pyrite with subordinate amounts of smectite and alunite of supergene origin.
This alteration mineral assemblage occurs in the lacustrine sedimentary rocks within the Furube, Takinosawa, Tokiwamatsu, Kobui and Okaji craters. The kaolinite zone seems to abut the other alteration zones (Fig. 4).

Smectite zone: This zone is characterized by abundant smectite and pyrite with or without minor amounts of kaolinite, chlorite and illite (Fig. 6). This zone fringes the advanced argillic alteration (pyrophyllite-diaspore-alunite, alunite-minamiite and alunite zones) through a thin dickite-kaolinite zone (Fig. 4). Among the lacustrine sedimentary rocks, only those of the Yagi crater suffer smectite alteration.

The transition from the pyrophyllite-diaspore-alunite zone to the smectite zone through the dickite-kaolinite zone is recognized not only in a map scale but also in an outcrop scale at the margin of the advanced argillic alteration. At Locality KB-110, northwest of the Kobui crater (Fig. 4), pyrophyllite-diaspore-kaolinite alteration of a couple of meters thickness within the andesite of the Kobui Lavas changes into the smectite zone through the 10 cm-thick kaolinite-rich zone (Fig. 7).

Propylitic alteration characterized by the assemblage of quartz, chlorite, illite, albite and pyrite with or without calcite and zeolite is recognized within the dacitic intrusions exposed along the Kobui River (Fig. 4). The relationship of the advanced argillic and propylitic alterations is not clear. Unconsolidated clay near to the sulfur- and arsenic-bearing hot spring contains cristobalite, tridymite, smectite, sulfur and orpiment.

3.4 Occurrence of alteration minerals

Pyrophyllite, diaspore, topaz and zunyite generally occur in vugs of the acid-leached rocks. Alunite-group minerals occur in vugs of acid-leached andesite (Plates 1c and 2b) as well as in a matrix of hydrothermal breccia (Plate 1d). Minamiite is generally coarser (0.1-0.5 mm; Plates 2e and f) than alunite and nratroalunite (< 0.2 mm; Plates 2a, b and c), and coexists occasionally with pyrite (Plates 2e and f). Some alunites include woodhouseite or svanbergite (Sr-Ba-woodhouseite), which are partially dissolved (Figs. 8a and b). These alunite crystals also enclose euhedral zunyite (Fig. 8a). Some alunites have oscillatory zoning of Ca-, Na- and K-rich bands (Fig. 8b).

Pyrite occurs in all the alteration zones except vuggy silica. In particular, pyrite is relatively concentrated in dike-like root zones of hydrothermal breccia, coexisting with alunite-group minerals (Plates 2e and f). Pyrite in the massively silicified zone and in the smectite zone is generally euhedral, whereas pyrite coexisting with alu-
3.5 Trace element variations

Trace element variations in non-altered andesite, each alteration zone, vuggy silica, massively silicified rocks, and lacustrine sedimentary rocks including unconsolidated hot spring sediments are shown in Figure 9. Gold concentrations in the non-altered andesites, argillic and advanced argillic rocks are lower than 20 ppb on average, except for the massively silicified rocks. The gold concentration tends to increase from the non-altered andesite to advanced argillic rocks, and the massively silicified rocks include ca. 100 ppb Au on average. The silver concentration has a similar occurrence to the gold, whereas the copper concentration has no significant variation among the non-altered andesites, argillic and advanced argillic rocks. Arsenic, mercury and antimony are concentrated in the lacustrine sedimentary rocks. Unconsolidated clay near to the active hot spring contains 1-2 wt. % of arsenic, 0.2 ppm to 38.6 ppm of mercury and 40 to 270 ppm of tellurium (Table 1).

4. K-Ar Ages of Andesite and Alunite

4.1 Samples analyzed

An andesite sample (a) of the Maruyama Lavas and six alunite samples (b, c, d, e, f and g) were dated by the K-Ar technique in order to determine the age of the lavas and the alteration. The sample localities are shown in Figures 3 and 4.

The andesite sample is taken from the lower unit that contains phenocrysts of plagioclase, clinopyroxene, orthopyroxene and opaque minerals. No hydrothermal alteration is recognized in the sample.

Three samples of alunite-group minerals (e, f and g) were taken from KAA (Fig. 4). Sample g was taken from the N50°W trending hydrothermal breccia and e from a hydrothermal breccia outcrop approximately 100 meters to the west. Sample f was taken from the N70°E trending hydrothermal breccia in the Kobui Lavas. Samples e, f and g consist of alunite-quartz (-minamiite), minamiite-alunite-pyrite-quartz, and alunite-quartz with minor amounts of zunyite, andalusite and geothite,
Fig. 10 Photographs of quartz-alunite samples used for K-Ar dating in the Furube advanced argillic subarea. The upper photograph (a) shows a fragment of hydrothermal breccia, which contains fragmented vuggy silica in a matrix of alunite (Sample b in Table 2). The lower photograph (b) is a fragment of acid-altered andesite lava in which vugs are filled with alunite (Sample c in Table 2).

respectively. The other alunite samples (b, c and d) were taken from TFMA; two (b and c) from the Furube and one (d) from the Tokiwa-matsu sub-areas (Fig. 4). Sample b is a fragment of hydrothermal breccia, which contains breccias of vuggy silica in an alunite matrix (Fig. 10a). Samples c (Fig. 10b) and d are fragments of acid-altered andesite lavas, of which vugs are filled with alunite-group minerals. Samples b and c consist of alunite-quartz, and sample d, minamiite-alunite-quartz.

Alunite-group minerals were separated with a dental drill and mineral compositions of the separated samples were checked by XRD. These samples contain a very small amount of quartz as well as the alunite-group minerals.

4.2 Result

The result of the measurements by the Central Research Institute, Mitsubishi Materials Corporation, are shown in Table 2. The constants used for the age calculation are \( \lambda_p = 4.962 \times 10^{-10}/y \), \( \lambda_\alpha = 0.581 \times 10^{-10}/y \) and \( ^{40}K/K = 0.01167 \) atom% (Steiger and Jäger, 1977). The calculation of the measurement error \((\sigma)\) was followed to Nagao et al. (1984). The measurement was performed twice for each sample, and the age with least air contamination for each sample is used in the paper. The age of the andesite is interpreted as the age of the andesite eruption, and those of the alunite samples as the ages of the mineral precipitation due to reaction between the hydrothermal solutions and host rocks.

These ages suggest that the Kobui Lavas erupted before ca. 2.9 Ma, the oldest age of the alunites in the advanced argillic alteration hosted by the Kobui Lavas, whereas the Maruyama Lavas are Late Pleistocene in age.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Name of formation or alteration area (sample location)</th>
<th>Rock type</th>
<th>Analyzed material</th>
<th>K wt%</th>
<th>Rad. (^{40})Ar (10(^6)cc/g)</th>
<th>K-Ar age (Ma)</th>
<th>Air Cont. (%)</th>
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<tr>
<td>a</td>
<td>Maruyama Lavas (41°51'00&quot;N, 141°06'36&quot;E)</td>
<td>Pyroxene andesite</td>
<td>Whole rock</td>
<td>0.76±0.05</td>
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<td>0.13±0.07</td>
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<tr>
<td>b</td>
<td>TFMA (41°51'40&quot;N, 141°07'09&quot;E)</td>
<td>Alunite-quartz</td>
<td>Alunite</td>
<td>6.30±0.13</td>
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<td>0.23±0.08</td>
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<td>c</td>
<td>TFMA (41°51'40&quot;N, 141°07'09&quot;E)</td>
<td>Alunite-quartz</td>
<td>Alunite</td>
<td>2.64±0.08</td>
<td>7.01±2.66</td>
<td>0.69±0.26</td>
<td>96.2</td>
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<td>d (Tokiwa-matsu)</td>
<td>TFMA (41°50'42&quot;N, 141°06'56&quot;E)</td>
<td>Minamiite-alunite-quartz</td>
<td>Minamiite-alunite</td>
<td>2.88±0.09</td>
<td>17.7±6.2</td>
<td>1.58±0.56</td>
<td>96.0</td>
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<td>e (940619-06)</td>
<td>KAA (41°51'21&quot;N, 141°03'29&quot;E)</td>
<td>Alunite-quartz (Minamiite)</td>
<td>Alunite</td>
<td>4.30±0.09</td>
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<td>f (kb-109)</td>
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<td>1.85±0.06</td>
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<td>g (940619-07)</td>
<td>KAA (41°51'23&quot;N, 141°03'33&quot;E)</td>
<td>Alunite-quartz (zunyite-andalusite-geothite)</td>
<td>Alunite</td>
<td>4.88±0.10</td>
<td>54.2±10.7</td>
<td>2.86±0.57</td>
<td>92.5</td>
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Table 3 $^{34}$Sv-CDT values of alunite-group minerals and pyrite in KAA and TFMAA.

<table>
<thead>
<tr>
<th>No.</th>
<th>Sample No.</th>
<th>Advanced argilic alteration</th>
<th>Location (Elevation)</th>
<th>Sample analyzed</th>
<th>Mineral assemblage</th>
<th>Mineral analysed</th>
<th>$^{34}$Sv-CDT (pyrite-alunite)</th>
<th>Isotopic T (°C)</th>
<th>K-Ar age (Ma)</th>
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<tr>
<td>1</td>
<td>Furube A</td>
<td>TFMAA</td>
<td>Furube (100m)</td>
<td>Hydrothermal breccia of vuggy silica in an alunite matrix.</td>
<td>quartz, alunite</td>
<td>alunite</td>
<td>38.0</td>
<td>0.2</td>
<td></td>
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<tr>
<td>2</td>
<td>Furube B</td>
<td>TFMAA</td>
<td>Furube (100m)</td>
<td>Andesite of which phenocrysts are replaced with alunite.</td>
<td>quartz, alunite</td>
<td>alunite</td>
<td>31.2</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>kb-173</td>
<td>TFMAA</td>
<td>Takinosawa (250m)</td>
<td>Andesite of which phenocrysts are replaced with alteration minerals.</td>
<td>tridymite, kaolinite, alunite, quartz</td>
<td>alunite</td>
<td>30.6</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Tokiawakatsu</td>
<td>TFMAA</td>
<td>Tokiawakatsu (400m)</td>
<td>Andesite of which phenocrysts are replaced with alteration minerals.</td>
<td>quartz, minamiite, alunite</td>
<td>minamiite, alunite</td>
<td>15.3</td>
<td>15.2</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>kb-021</td>
<td>KAA</td>
<td>Kobui River (380m)</td>
<td>Andesite of which phenocrysts are replaced with alteration minerals. The alteration minerals also occur in vugs (&lt;15 mm in size).</td>
<td>quartz, pyrophyllite, alunite, kaolinite</td>
<td>alunite</td>
<td>27.1</td>
<td>0.8</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>kb-034</td>
<td>KAA</td>
<td>Kobui River (410m)</td>
<td>Hydrothermal breccias of vuggy silica and compact quartz with a matrix of alteration minerals.</td>
<td>quartz, minamiite, zunyite, topaz</td>
<td>minamiite</td>
<td>30.4</td>
<td>2.7</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>kb-059</td>
<td>KAA</td>
<td>Kobui-kita (410m)</td>
<td>Andesite of which phenocrysts are replaced with alunite.</td>
<td>alunite-natrolalunite s. s.</td>
<td>alunite (60%)-natrolalunite (40%) s. s.</td>
<td>26.6</td>
<td>0.8</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>kb-074</td>
<td>KAA</td>
<td>Kobui-kita (520m)</td>
<td>Silicified andesite with pods of alteration minerals.</td>
<td>quartz, pyrophyllite, alunite, kaolinite</td>
<td>alunite</td>
<td>24.8</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>kb-107</td>
<td>KAA</td>
<td>Shirai River (170m)</td>
<td>Andesite of which phenocrysts are replaced with minamiite.</td>
<td>quartz, minamiite, quartz</td>
<td>minamiite</td>
<td>25.4</td>
<td>21.0</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>kb-109</td>
<td>KAA</td>
<td>Shirai River (170m)</td>
<td>Pyrite-disseminated silicified andesite of which phenocrysts are replaced with minamiite and kaolinite. Minamiite veins are present.</td>
<td>quartz, minamiite, kaolinite, pyrite, rutile</td>
<td>minamiite (veinlet) pyrite</td>
<td>19.8</td>
<td>19.8</td>
<td>319</td>
</tr>
<tr>
<td>11</td>
<td>kb-110</td>
<td>KAA</td>
<td>Shirai River (170m)</td>
<td>Pyrite-disseminated silicified andesite of which phenocrysts are replaced with minamiite.</td>
<td>quartz, minamiite, pyrite, rutile</td>
<td>minamiite</td>
<td>23.5</td>
<td>23.5</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>kb-154</td>
<td>KAA</td>
<td>Kinoashi River (320m)</td>
<td>Pyrite-disseminated silicified andesite of which phenocrysts are replaced with natrolalunite-alunite.</td>
<td>quartz, alunite-natrolalunite, pyrite, rutile</td>
<td>natrolalunite(80%) alunite (20%) s. s. pyrite</td>
<td>20.1</td>
<td>291</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>kb-155</td>
<td>KAA</td>
<td>Shirai River (360m)</td>
<td>Andesite of which phenocrysts are replaced with alteration minerals. The alteration minerals also occur in vugs (&lt;20 mm).</td>
<td>quartz, minamiite, alunite, kaolinite</td>
<td>alunite (minamiite)</td>
<td>26.2</td>
<td>26.2</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>kb-156</td>
<td>KAA</td>
<td>Shirai River (360m)</td>
<td>Hydrothermal breccia of compact quartz in a minamiite-alunite matrix.</td>
<td>alunite, minamiite, quartz</td>
<td>alunite, minamiite</td>
<td>28.6</td>
<td>28.6</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>kb-222</td>
<td>KAA</td>
<td>Kobui (530m)</td>
<td>Hydrothermal breccia of acid altered andesite.</td>
<td>naclrite, alunite-natrolalunite s.s., quartz, pyrophyllite, zunyite</td>
<td>alunite (70%)-natrolalunite (30%) s.s.</td>
<td>25.4</td>
<td>25.4</td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>kb-226</td>
<td>KAA</td>
<td>Kobui (560m)</td>
<td>Pyroclastic rock (?) with acid alteration.</td>
<td>quartz, minamiite, alunite, rutile</td>
<td>minamiite (alunite)</td>
<td>29.3</td>
<td>29.3</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>kb-326</td>
<td>KAA</td>
<td>Ponkinaishi River (310m)</td>
<td>Pyrite-disseminated andesite of which phenocrysts are replaced with alteration minerals.</td>
<td>quartz, minamiite, dickite, pyrite</td>
<td>minamiite</td>
<td>19.9</td>
<td>0.1</td>
<td>306</td>
</tr>
</tbody>
</table>

s.s.: solid solution
Isotopic temperature is calculated, following to Ohmoto and Lasaga (1982).
The younger age of the Maruyama Lavas than those of the alunites is concordant with the fact that the lavas are not affected by advanced argillic alteration. The different ages of the alunites (0.23±0.08 Ma and 0.69±0.26 Ma) near to the Furube crater are ascribed to the different stages of the alteration; an infiltrative advanced argillic alteration at ca. 0.7 Ma was followed by hydrothermal brecciation, which fragmented the advanced argillic rocks, at ca. 0.2 Ma. In spite of the close proximity of the alunite-sample localities in KAA, the age obtained (1.80±0.06 Ma and 2.92±0.31 Ma) are significantly different. These ages may be indicative of different pulses of hydrothermal activity, which is reflected in higher (alunite-zunyite-andalusite) and lower temperature (alunite-minamiite) mineral assemblages. These alunite ages suggest that KAA and TFAA formed from 2.9 Ma to 1.8 Ma and from 1.3 Ma to 0.2 Ma, respectively.

5. Sulfur Isotopes

Sulfur isotopes were measured on alunite-group minerals and coexisting pyrite, following to the methods described by Sasaki et al. (1979). The results (Table 3) indicate that all of the analyzed alunite-group minerals, including the samples used for the K-Ar dating, are greater than 15 ‰ in δ34S_CDT. This result suggests that the alunite-group minerals are not the product of supergene or steam-heated environment, but were formed in a magmatic hydrothermal environment (Rye et al., 1992). There is no clear difference in δ34S_CDT values among alunite, natroalunite and minamiite (Table 3).

Alunite-group minerals from TFAA have δ34S_CDT values greater than 30 ‰, with the exception of a minamiite sample from the Tokiwamatsu subarea, whereas those from KAA are less than 31 ‰ (Table 3). In KAA, relatively low δ34S_CDT values (approximately 20 ‰) were obtained from alunite-group minerals in the root zones of hydrothermal breccia that occur in the topographic lows (at heights below approximately 320 m above sea level), whereas relatively high values (25-31 ‰) at elevation above 320 m (Table 3). In TFAA, the δ34S_CDT values for the alunite-group minerals increase towards the north-northeast from the Tokiwamatsu to Furube subareas. A variation in the δ34S_CDT value is also recognized within a sample as well as within a locality. At Locality KB-150 (Fig. 4), one of the root zones to KAA, a 10 m-wide hydrothermal breccia is surrounded by smectite alteration. This root zone includes pyrite-rich quartz-minamiite fragments within relatively pyrite-poor quartz-minamiite rocks (Fig. 11). This occurrence indicates that the pyrite-rich quartz-minamiite rock was fragmented by a later brecciation associated with minamiite-quartz deposition. The samples of the pyrite-rich fragments (kb-109 and kb-110) have δ34S_CDT values of 20.2 ‰ and 23.9 ‰, respectively.

Sample kb-109 is intruded by later minamiite veinlets with a δ34S_CDT value of 27.0 ‰. Sample kb-107 from the pyrite-poor matrix part has a δ34S_CDT value of 25.8 ‰. This suggests that the δ34S_CDT value of the early-stage minamiite is smaller than that of the late-stage one at this locality.

Isotopic equilibrium temperatures of coexisting alunite-group minerals and pyrite from these hydrothermal breccias in KAA are between 290°C to 320°C (Table 3).

6. Discussion

6.1 Hydrothermal fluids in KAA and TFAA

The characteristics of KAA and TFAA are summarized in Table 4. The hydrothermal alteration occurred mainly within the subaerial Pliocene Kobui volcano(es). The mineral assemblage and alteration zones are quite similar to those of high-sulfidation epithermal gold deposits (Hedenquist, 1987; Arribas, 1995), although the gold concentration in the silicified rocks in KAA is not high enough for economic mining. Advanced argillic alteration is speculated to form above subvolcanic intrusions (Sillitoe, 1991; Hedenquist and Lowenstern, 1994), although no porphyritic intrusion of the same age is recognized at the surface. The lateral zonation from alunite-bearing zones (pyrophyllite-diaspore-alunite, alunite-minamiite and alunite zones) to smectite zone through dickite-kaolinite zone in KAA and TFAA is explained by reaction of the wall rock with hydrothermal fluids of decreasing acidity (Arribas, 1995).

Kaolinite and its higher temperature polymorph, dickite and nacrite, are common in KAA, whereas dickite and nacrite are restricted in the southern sub-area in TFAA. In the acid zone of active Philippine geothermal systems, kaolinite has a maximum occurrence temperature of about 200°C, whereas dickite is stable from 120°C to as high as 280°C (Reyes, 1990, 1991). Reyes (1990, 1991) also notes that the lowest temperature for pyrophyllite formation is about 200°C; this allows pyrophyllite to be used to identify the low-temperature limit.
Y. WATANABE, M. AOKI and K. YAMAMOTO

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Table 4 Characteristics of KAA and TFAA.

<table>
<thead>
<tr>
<th></th>
<th>KAA</th>
<th>TFAA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Age</td>
<td>2.9-1.8 Ma</td>
<td>1.3-0.2 Ma</td>
</tr>
<tr>
<td>Area</td>
<td>4.1 km²</td>
<td>1.2 km²</td>
</tr>
<tr>
<td>Distribution</td>
<td>continuous</td>
<td>separate</td>
</tr>
<tr>
<td>Associated explosion crater</td>
<td>Yagi, Kobui</td>
<td>Tokiwamatsu, Takinosawa, Furube</td>
</tr>
<tr>
<td>Strike of explosion breccia</td>
<td>N50°W, N70°E</td>
<td>N5-20°E</td>
</tr>
<tr>
<td>Type of advanced argillic rocks</td>
<td>massively altered andesite, hydrothermal breccia, vuggy silica and massively silicified rock</td>
<td>massively altered andesite, hydrothermal breccia and vuggy silica</td>
</tr>
<tr>
<td>Alteration mineral zone</td>
<td>pyrophyllite-diaspore-alunite zone, alunite-minamitite zone, alunite zone, dickite-kaolinite zone, kaolinite zone, smectite zone</td>
<td>alunite-minamitite zone, alunite zone, dickite-kaolinite zone, kaolinite zone, smectite zone</td>
</tr>
<tr>
<td>Alteration minerals</td>
<td>pyrophyllite, diaspore, zunyite, topaz, minamitite, natroalunite, alunite, nacrite, dickite, kaolinite, pyrite, quartz, (native sulfur, andalusite, sanvergite, woodhouseite)</td>
<td>minamitite, natroalunite, alunite, kaolinite, (nacrite, dickite), pyrite, quartz, (cristobalite, tridymite, native sulfur, orpiment)</td>
</tr>
<tr>
<td>$\delta^{34}$S&lt;sub&gt;CDT&lt;/sub&gt; of alunite-group minerals</td>
<td>20-30 ‰</td>
<td>15-38 ‰ (mostly 30-38‰)</td>
</tr>
<tr>
<td>Sulfur isotopic temperature</td>
<td>−320° C</td>
<td>−200° C</td>
</tr>
<tr>
<td>Estimated temperature</td>
<td>−320° C</td>
<td>−200° C</td>
</tr>
</tbody>
</table>

of a dickite-pyrophyllite assemblage, whereas the dickite-kaolinite assemblage is stable at lower temperatures. Experimental results suggest pyrophyllite and diaspore should not coexist at temperatures less than 280°C (HEMLEY et al., 1980) and zunyite is stable up to 450°C where it is replaced by topaz at higher temperatures (HSU, 1986). On the other hand, REYES (1990, 1991) observed zunyite, topaz and andalusite at measured temperatures of 250°-300°C, 280°-300°C and >260°C, respectively, and cristobalite and tridymite in the active Philippine geothermal systems. Based on these constraints, the temperature of the hydrothermal fluids which formed the pyrophyllite-diaspore-alunite zone in KAA is estimated to be higher than 200°C, because dickite (or nacrite)-pyrophyllite assemblage is common in the zone and kaolinite-pyrophyllite assemblage occurs only at the margin of the zone. The temperature may have exceeded 280°C in the Yagi crater where it is rich in topaz and zunyite, and pyrophyllite coexists with diaspore. In the root zone of the advanced argillic alteration, the temperature of the hydrothermal fluids responsible for the precipitation of alunite-group minerals is estimated to be 290°C to 320°C, based on the sulfur isotopic equilibrium. Homogenization temperatures of fluid inclusions in quartz veinlets within a topaz-bearing advanced argillic rock in KAA are 260°C to 350°C with a mode of 320°C (MITI, 1995). These results are roughly compatible with the estimated temperature.

In contrast, the temperature of the hydrothermal fluids in TFAA is estimated to have been lower than 200°C, because pyrophyllite, diaspore, zunyite and topaz are absent in TFAA. In particular, a lack of dickite and nacrite, and the presence of cristobalite and tridymite in the Takinosawa and Furube subareas suggest that the temperature during the alteration may have not exceeded 160°C locally.

If the hydrothermal fluids were in a closed system with respect to sulfur during the alteration and isotope equilibrium was maintained between sulfides and sulfates, the sulfur isotopic values of sulfides and sulfates are expected to decrease and increase, respectively, as the temperature of the fluids decreases. Although alunite-group minerals coexisting with sulfides were not found in most cases, the relatively lower and higher values in $\delta^{34}$S<sub>VDPT</sub> of the alunites in KAA and TFAA are consistent with the higher and lower temperatures estimated from the mineral assemblages.

Alunite-group minerals and their P-bearing analogues are common in advanced argillic alteration (ARRIBAS, 1995). Among these minerals, crandallite, woodhouseite and svanbergite form when the early acid fluids dissolve primary apatite (STOFFREGEN and ALPERS, 1987), releasing P and REE, where Sr and Ba are likely derived from plagioclase and alkali feldspar. Once all apatite dissolved, these phosphate-sulfate minerals stop forming, and even dissolve prior to precipitation of alunite and natroalunite (HEDENQUIST et al., 1994). Alunite-group minerals in the advanced argillic alteration are formed by acid and relatively oxidizing fluids, resulted from disproportionation of magmatic SO₂ at temperatures < 400°C to form H₂SO₄ and H₂S (SAKAI and MATSUBAYA, 1977) with progressive dissociation of both H₂SO₄ and HCl as the temperature decreased (HEDENQUIST, 1995; ARRIBAS et al., 1995). Alunite-group minerals in KAA include partially dissolved woodhouseite and svanbergite, as well as euhedral zunyite (Figs. 8a and b). This suggests that...
the hydrothermal fluids in KAA increased acidity and decreased temperature with time. The coexistence of alunite and pyrite at the root zone at temperature of 290-320°C indicates that the solutions followed to the pyrite-alunite equilibrium line in the redox potential: $R_H = \log (f_{H_2}/f_{H_2O})$-temperature diagram (Fig. 12). The solution may have decreased $R_H$ (oxidized) with time, because the pyrite-rich rock was brecciated by the later hydrothermal activity that precipitated mainly alunite-group minerals. The solution was also oxidized at the upper part of the KAA, because the pyrite/alunite volume ratio decreases (Plate 2a) and the pyrite exhibits a severely dissolved shape (Plate 2c). The occurrence of massively silicified rocks, which are characterized by the association of euhedral pyrite with granular quartz and the absence of alunite and anhydrite, suggests that the fluids were reduced at the latest stage of the hydrothermal activity (Fig. 12).

Sulfur dissemination in the crater-filling lacustrine sedimentary rocks commonly occurs by a vapor phase activity relatively rich in H$_2$S, separated from the liquid phase at the water table (White, 1967). Near the surface the H$_2$S oxidizes to form native sulfur and sulfuric acid. Leaching by sulfuric acid above and near the water table produces a characteristic suite of minerals that includes amorphous opal, cristobalite, anatase, kaolinite, alunite, and a variety of soluble sulfates, with sparse fine-grained cinnabar (White, 1967). The sulfur dissemination and mineral assemblages of the kaolinite zone in the lacustrine sedimentary rocks is ascribed to H$_2$S-dominant vapor-phase fluids.

6.2 Tectonic control on the hydrothermal systems
Flow path of a hydrothermal fluid is influenced by hydraulic gradient, by existing permeability anisotropy arising from different lithology, bedding and other forms of crustal layering, and by structural permeability developed under the prevailing stress field (Sibson, 1995). The linear structures of the root zones of hydrothermal breccia in KAA, and the linear alignment of the advanced argillic subareas with hydrothermal breccias subparallel to the alignment in TFAA suggest that structural permeability was most responsible for the fluid paths, whereas lateral flow of hydrothermal solutions is also expected for the upper part of KAA. Watanabe (1993a, b, 1995) suggested that a regional horizontal compressive stress in the upper crust of southwestern Hokkaido rotated rapidly in orientation from N70°W to N30°W at approximately 1.5-1.2 Ma, based on the analyses of igneous dikes, alignments of volcanic craters and conjugate sets of strike-slip faults. According to this change, E-W right-lateral and NW-SE left-lateral strike-slip faults are expected before the rapid rotation, and NW-SE right-lateral and N-S left-lateral faults after the rotation (Fig. 13). The strikes and ages of the hydrothermal breccias in the Kobui area are N50°W (2.9 Ma) and N70°E (2.7 Ma) in KAA, and N5°E (1.3 Ma) and N20°E (0.2 Ma) in TFAA. The variable orientations of the hydrothermal breccias are, therefore, interpreted to be due to the rotation of the regional horizontal compressive stress between 1.5 and 1.2 Ma. The hydrothermal breccias in KAA formed before the rapid rotation of the
6. 3 Evolution of the hydrothermal systems

The evolution of the hydrothermal systems in the Kobui area is illustrated in Figure 14. During the Pliocene, andesitic magmas intruded and extruded in the Kobui area and formed "Kobui volcano or volcanoes". At the latest stage of the magmatic intrusions (ca. 2.9 Ma), the crystallizing magma discharged hydrothermal solutions, which exolved vapor rich in magmatic volatiles such as SO$_2$, H$_2$S, CO$_2$, HCl and HF. The vapor was absorbed by circulating meteoric water, forming acid fluid due to dissociation of HCl and disproportionation of SO$_2$ to form H$_2$SO$_4$ and H$_2$S at temperatures below approximately 400°C. The acid fluids rose through the strike-slip fractures and reacted with the previously erupted Kobui andesite lavas to form KAA (Fig. 14a). The injection of the acid fluids occurred repeatedly over a million years, and the fluids increased acidity and decreased temperature with time. At the waning stage of this hydrothermal activity, abundant meteoric water may have mixed with the magma flux and formed more reduced fluids, which precipitated silica with a small amount of pyrite and traces of gold within the previously acid-leached rocks. After the completion of KAA, an upper portion of the "Kobui volcano" was eroded away and large explosions formed craters (Kobui and Yagi), destroying KAA. Lacustrine sediments were deposited within these craters, often disseminated with sulfur, arsenic, antimony and mercury which were carried by more neutralized and reduced, H$_2$S dominant hydrothermal fluids with lower temperatures (Fig. 14b). This change in the chemical characteristics of the hydrothermal solutions might have been caused by a change in the hydrology, as the craters could have provided a loci for abundant meteoric water inflow. A lack of the sulfur disseminated lacustrine sediments in the Yagi crater may be due to deeper erosion than in the other craters. This is inferred from the abundant occurrence of relatively...
high temperature minerals such as topaz and zunyite
within the crater and the oldest K-Ar age for the alunite
(2.9 Ma) near to the crater.

Since ca. 1.3 Ma, acid fluids have risen at the east of
KAA through NNE-SSW striking fractures and formed
TFAA (Fig. 14b). The earliest alteration occurred in the
Tokiwamatsu subarea, followed by the erosion of an up-
per portion of the alteration zone. In this subarea, vapor-
rich acid fluids may have been dominant, because
hydrothermal breccia and vuggy silica have a wide dis-
tribution and silica precipitation overprinting the vuggy
silica is not recognized. This advanced argillic alter-
ation was also followed by a crater-forming explosion
(Tokiwamatsu crater), and lacustrine sediments accu-
mulated within the crater. More recently (0.7-0.2 Ma),
advanced argillic alteration and successive eruptions
occurred at the Takinawaso and Furube subareas along
the NNE-SSW fracture. These areas of advanced argillic
alteration have been only slightly eroded and still pre-
serves the low temperature (<160°C) mineral assemblage
at the surface. Within these craters, H2S-dominant
hydrothermal fluids disseminated sulfur and mercury
in the lacustrine sediments.

Thus, the hydrothermal activities in the Kobui area
began with advanced argillic alteration followed by a
crater-forming explosion, and ended with sulfur (plus
arsenic, antimony and mercury) dissemination in the
crater-filling lacustrine sediments. This style is similar
to that of the Osorezan lithocap, where advanced argillic
alteration at 0.6 Ma was followed by crater-lake forma-
tion at 0.2 Ma (Aoki, 1991). The present hydrothermal
activity near to the Osore crater lake is characterized by
a H2S-dominant, neutral pH solution, which precipitates
metal-rich clay, including mercury, arsenic, antimony
and tellurium at the surface, as well as gold in the sub-
surface (Aoki, 1992). Thus, low-sulfidation epithermal
gold mineralization might also be expected beneath the
crater-fill sediments in the Kobui area (Fig. 14b).

Advanced argillic alteration and sulfur precipitation
is presently active in the crater of Esan volcano, 6 km
southeast from the Kobui area. The loci of advanced
argillic alteration and sulfur precipitation has shifted
eastward since 2.9 Ma.

7. Conclusions

1) The advanced argillic alteration in the Kobui area
is hosted by the Pliocene Kobui andesite lavas and is
divided into Kobui (KAA) and Tokiwamatsu-Furube
advanced argillic alteration areas (TFAA). KAA formed
during the period from 2.9 to 1.8 Ma and TFAA from 1.3
to 0.2 Ma.

2) The alteration is divided into pyrophyllite-dias-
pore-alunite, alunite-minamitten, alunite, dickite-kaolinite
and smectite zones from inner to outer alteration zones.

KAA is characterized by the presence of pyrophyllite,
diaspore, topaz and zunyite, whereas TFAA is lacking in
these minerals, but contains tridymite and cristob-
alite. The mineral assemblage and sequence and sulfur
isotopic temperatures suggest that the temperature of
the hydrothermal fluids during the alteration in KAA
was higher (200°C-320°C) than in TFAA (~200°C).

3) Hydrothermal solutions rose through conjugate
strike-slip fractures and diffused near the surface. The
fractures changed strike with time, related to the rotation
of the regional horizontal compressive stress exerted in
southwestern Hokkaido during the Pliocene and Pleis-
tocene.

4) The hydrothermal activity in the Kobui area began
with acidic and higher-temperature fluids which formed
the advanced argillic alteration, followed by crater-
forming eruptions, and ending with sulfur-dissemina-
tion in crater-filling lacustrine sediments by H2S domi-
nant, more reduced solutions. The loci of the activity
has shifted to the east since 2.9 Ma.

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Appendix 1 A comparison of the stratigraphic units reported by SHOYA and TAKAHASHI (1967) and this study.

<table>
<thead>
<tr>
<th>Age</th>
<th>Formation name</th>
<th>Lithology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Holocene</td>
<td>Talus deposits(^1)</td>
<td>gravel, sand, volcanic ash and clay</td>
</tr>
<tr>
<td>Pleistocene</td>
<td>Maruyama Volcanic Rocks(^2)</td>
<td>andesitic lava and tuff breccia</td>
</tr>
<tr>
<td>Miocene</td>
<td>Kinaoshi Formation(^3)</td>
<td>volcanic breccia and tuff breccia, intercalating andesite lavas</td>
</tr>
<tr>
<td></td>
<td>Shiodomarigawa Formation(^4)</td>
<td>siltstone, mudstone, tuff and tuff breccia</td>
</tr>
<tr>
<td></td>
<td>Kakkumi Formation(^5)</td>
<td>unstratified massive tuff</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1: "Talus deposits" (Shoya and Takahashi, 1967) mainly consists of alternating beds of lacustrine mudstone, siltstone and sandstone, which underlie the Pleistocene Maruyama Lavas and overlie the Pliocene Kobui Lava.

2: The Pleistocene "Maruyama volcanic Rocks" (Shoya and Takahashi, 1967) is divided into the Pleistocene Maruyama and Futago Lavas, based on K-Ar dating. The age of the Kobui Lava is older than 2.9 Ma, because allunites in the advanced argillic alteration in the lavas are dated at 1.8 Ma, 2.7 Ma and 2.9 Ma (Table 2). The K-Ar age of the lower unit of the Maruyama Lavas (0.1 Ma, Table 2) suggests Maruyama Lavas are Late Pleistocene in age.

3: Dacitic porphyries are included within the Kinaoshi Formation by Shoya and Takahashi (1967), whereas no extrusive facies of the dacite is recognized in the Kobui area.

4: Shoya and Takahashi (1967) divided the Miocene series into the Kakkumi, Shiodomarigawa and Kinaoshi Formations in ascending order. However, it is difficult to apply the division of Shoya and Takahashi (1967) for the Kobui area, because the Miocene series are composed of alternation of andesitic lava, tuff breccia, tuff, siltstone and mudstone. This paper uses the name "Shiodomarigawa Formation" for all the Middle to Late Miocene strata in the Kobui area.
Plate 1  
a) Maruyama volcano (upper unit of the Maruyama Lavas). There is an explosion crater on the northwest of Mt. Maruyama (left side of the Mt. Maruyama). 
b) Dike-like hydrothermal breccia in the Tokiwamatsu subarea in TFAA. The hydrothermal breccia consists of completely acid-leached andesite. 
c) Massively altered andesite at a root zone of KAA. The phenocrysts of the andesite are replaced by minamiite and alunite, and the groundmass by quartz and pyrite (kb-155). 
d) Hydrothermal breccia in KAA. Breccias of silicified and acid-leached andesite occur in a matrix of minamiite and alunite (kb-051). 
e) Vuggy silica in the Tokiwamatsu subarea in TFAA (kb-176). Only quartz and anatase are detected by XRD. 
f) Massively silicified rock composed mainly of quartz with minor pyrite in the KAA (kb-082).
Plate 2 Photomicrographs of altered and silicified rocks in the KAA. Abbreviations: Al: alunite-group minerals, Py: pyrite, Qz: quartz, V: vug. a) Vuggy altered andesite composed of quartz, alunite-group minerals and pyrite from the upper portion of KAA. b) Altered andesite composed of quartz, pyrite and alunite-group minerals from the upper portion of KAA. Phenocrysts of the andesite is completely replaced by alunite-group minerals and pyrite. c) Co-occurrence of pyrite and alunite in altered andesite from the upper portion of KAA. Pyrites show an anhedral, colloform shape. d) Massively silicified rock composed of granular, anhedral quartz. e) Pyrite-rich quartz-minamiite rock at a root zone of KAA. f) Pyrite-rich quartz-minamiite rock at a root zone of KAA. Minamiite and pyrite occur in vugs.