Provenance identification based on EPMA analyses of heavy minerals: Case study of the Toki Sand and Gravel Formation, central Japan

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Abstract
To perform efficient provenance analysis, a method for quick identification of heavy minerals by using an electron probe microanalyzer (EPMA) was developed. The concentrations of 28 elements were measured in individual grains to identify the mineral species based on their chemical composition. Provenance rocks can be identified based on the abundance ratio of identified minerals and the chemical composition of specific minerals. This method was applied to samples from the Toki Sand and Gravel Formation in the Tono area, central Japan. At the sampling location, gravels of the Naegi-Agematsu Granite and the Nohi Rhyolite were observed in the upper member, whereas gravels of only the Nohi Rhyolite were observed in the lower member. Heavy-mineral compositions of the bedrock samples of the Nohi Rhyolite and the Naegi-Agematsu Granite, as well as the sediments at the sampling location, were measured by EPMA analysis. Both bedrocks commonly contain ilmenite, biotite, allanite, and zircon, and the abundance ratios of heavy minerals were similar. Instead, MnO contents of ilmenite grains and Y2O3 contents of zircon grains were used as indexes to distinguish the two bedrocks. As a result, the Y2O3-rich zircon grains (≥1 wt%) observed in the Naegi-Agematsu Granite were obtained in the upper member but not in the lower member. The MnO contents of ilmenite grains were higher in the Naegi-Agematsu Granite and the upper-member sediments than those in the Nohi Rhyolite and the lower-member sediments, although the difference was small probably because of Mn dissolution due to secondary alteration. These results indicate that the Naegi-Agematsu Granite was added to the provenance of the sampling location after deposition of the lower member, which is consistent with the geological description. The change in provenance could have been caused by uplift and denudation of the Atera Mountains associated with reactivation of the nearby Atera Fault.

KEYWORDS
Atera Fault, EPMA, heavy mineral, mineral identification, mineralogical composition, provenance analysis

1 | INTRODUCTION
Assessment of uplift and the associated denudation of mountains is significant from the viewpoint of geological disposal of radioactive wastes. One of the reasons is that uplift and denudation can cause exposure of repositories (McKinley & Chapman, 2009). Furthermore, uplift and denudation can cause nuclide migration by affecting the hydraulic gradient and the groundwater flow paths at depths of several hundred meters below ground level (e.g. Onoe, Sasao, Saegusa, & Kosaka, 2009).
Uplift occurs in response to tectonic and non-tectonic processes (e.g. glacial unloading, igneous intrusion). Along active plate boundaries, following uplifts resulting from tectonic processes, regional-scale (hundreds to thousands of kilometers) uplift is caused by the convergence of tectonic plates via mechanisms such as collision and subduction, and local-scale (kilometer) uplift is generally caused by faulting and folding (Litchfield, Ota, & Merritts, 2009). Generally, mountain formation occurs over several hundred thousand years or longer, even in regions with active plate convergence such as the Japanese Islands, where the general uplift rate in the Japanese Islands ranges from < 0.1 mm/year to 1 mm/year (Committee for Geosphere Stability Research, 2011). However, more than a hundred thousand years are required until the radioactivity of wastes becomes equivalent to that of a natural uranium ore. Therefore, relatively long-term geological events, such as uplift and erosion of a mountain, should be considered and their effects should be evaluated.

Provenance analysis based on sedimentological, petrological, mineralogical, geochemical, and geochronological approaches in the downstream area is often helpful for understanding onset times and uplift processes, as well as the associated denudation of mountains in the upstream area (e.g. Nichols, Williams, & Paola, 2007). Conventional provenance analysis is based mainly on gravel composition and paleocurrents reconstructed using sedimentary structures. Moreover, modal analysis is helpful for revealing the provenance of sandy sediments and sandstone, as well as gravel composition (Zuffa, 1985). However, mineral composition in downstream sediments can change from that of the host rock upstream because several types of minerals can be dissolved by weathering and because specific minerals can be concentrated owing to density differences (Morton & Hallsworth, 1999). In contrast, discrimination of heavy minerals based on chemical features can be useful because several heavy minerals, such as zircon and Fe–Ti oxides, are resistant to weathering. Moreover, heavy minerals could be concentrated, but their chemical compositions have the potential to serve as indexes for provenance analysis. Many studies have revealed provenance based on the chemical compositions of heavy minerals, such as chromium spinel (Arai & Hisada, 1991; Hisada, Arai, & Lee, 1999; Hisada, Takashima, Arai, & Lee, 2008) and garnet (Takeuchi, 1994). However, the methods used in these studies are inconvenient for the following three reasons: (i) specific skills for mineral identification are needed; (ii) the methods are applicable to limited types of rocks, including target minerals; and (iii) extraction and concentration of target minerals are time-consuming when large quantities of samples are employed.

Recently, tremendous advances have been made in quantitative elemental analysis of micro domains by using techniques such as electron probe microanalysis (EPMA), secondary ion mass spectrometry, and laser ablation inductively coupled plasma mass spectrometry (LA-ICP–MS) (e.g. Hirata et al., 2004; Kilburn & Wacey, 2014; McGee & Keil, 2001). These techniques have made it possible to perform in-depth chemical analyses of individual mineral grains in sediments. The classification of chemical features by multiple single-grain analyses of heavy minerals resistant to weathering has great potential for wide-range application to provenance analysis.

In the present study, rapid analyses of a multitude of heavy minerals are performed using a field-emission electron probe microanalyzer (FE-EPMA) to reveal provenance changes. These analyses, proposed by M. Shimizu, Sano, and Suzuki (2017), are independent of the observational ability for mineral identification, free of restrictions on rock types, and they shorten treating time. A case study is conducted on the Toki Sand and Gravel Formation distributed in Tono area, Gifu prefecture, central Japan. Spatial distribution of sediments is constrained by the process of formation of mountains, which are the sources of the sediments. In cases where faulting could have led to mountain formation, revealing the source of sediments can help reveal the history of faulting and the associated uplift. We discuss the relationship among the characteristics of the heavy minerals in the sediments with uplift controlled by faulting, as well as the usefulness of rapid analyses using EPMA for provenance analysis.

## 2 | GEOLOGICAL SETTING

### 2.1 | Overview

The Toki Sand and Gravel Formation comprises Pliocene to lower-Pleistocene terrestrial sediments (Editorial Committee of CHUBU II, 1988). These sediments are distributed in the southeastern part of Gifu prefecture on the western side of the Atera Fault (Figure 1a,b). The general thickness of these sediments ranges from several tens of meters to several hundreds of meters. The gravels consist mainly of felsic volcanic clastics of the Nohi Rhyolite of the Late Cretaceous, chert, and metamorphosed clastic rocks derived from a Jurassic accretionary complex in the Mino Belt, as well as granite and granite phryry (Editorial Committee of CHUBU II, 1988).

The approximately 35 km-long Atera Fault strikes northwest-southeast, and it is subvertical with left-lateral strike-slip displacement and northeastern uplift (Sugimura & Matsuda, 1965; Tsukuda et al., 1993). The fault has evolved through reactivation of ancient shear zones under east-west compression during the Quaternary (Niwa, Mizuochi, & Tanase, 2009; Sugimura & Matsuda, 1965). The total horizontal displacement along the central part of the fault has been estimated to be 6–7 km based on the displacement of a granite porphyry dike swarm (N. Yamada, 1981). The Atera Mountains, northeast of the Atera Fault, are 600–1 200 m higher than the Mino Plateau southwest of the fault (N. Yamada, 1981). The Atera Fault separates the Gero Ash-Flow Sheet of the Nohi Rhyolite from the fine-to-coarse biotite granite of the Naegi-Agematu Granite in the study area (Figure 1a,b).

We performed sediment sampling in the stone mine in Nakatsugawa City (Figure 1a). The Toki Sand and Gravel Formation overlies the welded tuff of the Nohi Rhyolite at this location. Basement rocks in and around the location consist of welded tuff of the Gero Ash-Flow Sheet of the Nohi Rhyolite, granite of the Naegi-Agematu Granite, and basalt of the Ueno basaltic rocks (Figure 1a, in dashed circles).

### 2.2 | Basement rock: Nohi Rhyolite

The Gero Ash-Flow Sheet is part of the Late Cretaceous Nohi Rhyolite composite volcanism in central Japan (N. Yamada et al., 2005), and its Rb-Sr whole-rock isochron age is (75.2 ±6.2) Ma (1σ; Shirahase, 2005; N. Yamada & Koido, 2005). The Nohi Rhyolite sample used in this study was obtained from the basement rock in the stone mine (Figure 1a).
FIGURE 1  Geological setting of studied area. (a) Geological map of areas in and around the sampled location. Location of the study area is shown in the upper left part of the figure. (b) The cross section along the NEN–SWS line in the central part of (a). The line is shown in (a) with a red line near the center of the geological map. (c) Columnar section of the sampled location of the sediments and Nohi Rhyolite
At the microscopic scale, welded tuff of the Nohi Rhyolite is phenocryst-rich and is composed of quartz, plagioclase, alkali-feldspar, and biotite phenocrysts in a glassy groundmass accompanied by quartz, chlorite, and biotite (Figure 2a). Minor amounts of orthopyroxene, clinopyroxene, hornblende, and olivine are included, although most of them are altered severely. The Nohi Rhyolite at the sampling locations experienced contact metamorphism from the Naegi-Agematsu Granite (N. Yamada & Akahane, 2005). Glomeroporphyritic-colored minerals are recrystallized biotite showing a decussate texture because of contact metamorphism from the Naegi-Agematsu Granite (N. Yamada et al., 2005). Most of the plagioclase is highly sericitized. Representative accessory minerals are allanite, ilmenite (Figure 2b), and zircon (Figure 2c). Clay minerals and calcite occur as secondary minerals owing to groundmass alteration.

2.3 | Basement rock: Naegi-Agematsu Granite

The Naegi-Agematsu Granite, which originally intrudes the Gero Ash-Flow Sheet, has a CHIME monazite age of \(67.2 \pm 3.2\) Ma (2σ; Suzuki et al., 1994). In this study, Naegi-Agematsu Granite samples were obtained from the two locations shown in Figure 1a. The sample from location 2 (denoted by '2 in black triangle' in Figure 1a) was used only for collecting additional ilmenite and zircon grains. The Naegi-Agematsu Granite is composed mainly of anhedral quartz, subhedral alkali-feldspar, euhedral plagioclase, and euhedral biotite (Figure 2d). Principal accessory minerals are ilmenite (Figure 2e), zircon (Figure 2f), and allanite. The sample was free of titanomagnetite, and thus, it belongs to the ilmenite series (Ishihara, 1977).
2.4 | Basement rock: Ueno basaltic rocks

The Ueno basaltic rocks are extrusive rocks from the late Pliocene to the early Pleistocene monogenetic volcano group. The basaltic mass nearest to the stone mine (Sakashita mass: Nakano, 1993) overlies the Toki Sand and Gravel Formation. Equivalent rock masses of the Ueno basaltic rocks are exposed in the Kiso province (Nakano, 1994; Figure 1a), which is approximately 20 km south of the Ontake volcano. The whole-rock K-Ar ages of basalt from the Sakashita mass are 1.7–1.4 Ma (S. Shimizu, Yamazaki, & Itaya, 1988; Ujike, Iizuka, & Nakano, 1992; Uto & Yamada, 1985), whereas the whole-rock K-Ar ages of basalts from the Kiso province are 2.8–2.1 Ma (Nakano, Uto, & Uchiumi, 2000; Ujike et al., 1992), except for one sample, which had an age of (1.54 ±0.02) Ma (Nakano et al., 2000). The basaltic rock consists of euhedral phenocrysts, which are composed mainly of olivine, augite, and plagioclase (Nakano, 1994). Minor amounts of micro-phenocrysts of Fe-Ti-oxide (titanomagnetite and ilmenite) are present as well (Nakano, 1994). The groundmass consists mainly of plagioclase, augite, and Fe-Ti-oxide, which exhibit various textures ranging from coarse-grained intergranular to fine-grained intersertal (Nakano, 1994).

2.5 | Sediment samples and sampling location

The Toki Sand and Gravel Formation in the stone mine is subdivided into lower and upper members (Figure 1c). The lower member consists mainly of gravel and sand beds. The gravel beds are composed of pebbles and cobbles, which are welded tuff of the Nohi Rhyolite, in a matrix of consolidated coarse sand. The gravels are rounded-to-subrounded, poorly sorted, and less weathered. The lowest part of the gravel bed overlies the basement rock of the welded tuff of the Nohi Rhyolite. The upper part of the basement rock is weathered severely and characterized by well-developed weathering rinds. Therefore, the boundary between the gravel bed and the basement rock is unclear. The gravel beds include lenticular intercalations of pale gray coaly silt (F-5). In addition, F-2, F-7, and F-8 can be found in the layers in this lower member, as shown in Figure 1c.

A gravel-bearing silt bed overlies the gravel bed in the upper member. The gravel bed in the upper member is composed of pebbles and cobbles, which are welded tuff of the Nohi Rhyolite, in a sandy matrix, in minor association with cobbles of basaltic rocks and granite. The pebbles and cobbles are subrounded-to-subangular, poorly sorted, and moderately weathered. The gravel-bearing silt bed is slightly viscous, and it mainly contains quartz and feldspar fragments. The uppermost part of the gravel-bearing silt bed is weathered severely and brownish in color. In this part, highly weathered cobbles, such as massive sand, are included. These cobbles are welded tuff and granite. F-10 to F-14 were obtained from the layers in this upper member, as shown in Figure 1c.

According to a previous study on gravel composition (Yasue et al., 2011), all (100 %) gravels in the lower member are from the Nohi Rhyolite. In the upper member, more than 90 % of the gravels are from the Nohi Rhyolite, and < 10 % of the gravels are composed of basalt (Yasue et al., 2011).

Paleocurrent analysis based on the imbrication structure of gravels, trough cross-bedding in sand beds, and anisotropy of magnetic susceptibility in silt beds indicates that the direction of the current is toward south-southwest or southwest in the upper member and toward east-southeast or southeast in the lower member (Agency for Natural Resources and Energy, 2015; Yasue et al., 2011). High concentrations of K2O (~ 5 wt%) were found in the glass inclusions in plagioclase grains from a sand bed in the lower member, suggesting that the grains could have been derived from Znp–Oht tephra (3.9 Ma; Kurokawa & Tomita, 1998; Tamura, Yamazaki, & Mizuno, 2005) or its correlatives (Yasue et al., 2011; Figure 1c).

3 | ANALYTICAL METHOD

The quick chemical quantification and mineral identification method proposed by M. Shimizu et al. (2017) was applied to the sediment and bedrock samples. Nine samples were collected from the sediment: F-2, F-5, F-7, and F-8 from the lower member, and F-10, F-11, F-12, F-13, and F-14 from the upper member (Figure 1c). The bedrock samples used in the analysis were the Nohi Rhyolite and the Naegi-Agematsu Granite (Figure 1a), and they were used to compare the mineral composition and chemical features of specific heavy minerals in the sediment samples.

All samples were crushed and sieved repeatedly (using the Mesh Cloth of 80 meshes; Nichika Inc.) to extract grains measuring < 250 μm. The grains were panned in water to concentrate the heavy minerals. The concentrated grains were dried, and heavy minerals with specific gravity of 2.7 or higher were separated using a heavy liquid (sodium polytungstate; SPT, SOMETU). The separated heavy minerals were dried and mounted linearly on a glass slide at intervals of approximately 2 mm by using Petropoxy 154 (Palouse Petro Products). The mounts were polished using diamond pads (#600, #1500); agate plates (#600, #1000, #2000, #4000, #6000); and then with 3 μm, 1 μm, and 0.25 μm diamond paste on a rotary polisher. The polished mounts were then covered with 15–20 nm-thick carbon in a vacuum evaporator.

The chemical compositions of the mineral grains on the glass slides were determined quantitatively by using a JEOL JXA-8530F FE-EPMA at the Tono Geoscience Center, Japan Atomic Energy Agency, without prescreening. Twenty-eight elements (Fe, Na, Mg, Al, Si, P, S, K, Ca, Ti, Cr, Mn, Fe, Y, Zr, Nb, La, Ce, Pr, Nd, Sm, Dy, Ho, Er, Yb, Hf, Ta, and Th) in each mineral grain were measured under the conditions listed in Table 1. The FE-EPMA comprises five wavelength-dispersive x-ray spectrometers, namely, CH1: TAPH-LDE2H, CH2: TAP-LDE1, CH3: LIF-PET, CH4: LIFH-PETH, and CH5: LIFH-PETH. The operating conditions were as follows: accelerating voltage = 15 kV, probe current = 50 nA, and beam diameter = 3 μm. In this study, the quickness of measurement was prioritized over precision because we were required to analyze a large number of grains for provenance estimation. To shorten analysis time, the background X-ray intensities of most elements were measured only on the lower side of the peaks (Table 1). The total measurement time for a spot was approximately 210 s. Data of < 80 wt% and > 110 wt% total
were rejected and excluded from the analytical results. At least 300 grains from each sample were measured. Mineral species were identified automatically on Microsoft® Excel® based on the criteria in Tables 2 and 3.

### RESULTS

A quick quantitative analysis of the bedrock samples showed that the heavy minerals in the Nohi Rhyolite and the Naegi-Agematsu Granite were mainly ilmenite, allanite, zircon (Figure 3), and biotite. These minerals are commonly included in welded tuff of the Nohi Rhyolite and the Naegi-Agematsu Granite, as reported in previous studies (Ishihara & Wu, 2001; Sonehara & Harayama, 2007; N. Yamada, 1977). There is no clear distinction between the mineral compositions of these two bedrocks.

Heavy minerals from the sediment samples consisted mainly of ilmenite, rutile, and zircon, but allanite and biotite were not present (Figure 4). Moreover, Al-vermiculite was abundant in the samples from the upper member, in contrast to those from the lower member, except for F-5. Zircon seemed to be more abundant in the upper member (except for F-13) than in the lower member. Because ilmenite and zircon were present in all analyzed samples, we compared the MnO content of ilmenite grains and the Y2O3 content of zircon grains among the samples. Ilmenite generally includes Mn as a solid solution, but it is occasionally changed to rutile owing to secondary alteration (Kuroda & Suwa, 1983). Therefore, ilmenite grains that contained anomalous amounts of TiO2 were deselected from the analytical results considering the possibility of alteration. Furthermore, if the atomic ratios of the results were not consistent with the chemical formula of ilmenite, the corresponding results were deselected.

### TABLE 1 Measurement condition of FE-EPMA analysis for each element

| Element | CH   | Rowland circle radius (mm) | Analyzing crystal | X-ray | Peak position (mm)/measurement time (s) | BG position (mm)/measurement time (s) | Slope of BG (cps/mm) | Procedure to eliminate interference |
|---------|------|---------------------------|-------------------|-------|----------------------------------------|--------------------------------------|---------------------|------------------------------------|
| Na      | CH1  | 100                       | TAPH              | Kα    | 129.522/10                             | 5.5/5                                | 5.5/5               | PHAa                               |
| F       | CH1  | 100                       | TAPH              | Kα    | 199.192/10                             | 5.0/5                                | 5.0/5               | PHA                               |
| P       | CH2  | 140                       | TAP               | Kα    | 66.727/10                              | 5.0/5                                | 5.0/5               | PHA                               |
| Si      | CH2  | 140                       | TAP               | Kα    | 77.343/10                              | 5.0/5                                | -3.6                | PHA                               |
| Al      | CH2  | 140                       | TAP               | Kα    | 90.661/10                              | 5.0/5                                | -1.5                | PHA                               |
| Ho      | CH2  | 140                       | TAP               | Mα    | 99.468/10                              | 5.0/5                                | 5.0/5               | PHA                               |
| Mg      | CH2  | 140                       | TAP               | Kα    | 107.591/10                             | 5.0/5                                | 5.0/5               | PHA                               |
| Ti      | CH3  | 140                       | PETJ              | Kα    | 87.997/10                              | 4.0/5                                | -2.6                | -                                  |
| Ca      | CH3  | 140                       | PETJ              | Kα    | 107.596/10                             | 3.0/5                                | -0.2                | -                                  |
| K       | CH3  | 140                       | PETJ              | Kα    | 119.855/10                             | 3.0/5                                | -1.06               | -                                  |
| Th      | CH3  | 140                       | PETJ              | Mα    | 132.521/10                             | 3.0/5                                | -0.8                | -                                  |
| S       | CH3  | 140                       | PETJ              | Kα    | 172.046/10                             | 5.0/5                                | -0.05               | -                                  |
| Zr      | CH3  | 140                       | PETJ              | Lα    | 194.480/10                             | 5.0/5                                | 0.02                | -                                  |
| Ta      | CH3  | 140                       | PETJ              | Mα    | 232.370/10                             | 1.5/5                                | -0.7                | -                                  |
| Hf      | CH4  | 100                       | LIFH              | Lα    | 108.863/10                             | 2.0/5                                | -2.7                | PHA                               |
| Er      | CH4  | 100                       | LIFH              | Lα    | 123.885/10                             | 5.0/5                                | -1.6                | Interference correction through the procedure described in Åmli and Griffin (1975) |
| Mn      | CH4  | 100                       | LIFH              | Kα    | 146.059/10                             | 5.0/5                                | -1.3                | -                                  |
| Nd      | CH4  | 100                       | LIFH              | Lβ    | 150.575/10                             | 5.0/5                                | -1.8                | -                                  |
| Ce      | CH4  | 100                       | LIFH              | Lα    | 178.102/10                             | 5.0/5                                | -2.0                | -                                  |
| Y       | CH4  | 100                       | PETH              | Lα    | 206.574/10                             | 5.0/5                                | -0.13               | -                                  |
| Nb      | CH4  | 100                       | PETH              | Lα    | 183.352/10                             | 2.0/5                                | -1.0                | -                                  |
| Yb      | CH5  | 100                       | LIFH              | Lα    | 116.531/10                             | 5.0/5                                | -1.17               | -                                  |
| Dy      | CH5  | 100                       | LIFH              | Lα    | 132.991/10                             | 5.0/5                                | -1.11               | Interference correction through the procedure described in Åmli and Griffin (1975) |
| Fe      | CH5  | 100                       | LIFH              | Kα    | 134.917/10                             | 3.0/5                                | -1.6                | -                                  |
| Sm      | CH5  | 100                       | LIFH              | Lβ    | 139.149/10                             | 2.0/5                                | -2.4                | -                                  |
| Pr      | CH5  | 100                       | LIFH              | Lβ    | 157.227/10                             | 5.0/5                                | -1.2                | -                                  |
| Cr      | CH5  | 100                       | LIFH              | Kα    | 159.462/10                             | 5.0/5                                | -1.02               | -                                  |
| La      | CH5  | 100                       | LIFH              | Lα    | 185.537/10                             | 3.0/5                                | -1.0                | -                                  |

- Pulse height analyzer.
The MnO contents of ilmenites from the Nohi Rhyolite range mainly from 2.5 wt% to 6.5 wt% (Figure 5a). In case of the Naegi-Agematsu Granite, the MnO content ranges mainly from 3.5 wt% to 7.5 wt%, which is slightly higher than that in the Nohi Rhyolite. The Y2O3 content of the zircons from the Nohi Rhyolite was mostly < 1 wt%, whereas the Naegi-Agematsu Granite contained zircons with ≥ 1 wt% Y2O3 (Table 4).

## DISCUSSION

### 5.1 Provenance analysis using EPMA

The results of a quick quantitative analysis and mineral identification showed that differences in heavy-mineral compositions between the Nohi Rhyolite and the Naegi-Agematsu Granite, both of which can be considered to be provenance rocks of the studied sediments (Figure 3), were not clearly recognized. The Nohi Rhyolite and the Naegi-Agematsu Granite were possibly derived from similar sources of magma (Sonehara & Harayama, 2007). In addition, their formation ages are not significantly different within the range of precision of radiometric age determination in previous studies (N. Yamada & Akahane, 2005; N. Yamada & Koido, 2005). Thus, further examination of the chemical compositions of specific minerals could be helpful, in addition to analysis of mineral compositions or ages.

The MnO content of ilmenite grains and the Y2O3 content of zircon grains differed between the upper and the lower members (Figure 5b,c; Table 5). In the samples from the lower member, ilmenite grains containing 2.5–5 wt% MnO were dominant (Figure 5b), although ilmenite grains containing 1 wt% MnO were common in F-8 as well. The ilmenite grains from the upper member showed two major peaks: grains containing < 1 wt% MnO and grains containing 3–6 wt% MnO (Figure 5c). Zircons containing ≥ 1 wt% Y2O3 were common in the upper member, whereas the Y2O3 content of most zircons from the lower member was < 1 wt% (Table 5).

### TABLE 2

| No. | Mineral | Component | Criteria |
|-----|---------|-----------|----------|
| 1   | Allanite| FeO       | ≥ 5 wt%  |
|     |         | Al2O3     | ≥ 5 wt%  |
|     |         | Ce2O3     | ≥ 3 wt%  |
| 2   | Apatite | P2O5      | ≥ 40 wt% |
|     |         | CaO       | ≥ 30 wt% |
| 3   | Columbite| Ta2O5     | ≥ 30 wt% |
| 4   | Epidote | CaO       | ≥ 5 wt%  |
|     |         | FeO       | ≥ 5 wt%  |
|     |         | Al2O3     | ≥ 20 wt% |
| 5   | Fergusonite| Nb2O5   | ≥ 30 wt% |
|     |         | Y2O3      | ≥ 10 wt% |
| 6   | Fluorite| CaO       | ≥ 30 wt% |
|     |         | F         | ≥ 30 wt% |
| 7   | Hornblende| MgO + FeO| ≥ 20 wt% |
|     |         | Al2O3     | 4–11 wt% |
|     |         | SiO2      | ≥ 40 wt% |
|     |         | CaO       | 9–12 wt% |
| 8–1 | Ilmenite| TiO2      | ≥ 40 wt% |
|     |         | MnO       | ≥ 0.1 wt%|
|     |         | FeO       | ≤ 30 wt% |
| 8–2 | Ilmenite| TiO2      | ≥ 40 wt% |
|     |         | MnO       | ≥ 15 wt% |
|     |         | FeO       | ≤ 30 wt% |
| 9   | Monazite| Ce2O3     | ≥ 20 wt% |
|     |         | Nd2O3     | ≥ 3 wt%  |
|     |         | La2O3     | ≥ 3 wt%  |
| 10  | Rutile  | TiO2      | ≥ 60 wt% |
| 11  | Thorite | ThO2      | ≥ 40 wt% |
| 12  | Xenotime| Y2O3      | ≥ 30 wt% |
|     |         | P2O5      | ≥ 20 wt% |
| 13  | Zircon  | ZrO2      | ≥ 20 wt% |
| 14  | Augite  | SiO2      | ≥ 45 wt% |
|     |         | MgO       | ≥ 7 wt%  |
|     |         | CaO       | ≥ 17 wt% |
|     |         | FeO       | ≥ 10 wt% |
|     |         | Al2O3     | ≤ 3 wt%  |
| 15  | Hypersthenesilicate| SiO2 | ≥ 45 wt% |
|     |         | MgO       | ≥ 10 wt% |
|     |         | FeO       | ≥ 25 wt% |
|     |         | Al2O3     | ≤ 3 wt%  |
|     |         | CaO       | ≤ 5 wt%  |
|     |         | MgO + FeO | ≥ 30 wt% |
| 16  | Titanomagnetite| TiO2 | 5–40 wt% |
|     |         | FeO       | ≥ 40 wt% |

The MnO contents of ilmenites from the Nohi Rhyolite range mainly from 2.5 wt% to 6.5 wt% (Figure 5a). In case of the Naegi-Agematsu Granite, the MnO content ranges mainly from 3.5 wt% to 7.5 wt%, which is slightly higher than that in the Nohi Rhyolite. The Y2O3 content of the zircons from the Nohi Rhyolite was mostly < 1 wt%, whereas the Naegi-Agematsu Granite contained zircons with ≥ 1 wt% Y2O3 (Table 4).
included commonly in the host rocks of the Nohi Rhyolite and the Naegi-Agematsu Granite, could be potentially useful from the viewpoint of estimating the provenance rocks in this study.

The MnO content of the ilmenites from the Naegi-Agematsu Granite ranged from 3.5 wt% to 7.5 wt%, which was slightly higher than that of the ilmenites from the Nohi Rhyolite (Figure 5a). Because ilmenite contains Mn as a solid solution, the MnO content of ilmenites can be used to determine the chemical composition of magma during ilmenite crystallization. Mineral crystallization temperature depends mainly on the chemical composition and water content of magma. Minerals with a higher initial magma water content crystallize at a lower temperature. Sonehara and Harayama (2007) suggested that the minerals found in the Nohi Rhyolite crystallized from hot (approximately 830 °C) and dry (approximately 1.0 wt% water content) felsic magma. We compared the approximate crystallization temperatures of the minerals calculated using MELTS (Asimow & Ghiorso, 1998; Ghiorso & Sack, 1995) in a manner similar to that employed by Christiansen (2005). Sonehara and Harayama examined the crystallization path of the Nohi Rhyolite by using MELTS. Based on the results of Sonehara and Harayama, mafic mineral crystallizes at higher

**FIGURE 3** Abundance ratio of minerals in bedrock samples identified using quick quantification method

**FIGURE 4** Abundance ratio of minerals in sediment samples identified using quick quantification method. The sample numbers are listed in order of their positions in the outcrop from upper to lower

**FIGURE 5** Legend on next page.
temperatures than ilmenite (Figure 6). This suggests that the Mn in the magma could be incorporated into mafic minerals before the crystallization of ilmenite. The crystallization temperature of ilmenite in the Nohi Rhyolite was estimated to be approximately 890 °C (Figure 6). Although mafic minerals were rare in the host rock samples used in the present study, large quantities of mafic minerals have been altered to chlorite in the common Nohi Rhyolite, as reported in previous studies (Sonehara & Harayama, 2007; N. Yamada & Koido, 2005). Thus, the order of crystallization of ilmenite and mafic minerals could explain why the ilmenite grains from the Nohi Rhyolite had lower MnO contents than the ilmenite grains from the Naegi-Agematsu Granite.

As described above, ilmenite grains rich in MnO are more abundant in the upper members of the studied sediments than in the lower members (Figure 5b,c). This observation implies that minerals and fragments from the Naegi-Agematsu Granite were included in the upper member in contrast to the lower member. However, as mentioned above, the difference between the MnO contents of ilmenite grains from the Naegi-Agematsu Granite and the Nohi Rhyolite are minor. The MnO content of ilmenite grains is controlled by several factors, such as oxygen fugacity (fO₂) and chemical composition of magma (Imaoka, Nakashima, & Murakami, 1982). Therefore, we cannot clearly identify the source rocks based only on the MnO content of ilmenite grains. By contrast, ilmenites with < 1 wt% MnO are included measurably in a few sediment samples. Their occurrence could have resulted from Mn dissolution due to alteration and weathering. Moreover, the unclear difference between the upper and lower members may be a result of Mn dissolution.

Zircons with ≥ 1 wt% Y₂O₃ are relatively common in the Naegi-Agematsu Granite (approximately 7.8 %) but are very rare in the Nohi Rhyolite (less than 1.0 %) (Table 4). This finding is consistent with the results of a previous study (Suzuki & Yogo, 1986), suggesting that Y₂O₃-rich zircon grains are unique to the Naegi-Agematsu Granite. The Nohi Rhyolite and the Naegi-Agematsu Granite are considered to have been formed by a series of large silicic magmatic events (Sonehara & Harayama, 2007). Yttrium (Y) is one of the incompatible elements, and it is concentrated preferentially in the melt phase of magma rather than in the solid phase. Thus, the Naegi-Agematsu Granite, which was formed after formation of the Gero Ash-Flow Sheet of the Nohi Rhyolite (N. Yamada & Akahane, 2005), could be rich in Y₂O₃. Rapid quantification of abundant grains from the sediments in the studied location showed that zircon grains with ≥ 1 wt% Y₂O₃ were present widely in the upper member in contrast with the lower member, where the Y₂O₃ contents of most zircons were < 1 wt% (Table 5). This suggests that the Naegi-Agematsu Granite was enclosed in the provenance of the studied location during deposition of the upper member. This result is consistent with the difference in gravel composition between the lower and upper members. Thus, the chemical analysis of heavy minerals is effective for provenance estimation in the studied area. Similar to the granite gravels in the Toki Sand and Gravel Formation in the study area, several types of gravels were found to be weathered severely, and it was difficult to identify their lithology. In addition, mineral identification using optical microscopy for massive quantities of fine sands is time-consuming, especially for beginners, and the risk of misidentification is non-negligible. In such cases, our examination method based on grain-by-grain chemical composition can be helpful for identifying source rocks in the sediments accurately and precisely.

### Table 4 Number of zircon grains from basement-rock samples sorted by Y₂O₃ content

| Basement rock           | Y₂O₃ < 1 wt% | 1 wt% ≤ Y₂O₃ < 3 wt% | Y₂O₃ ≥ 3 wt% | n |
|-------------------------|--------------|----------------------|--------------|---|
| Naegi-Agematsu Granite  | 214          | 12                   | 6            | 232|
| Nohi Rhyolite           | 102          | 1                    | 0            | 103|

### Table 5 Number of zircon grains from sediment samples sorted by Y₂O₃ content

| Position in the outcrop | Sample | Y₂O₃ < 1 wt% | 1 wt% ≤ Y₂O₃ < 3 wt% | Y₂O₃ ≥ 3 wt% | n |
|-------------------------|--------|--------------|----------------------|--------------|---|
| Upper                   | F-13   | 2            | 0                    | 4            | 6 |
|                         | F-12   | 15           | 1                    | 9            | 25|
|                         | F-11   | 17           | 10                   | 21           | 48|
| Lower                   | F-10   | 24           | 5                    | 13           | 42|
|                         | F-14   | 10           | 9                    | 7            | 26|
|                         | F-8    | 8            | 0                    | 0            | 8 |
|                         | F-7    | 9            | 0                    | 0            | 9 |
|                         | F-5    | 4            | 1                    | 0            | 5 |
|                         | F-2    | 14           | 0                    | 0            | 14|

Sample numbers in descending order of positions in the outcrop.

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**Figure 5** Histogram of MnO content in ilmenites from basement rocks and sediments. (a) Basement-rock samples. (b) Lower members of the Toki Sand and Gravel Formation at the sampling location. (c) Upper members of the Toki Sand and Gravel Formation at the sampling location. In horizontal axes, 0.5 represents 0 wt % ≤ MnO ≤ 0.5 wt%, whereas 1, 1.5, 2, ... represent 0.5 wt % < MnO ≤ 1.0 wt%, 1.0 wt% < MnO ≤ 1.5 wt%, 1.5 wt % < MnO ≤ 2.0 wt%, ..., respectively. Along the vertical axes, relative frequency (%) is defined as the number of ilmenite grains divided by the total number of ilmenite grains in each sample. The sample numbers in (b) and (c) are listed in order of positions in the outcrop from upper to lower.

**Figure 6** Crystallization temperatures of minerals in Nohi Rhyolite based on calculations using MELTS.
5.2 | Relationship between provenance changes and history of fault activity

Changes in provenance from the lower member to the upper member were ascertained by analyzing the chemical composition of heavy minerals in this study, as well as based on the gravel composition observed in the outcrop. Namely, sediments of the lower member were sourced mainly from the Nohi Rhyolite, whereas those of the upper member were sourced from both the Naegi-Agematsu Granite and the Nohi Rhyolite. Znp–Oht tephra (3.9 Ma; Kurokawa & Tomita, 1998; Tamura et al., 2005) or its equivalents were included in the sandy layer near F-8 in the lower member (Yasue et al., 2011). In addition, the plagioclase K–Ar age of the basalt gravel from the upper member was found to be (2.08 ±0.30) Ma (Appendix, Table S2), which was within the age range of the Ueno basaltic rocks. These age data show that the changes in provenance from the lower to the upper members occurred over the period from 3.9 Ma to < 2.08 Ma.

Although the activity of the Atera Fault near the eastern side of the sediment sampling location was initiated in the Late Cretaceous (Kanaori, Endo, Yairi, & Kawakami, 1990), displacement of the fault with left-lateral strike-slip and uplift of the eastern side were reactivated since approximately 3 Ma (Yasue, Bateer, & Takeuchi, 2003) (Figure 7a). The Tsukechi River was displaced by 7–7.5 km owing to sinistral movement of the Atera Fault (Tsukuda et al., 1993) (Figure 7b). The eastern side of the Atera Fault was uplifted by ~1 km (Sugimura & Matsuda, 1965; Tsukuda et al., 1993; N. Yamada, 1981). Moreover, K. Yamada et al. (2012) calculated erosion across the Atera Fault and true dip displacement of the fault based on differences in zircon fission-track ages across the fault and suggested that dip-slip displacement of the fault since 70 Ma was approximately 1 km. This result is consistent with that estimated from the topography across the fault (N. Yamada, 1981). The paleocurrent of the upper member is directed from northeast to southwest. Therefore, the activity of the Atera Fault seemingly caused exposure of the Naegi-Agematsu Granite east of the study location (Figure 7b, cross-section view), and the clastics from the granite were mixed in the upper member. The Ueno basaltic rocks occur not only in the neighborhood of the studied location (Sakashita mass) but also in an upstream area in the northeast (Kiso province) (Figure 7b, plane view), because of which the basalt gravel may have mixed with the upper member.

6 | CONCLUSIONS

To perform efficient provenance analysis, we developed a method for quick identification of heavy minerals by using EPMA. This method was applied to sediments of the Toki Sand and Gravel Formation, central Japan. Because the Nohi Rhyolite and the Naegi-Agematsu Granite of the candidate provenance rocks in the study area have similar formation ages, it is difficult to distinguish between them by performing grain-by-grain radiometric dating. Hence, we focused on mineral and chemical compositions.

FIGURE 7 Schematics showing changes in provenance in and around the studied location. (a) Gravels of the Nohi Rhyolite were provided mainly to the studied location until sometime in the period from 3.9 Ma to < 2.08 Ma. (b) After that time, gravels of the Naegi-Agematsu Granite and the Ueno basaltic rocks started to be provided to the studied location as well.
By using the method for quick identification of heavy minerals with an EPMA, we estimated the heavy-mineral compositions of the Nohi Rhyolite and the Naegi-Agematsu Granite. Both contained biotite, ilmenite, allanite, and zircon, which is consistent with their previous petrographic descriptions. We focused on variations in the chemical compositions of ilmenite and zircon because the heavy-mineral compositions of the Nohi Rhyolite and the Naegi-Agematsu Granite were similar. Zircon grains with > 1 wt% Y2O3 were common in the Naegi-Agematsu Granite, whereas most of the zircon grains from the Nohi Rhyolite contained < 1 wt% of Y2O3. In addition, zircons with > 1 wt% Y2O3 were common in the upper member but very rare in the lower member of the Toki Sand and Gravel Formation at the sampling location. The MnO content of the ilmenite grains from the Naegi-Agematsu Granite was slightly higher than that of the ilmenite grains from the Nohi Rhyolite. However, the difference was too small to serve as a discriminator between these two bedrocks. This result indicates that the Naegi-Agematsu Granite was exposed to the land surface in the provenance of the sampling location after deposition of the lower member, which is consistent with its gravel composition. This provenance change could be explained by uplift and denudation of the Atera Mountains associated with reactivation of the nearby Atera Fault (Kanaori et al., 1990; Yasue et al., 2003).

The proposed quick quantification and mineral identification method based on EPMA can contribute to provenance analysis based on abundance ratio and chemical composition of heavy minerals. This method is especially helpful for fine-grained sediments without gravels, that is, in cases where identification of provenance rocks by mesoscopic examination is difficult. This method can provide some clues for provenance estimation, as well as geological description, tephrochronology, and grain-by-grain radiometric dating.

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APPENDIX A

K–Ar dating of a basalt gravel from a gravel bed in the upper member of the Toki Sand and Gravel Formation (Figure 1c) was performed at Hiruzen Institute for Geology and Chronology. We selected plagioclase phenocrysts for K–Ar dating because other phenocrysts such as augite and olivine are very poor in potassium and were generally altered, and the groundmass was affected largely by alteration. The sample was crushed and sieved into a fraction between 200 meshes and 300 meshes (50–75 μm in range) to concentrate plagioclase phenocrysts. The procedure for mineral separation was based on Yagi (2006) and Yagi and Itaya (2011).

Quantitative determination of potassium was performed with flame photometry by using a Model 180-30 atomic absorption spectrometer manufactured by Hitachi (Nagao, Nishido, Itaya, & Ogata, 1984). The solution of acid-degraded plagioclase sample in HCl, with added cesium for inhibiting ionization interference, was measured using flame photometry. The potassium content in the solution was calculated by averaging two measurements for one sample. Error in the potassium content was taken to be 2 % based on multiple analyses of standard samples (JG-1 and JB-1) issued by the Geological Survey of Japan.

Argon isotopic ratio was determined using the mass spectrometer in Okayama University of Science (HIRU; Itaya et al., 1991; Nagao & Itaya, 1988; Nagao et al., 1984). We adopted the isotope dilution method and employed $^{38}\text{Ar}$ as a spike. We calculated the argon isotopic ratio by referring to Itaya et al. (1991). We used the decay constants of $\lambda_e = 0.581 \times 10^{-10}$/year and $\lambda_\beta = 4.962 \times 10^{-10}$/year, as recommended by Steiger and Jäger (1977). Here, $\lambda_e$ and $\lambda_\beta$, respectively, represent the decay constant for the $^{40}\text{K} \rightarrow ^{40}\text{Ar}$ electron capture decay and the decay constant for the $^{40}\text{K} \rightarrow ^{40}\text{Ca} \beta^-$ decay.

The result of K analysis of plagioclase K–Ar dating of the basalt gravel showed that the difference between repeated K measurements of the same sample was within 2 % of the mean (Table S1). The K–Ar age of plagioclase phenocrysts in the basalt gravel was found to be (2.08 ±0.30) Ma (Table S2).