

Article



Age of Initial Submarine Volcanism in the Paleo-Tsushima Basin and Implications for Submarine Volcanism in the Opening Stage of the Japan Sea in Northern Kyushu

Takashi Ninomiya ^{1,*}, Shoichi Shimoyama ², Sho Taniguchi ³, Toshihiro Takahashi ⁴, Tohru Danhara ⁵ and Hideki Iwano ⁵

- ¹ Office for Graduate School Education, Graduate School of Science, Kyushu University, Fukuoka 819-0395, Japan
- ² Department of Civil Engineering and Architecture, Faculty of Science and Engineering, Saga University, Saga 840-8502, Japan; cshmym@diary.ocn.ne.jp
- ³ Hazama Ando Corporation, Tokyo 107-8658, Japan; taniguchi.sho@ad-hzm.co.jp
- ⁴ Japan Petroleum Exploration Co., Ltd., Tokyo 100-0005, Japan; toshihiro.takahashi@japex.co.jp
 ⁵ Kyoto Fission-Track Co., Ltd., Kyoto 603-8832, Japan; kft-home@r5.dion.ne.jp (T.D.);
- iwano_hide@zeus.eonet.ne.jp (H.I.) Correspondence: t.ninomy@geo.kyushu-u.ac.jp; Tel.: +81-92-802-4027

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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). **Abstract:** The Tsushima Lapilli Tuff, the thickest tuff in the Taishu Group on Tsushima Island, underwent a thermal event after deposition, and has not previously yielded a reliable age because various ages have been reported. This study clarifies the eruption age and thermal history of the Tsushima Lapilli Tuff based on fission-track (FT) and U–Pb dating of zircon grains using laser ablation inductively coupled plasma mass spectrometry (ICP-LA-MS) and evaluates submarine volcanism during deposition of the Taishu Group in the southwestern Japan Sea, as well as volcanism change on Tsushima Island. This study revealed that thermal events caused rejuvenation in some single-grain FT ages after deposition in the Tsushima Group, and that the eruption age of the Tsushima Lapilli Tuff was 16.2 ± 0.7 Ma; the age of the largest submarine volcanism event in the Taishu Group in Tsushima Island was thus determined. On the basis of our previous studies, this age and tectonism strongly indicate that felsic submarine volcanism occurred between 18 and 16 Ma, accompanied by rapid subsidence, and the volcanism changed from felsic volcanism originating from melting of old continental crust by asthenospheric upwelling to mafic volcanism originating from small-scale lithospheric mantle upwelling from 13.6 Ma onward.

Keywords: fission-track dating; U–Pb dating; Taishu Group; 16.2 \pm 0.7 Ma; eruption age

1. Introduction

Strata containing abundant pyroclastic rocks deposited during the opening stage of the Japan Sea (23–15 Ma) are distributed along the Japan Sea coast from southern Hokkaido to northern Kyushu (e.g., [1]). The pyroclastic rocks showing greenish gray color caused by alteration after deposition are called green tuff, and their distribution area is called the Green Tuff region (Figure 1). The pyroclastic strata are key to clarifying the volcanism and tectonism sequence of the opening stages of the Japan Sea, and numerous studies have been performed for that purpose (e.g., [2–4]). The Miocene Taishu Group, widely distributed in the southwestern Japan Sea, is an important group because it connects the Green Tuff region and the Miocene groups distributed in northern Kyushu such as the Goto Group in the Goto Islands and Nojima Group in the Sasebo district [5]. The Taishu Group is a thick marine group (>5000 m) containing abundant pyroclastic rocks. The Taishu Group has been suggested to have been regarded as evidence supporting the idea that the Japan Sea was formed by long-term tectonism (e.g., [6]). However, the Taishu Group could not

contribute to investigation related to the opening of the Japan Sea because of this proposed long-term age and unusual thickness of the group dominated by mudstone indicating unique sedimentary basin formation [5,6].

Recently, re-examination of the formation age of the Taishu Group was performed based on U–Pb dating using SHRIMP (sensitive high-resolution ion micro probe) [5]. Ninomiya et al. [5] clarified that the Taishu Group was deposited in the opening stage of the Japan Sea at 17.9–15.9 Ma by rapid sedimentation, mainly composed of deep-sea rocks, without a thermal event causing rejuvenation of the U–Pb age after deposition, and that this group can be compared with the strata in the Green Tuff region. The Tsushima Lapilli Tuff (also called the T3 or Tk tuff; [7]) which is an important dacitic submarine pyroclastic rock unit, can be traced over 40 km, and can be considered a key tuff in the stratigraphy of the Taishu Group. In addition, the age of the Tsushima Lapilli Tuff constrains the formation age of the deep-sea basin called the Paleo-Tsushima Basin (water depth > 800 m) because the tuff was formed by the initial submarine volcanism in the deep-sea basin, which appeared by rapid subsidence associated with the opening of the Japan Sea [5,8].

However, various ages have been proposed for the Tsushima Lapilli Tuff, and a reliable age has not previously been obtained to constrains the formation age of the deepsea basin [9–12]. Its age was regarded as ca. 20 Ma based on zircon fission-track (FT) age dating conducted in the 1980s [9,10]. Subsequently, Sakai and Yuasa [11] reported a range between 46 Ma and 15 Ma based on samples from the same locality and whole-rock K-Ar dating, and proposed that the oldest age (46 Ma) was the true eruption age of the Tsushima Lapilli Tuff. This age is evidence showing that the Taishu Group was deposited in the middle Eocene. In addition, Sakai and Yuasa [11] noted the possibility that the young ages contained in the FT age data were rejuvenated by thermal influence originating from intrusive rocks and indicated that a thermal event had caused rejuvenation of the previously obtained FT ages. Therefore, we aimed to determine the eruption age of the Tsushima Lapilli Tuff based on U–Pb and FT dating to constrain the formation age of the Paleo-Tsushima Basin and confirm the occurrence of rejuvenation of the FT ages (thermal history after deposition) with a reliable age to solve the above problems. In addition, we evaluated the formation age of the Paleo-Tsushima Basin and the volcanism changes of the southwestern Japan Sea in the opening stage of the Japan Sea based on the age of the Tsushima Lapilli Tuff, previously obtained radiometric ages and tectonism in the southwestern Japan Sea [5,13,14].

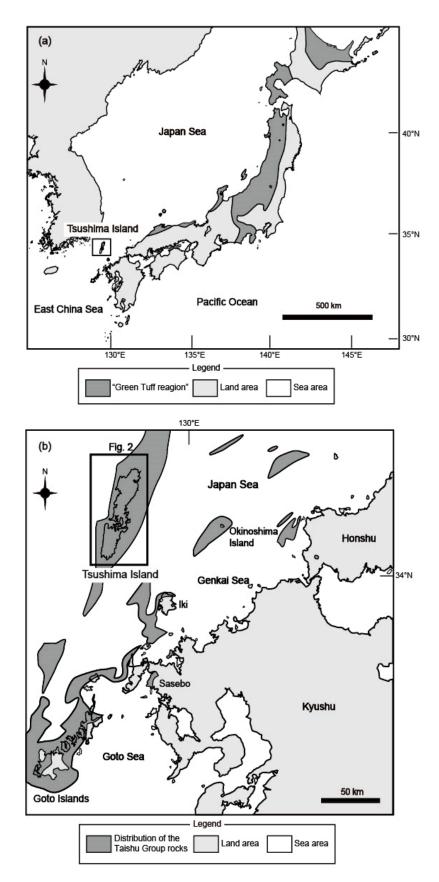


Figure 1. (a) "Green Tuff region" (distribution of lower–middle Miocene rocks) and study area. Rectangle indicates the study area. Modified from Fujioka [15]. (b) Maps showing distributions of the Taishu Group near Tsushima Island. Modified from the Geological Survey of Japan [16].

2. Geology of Tsushima Island

Tsushima Island is located in the Japan Sea off of northwestern Kyushu (Figure 2). The Taishu Group on Tsushima Island comprises over 5000 m of thick, mudstone-dominated marine strata classified into Lower, Middle and Upper units [17–20]. The Lower unit is subdivided into basal and upper parts. The basal part was deposited in a delta system, based on sedimentary facies analysis [21].

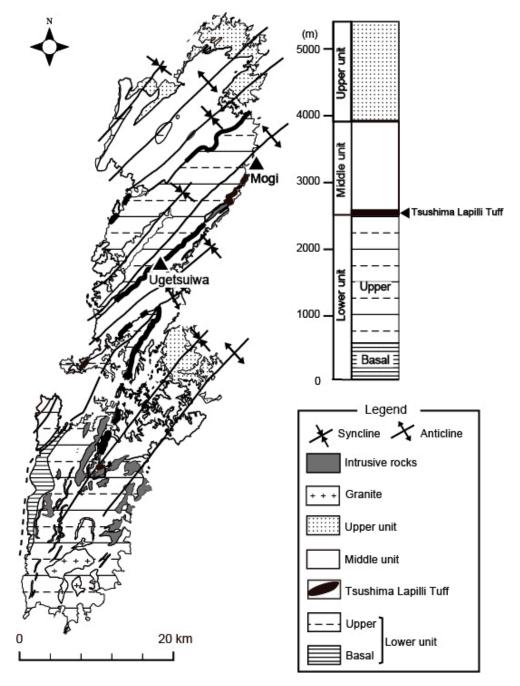


Figure 2. Geologic map of Tsushima Island. Modified from Ninomiya et al. [5]. Triangles show the sampling point of the Tsushima Lapilli Tuff.

The succession comprising the upper part of the Lower Unit and the Upper Unit is mainly regarded as deep-sea sedimentary rocks because no sedimentary environmental change is observed (e.g., [5,22]), fossil assemblages indicating a deep-sea environment were reported ([23–26]), and the Upper Unit is regarded as deep-sea rocks [23]. However,

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the Upper Unit distributed in central Tsushima Island and the Upper Unit distributed on northern Tsushima Island were regarded as a homogenous facies by Ninomiya et al. [5] based on the previously performed mapping (e.g., [20,27]). Anticlinorium–synclinorium structures, which are open folded with a wavelength of less than 2 km and northeast– southwest axes, plunge roughly to the northeast (e.g., [26,28]). These folds are considered to be part of the Taiwan–Shinji Fold Belt (e.g., [29]). In the southern area of Tsushima Island, a halo of hornfels is observed (e.g., [30]). Rhyolite and dacite related to granitic activity and dolerite are observed as intrusive rocks [31]. Pb–Zn mineralization is considered to be formed by granite intrusion [32], but many small-scale mineralized zones are observed on the island [33].

3. Materials and Methods

3.1. Material Description

The facies and lithology of the Tsushima Lapilli Tuff were well studied by Sakai and Kawahara [7]. They clarified that the Tsushima Lapilli Tuff is composed of a lower part, named the Mogi subaqueous pyroclastic flow deposit, which is locally distributed, and an upper part, named the Ugetsuiwa subaqueous pyroclastic flow deposit, which is widely distributed on Tsushima Island. In addition, Sakai and Kawahara [7] proposed that the Tsushima Lapilli Tuff originated from a large-scale terrestrial volcanic eruption, because rhyolitic-andesitic fragments present in the tuff were not observed at the chilled margin, and because of the presence of well-vesiculated pumice. In this study, we dated samples obtained from Ugetsuiwa and Mogi, type localities of the Tsushima Lapilli Tuff (Figure 3). The Tsushima Lapilli Tuff at Mogi, the type locality of the Mogi subaqueous pyroclastic flow deposit, is 35 m in thickness and includes both the Mogi subaqueous pyroclastic flow deposit and the Ugetsuiwa subaqueous pyroclastic flow deposit. The Mogi subaqueous pyroclastic flow deposit is light gray in fresh parts and white to dark brown in weathered parts, and is divided into the lower part, which is massive, and the upper part, which is stratified (Figure 4a). The Mogi subaqueous pyroclastic flow deposit exhibits vein structures filled with plagioclase crystals associated with fissures in mudstone that formed during deposition (Figure 4b; [7]). The basal part of the succession contains unconsolidated fragments of sandstone, rhyolite and quartz porphyry (Figure 4c). The other part mainly contains plagioclase and pumice with a small amount of rock fragments composed of mudstone and sandstone. The samples used for radiometric dating were obtained from the part with fewer rock fragments based on macroscopic observation (Figure 4d; TSU-Mg). The sample material is porphyritic with phenocrysts of coarse-grained plagioclase and minor quartz in a matrix of quartz plagioclase; secondary minerals represented by calcite in the thin sections indicate alteration after deposition (Figure 5a,b). Ugetsuiwa is the type locality of the Ugetsuiwa subaqueous pyroclastic flow deposit, which is distributed across a wide area of Tsushima Island. The Ugetsuiwa subaqueous pyroclastic flow deposit is light gray in fresh parts and white to brown in weathered parts. The succession is divided into the lower part, which is massive, and the upper part, which is stratified and tabular with cross-stratification. Vein structures filled with plagioclase crystals associated with fissures in mudstone that formed during deposition are also observed (Figure 6a,b). The basal part of the succession contains unconsolidated fragments of sandstone, rhyolite, pumice and chloritized pumice (Figure 6c). The other part is composed of plagioclase, pumice, quartz and rock fragments, mainly mudstone. The samples used for radiometric dating were obtained from the part with fewer rock fragments based on macroscopic observation, as was carried out for the Mogi site (Figure 6d; TAT-2-058). The sample material is porphyritic with phenocrysts of plagioclase and minor quartz in a matrix of quartz and plagioclase in the thin sections (Figure 5c,d). The plagioclases are often accompanied by calcite as a secondary mineral, indicating strong alteration after deposition compared with TSU-Mg.

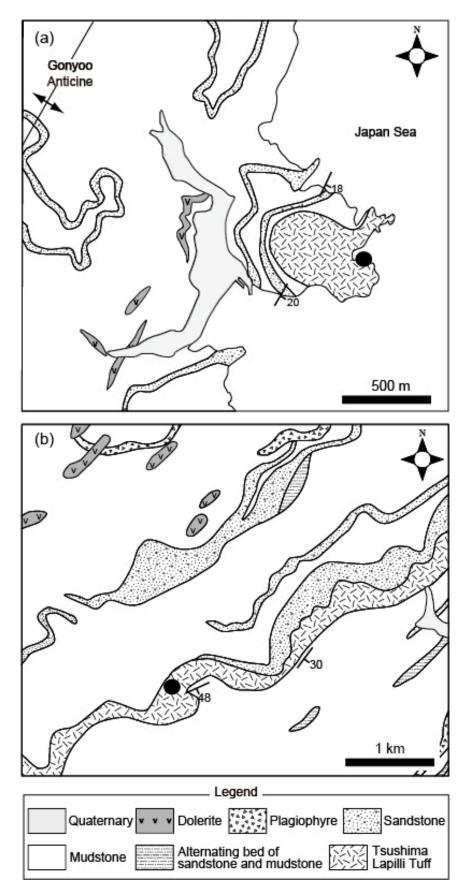


Figure 3. Geologic maps at (a) Mogi and (b) Ugetsuiwa, modified from [14]. Circles show sampling points.

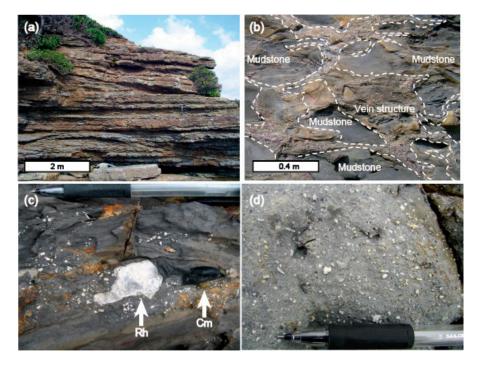


Figure 4. Photographs of the Tsushima Lapilli Tuff at Mogi. (**a**) Basal part of the Tsushima Lapilli Tuff. (**b**) Rhyolite and carbonaceous material as gravel. (**c**) Massive part and sampling point in the lower part. (**d**) Stratified part in the upper part.

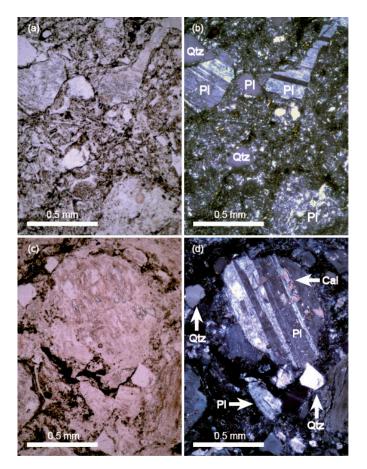


Figure 5. Photomicrographs showing microscopic features of the Tsushima Lapilli Tuff. (**a**,**b**) TSU-Mg. (**c**,**d**) TAT-2-058. (**a**,**c**) Under plane-polarized light. (**b**,**d**) Under cross-polarized light. Cal, calcite; Kf, K-feldspar; Pl, plagioclase; Qtz, quartz.

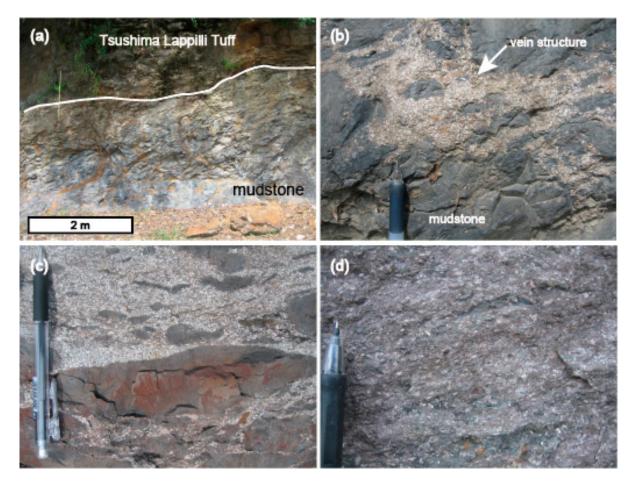


Figure 6. Photographs of the Tsushima Lapilli Tuff at Ugetsuiwa. (a) Boundary between the Tsushima Lapilli Tuff and mudstone comprising the Lower Unit. (b) Vein structure consisting of plagioclase crystals in the mudstone. (c) Basal part. (d) Massive part and sampling point in the lower part.

3.2. Methodology of LA-ICP-MS U–Pb and Fission-Track Double Dating

A sufficient number of zircon grains (>5000 grains) were extracted from each 0.3 kg rock sample (TSU-Mg and TAT-2-058) by standard mineral separation techniques, which involved crushing, sieving, panning, magnetic separation, gravity separation, and handpicking. The zircon grains were mounted on a PFA (copolymer of tetrafluoroethylene–perfluoroalkoxyethylene) sheet and polished with diamond pastes. They were then etched with KOH:NaOH (1:1) eutectic etchant at 225 °C until the etched tracks were isotropically distributed (ca. 30 h). Spontaneous track density measurement was performed for 30 grains, which were randomly selected using a high-resolution touch-monitor screen through a digital camera and an optical microscope, as described by Danhara and Iwano [34].

The zircon grains were analyzed using two laser ablation-inductively plasma mass spectrometry (LA–ICP–MS) systems installed at the Division of Earth Planetary Science of Kyoto University, Japan (Table S1). For sample TSU-Mg, the LA-ICP-MS system was an iCAP Qc quadrupole ICP-MS (Thermo Fisher Scientific, Waltham, MA, USA) combined with a New Wave Research NWR-193 ArF excimer laser-ablation system (Fremont, CA, USA) with a 25 µm diameter laser spot. The forward power of the ICP-MS was 1400 W. We used the standard mode of the ICP-MS, without collision reaction gases. Helium gas was used as the carrier gas inside the ablation cell and mixed with argon gas before entering the ICP-MS. Signal intensities for ²⁹Si, ²⁰²Hg, ²⁰⁴(Pb+Hg), ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb, ²³²Th and ²³⁸U were obtained from zircon grains. LA-ICP-MS sessions involved 2–3 analyses of each reference standard [NIST SRM610 and 91,500 zircon; 35] and 15–17 analyses of unknown samples or age standards, followed by repeated analyses of the reference standards. The

operating conditions are summarized in Table S1. The isotope ²⁹Si was used as an internal standard for normalizing ²³⁸U signal intensities for U concentration calculation.

For sample TAT-2-058, another system was used: an AttoM high-resolution magnetic sector field single-collector ICP-MS (Nu Instruments, Wrexham, UK) combined with the above-mentioned excimer laser ablation system with a 20 µm LA spot. The forward power of the ICP-MS was 1300 W. With this LA-ICP-MS, signal intensities for ²⁰²Hg, ²⁰⁴(Pb+Hg), ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb, ²³²Th and ²³⁸U were obtained without ²⁹Si. The operating conditions are summarized in Table S1. We calculated U–Pb ages and also approximate U concentration for each grain by means of the ²³⁸U signal data without internal standard correction based on Si following the method described by Noda et al. [35,36]. For both samples, fission track age calibration was based on the zeta approach [37–39] using the age standard Fish Canyon Tuff with a reference age of 28.45 Ma [40] and zeta values for 91,500 zircons as a uranium standard. Secondary standard zircons from OD-3 [41,42] and the Tardree Rhyolite [43] were also analyzed to assess the accuracy of U–Pb and FT dating, respectively.

4. Results

The dating results obtained in this study are shown in Tables S2–S7 and Figures 7–9. For sample TSU-Mg, the resulting U–Pb data showed 26 concordant grains ranging from 13.9 to 355.9 Ma with four discordant grains (Nos. 9, 26, 28 and 30). Among them, two older grains (Nos. 4 and 6) were excluded as detrital grains or xenocrysts. The remaining 24 grains were regarded as the youngest U–Pb grain population within a 3 σ error range, giving a weighted mean U–Pb age of 16.2 ± 0.3 Ma (2 σ) (Table S6). For FT age calculation, Figure 9a revealed that most of the grains had consistent FT ages regardless of their U–Pb ages. However, grain No. 6 was excluded as an outlier with a remarkably older FT age. As a result, the remaining 29 grains yielded a pooled FT age of 15.8 ± 0.7 Ma (1 σ) (Table S2). We confirmed that the resulting FT age for the Tardree Rhyolite was in agreement with the reference age.

For sample TAT-2-058, the resulting U–Pb data showed 19 concordant grains ranging from 14.3 to 1880 Ma with eleven discordant grains (Nos. 6, 7, 10, 13, 14, 15, 19, 20, 25, 27 and 30). Among them, two older grains (Nos. 1 and 4) were excluded as detrital grains or xenocrysts (Table S7). The remaining 17 grains were regarded as the youngest U–Pb grain population within 3σ error, giving a weighted mean U–Pb age of 16.1 ± 0.3 Ma (2σ) (Table S7). For FT age calculation, Figure 9b revealed that all grains had consistent FT ages regardless of their U–Pb ages. Therefore, 30 grains can be regarded as a single population with a mean age of 16.6 ± 1.0 Ma (1σ) (Table S3).

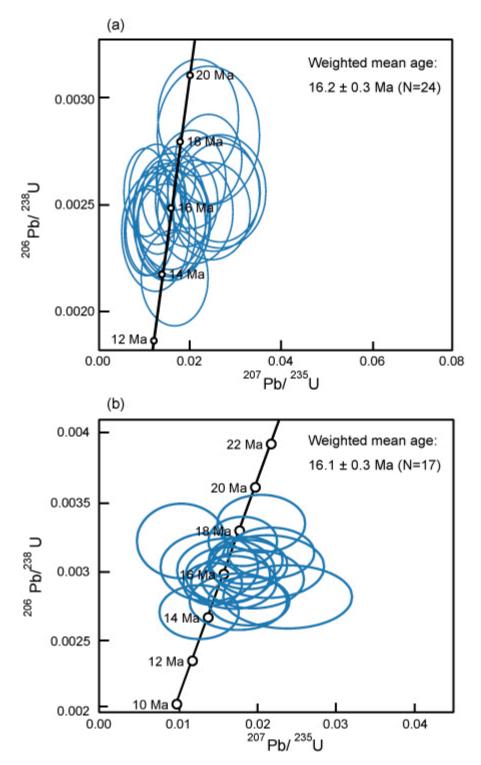


Figure 7. U–Pb concordia plot of the youngest concordant clusters. (a) TSU-Mg. (b) TAT-2-058.

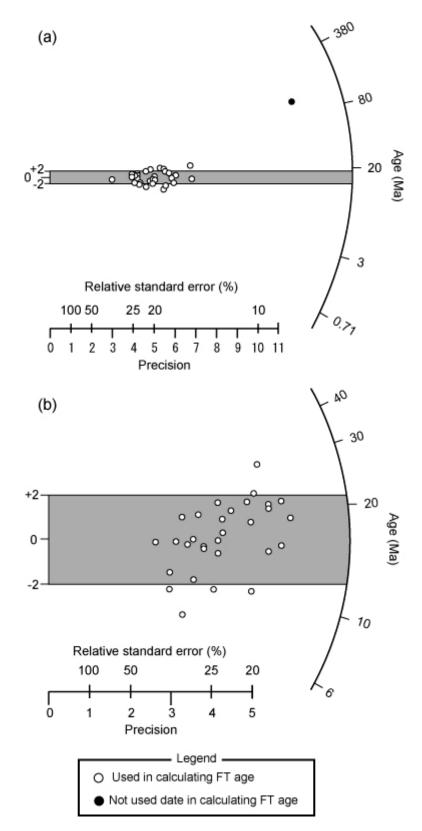


Figure 8. Radial plot of FT dating. (a) TSU-Mg. (b) TAT-2-058.

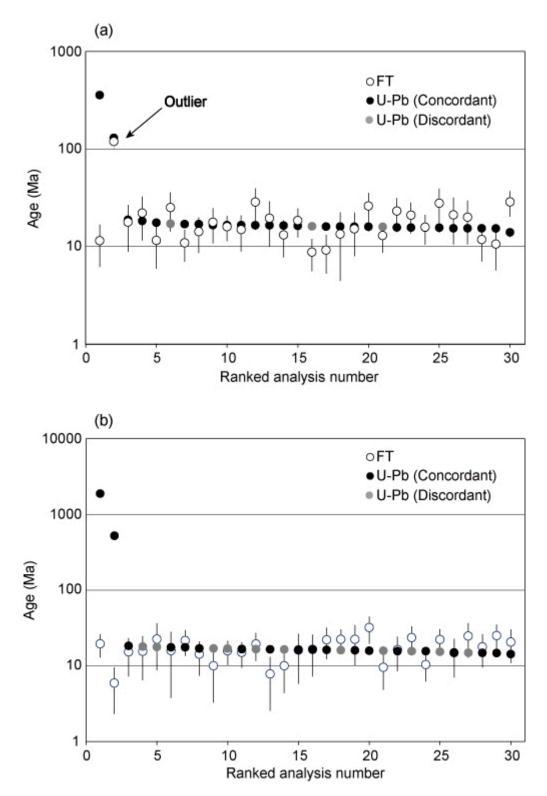


Figure 9. Grain-by-grain relationship between FT and U–Pb ages for (**a**) sample TSU-Mg and (**b**) TAT-2-058 in descending order of grain U–Pb age. Error bars represent 2σ , and most U–Pb age errors are within the size of the symbol or smaller.

5. Discussion

5.1. Thermal History of the Tsushima Lapilli Tuff

The Ugetsuiwa sample (TAT-2-058) contained remarkably old zircon grains that originated from xenocrysts that were all much older than the age of the tuff (1.9 Ga and 522 Ma) based on the U–Pb age data. The Mogi sample (TSU-Mg) also contained remarkably old zircon grains that originated from xenocrysts that were all much older than the age of the tuff in the U–Pb age data (356 Ma and 130 Ma). The presence of these old grains indicates that there were no thermal events above the closure temperature of U–Pb dating (900 °C; [44]), and these ages are considered to represent the rock formation ages because it is unrealistic to suggest that thermal events induced by tectonics could exceed 900 °C, close to the temperature of magma.

However, grain No. 4 in TSU-Mg yielded a U–Pb age of 370 Ma, whereas the FT age was 11.4 Ma, younger than the U–Pb age. Therefore, a thermal event is considered to have caused incomplete FT age rejuvenation, because many FT ages coincide with the U–Pb age.

In contrast, the FT age data of TAT-2-058 contained grains discordant in U–Pb age, but the grains were regarded as a single population showing 16.6 ± 1.0 Ma as the mean age. This mean age included two old zircon grains considered detrital grains or xenocrysts in the U–Pb age, but their FT ages were indistinguishable from those of the other grains. Therefore, those two grains were likely rejuvenated because the temperature reached a level high enough to cause rejuvenation during the tuff formation or later. Therefore, the FT age of TAT-2-058 (16.6 ± 1.0 Ma) can be also regarded as the eruption age.

The heat source that caused this rejuvenation was investigated. Granite, rhyolite, dacite and dolerite are known as intrusive rocks on Tsushima Island. The K–Ar age of the granite is between 19 and 13 Ma [45], and the Pb–Zn mineralization age is ca. 15 Ma [32]. The K–Ar ages of the rhyolite and dacite are between 19.0 and 14.1 Ma [45]. The age of the dolerite is unknown. The folding structure on Tsushima Island is part of the Taiwan–Shinji Fold Belt (e.g., [29]), and its formation age is considered to be from 13.6 Ma onward, based on the planktonic foraminifera in a core obtained off the San-in district [46]. The distribution of granite has been deformed by the folding structures, indicating that these folding structures on Tsushima Island formed after granite intrusion (Figure 2).

However, dolerite cuts the folding structures, as well as the dacite and rhyolite [17,31]. Therefore, the folding structures were formed after the granite intrusion but before the dolerite intrusion. In addition, the Taishu Group has been strongly affected by diagenesis (late diagenetic zone–low anchizone; ≥ 200 °C), based on illite crystallinity in the mudstone [28]. Therefore, the heat source that caused rejuvenation of the FT age is considered to be multi-factorial because of the formation of cleavage parallel to the folding axes, dolerite intrusion and acidic volcanic rock intrusion related to the granite. If the temperature of the thermal event was approximately 200 °C, several million years would be required for annealing to occur (e.g., [47]). Therefore, there is a possibility that the Taishu Group was subjected to this heat for several million years after deposition.

5.2. Age of Tsushima Lapilli Tuff and Submarine Volcanism in the Southwestern Japan Sea

In this section, the age of the Tsushima Lapilli Tuff and submarine volcanism in the southwestern Japan Sea are discussed. The FT and $^{238}U-^{206}Pb$ ages represent the eruption age of the Tsushima Lapilli Tuff. Therefore, 16.2 ± 0.7 Ma, the weighted mean age of these four ages, is considered to be the eruption age of the Tsushima Lapilli Tuff, determined for the first time, and is regarded as reliable.

Based on the age obtained by this study, the Tsushima Lapilli Tuff can be compared with the Okinoshima Formation and its thick (>200 m) subaqueous pyroclastic rock formed by acidic submarine volcanism, because the ages coincide with 16.2 ± 0.4 Ma [6]. This result shows that the Tsushima Lapilli Tuff and the subaqueous pyroclastic rock in the Okinoshima Formation represent submarine volcanism of 16.2 ± 0.6 Ma in the southwestern Japan Sea. Based on the previously obtained age of the Taishu Group [5,6] and the tectonism of the opening of the Japan Sea (e.g., [23,48,49]), rapid subsidence associated with submarine volcanism is considered to have occurred in the southwestern Japan Sea, including the Genka Sea region. As a result, the Paleo-Tsushima Basin was formed, and northern Kyushu separated from the Eurasian continent between 17.9 Ma and 16.2 Ma. This result provides a new constraint to determine the formation age of the Paleo-Tsushima Basin.

5.3. Volcanism Change in the Southwestern Japan Sea

Volcanism in northeastern Japan during the initial stages of the formation of the Japan Sea was characterized by bimodal volcanism originating from asthenospheric upwelling and lateral propagation (e.g., [48–51]). However, Ninomiya et al. [6] proposed that the volcanism in the southwestern Japan Sea region was characterized by felsic volcanism formed by the melting of existing old continental crust, because older inherited zircon grains with ages of 180 Ma and 2.0 Ga are contained in submarine lava [5]. These older inherited zircon grains indicate that the melting of existing old continental crust resulted from the occurrence of asthenospheric upwelling, as observed in the Izu–Bonin Arc [52]. Consequently, felsic magma was formed and caused felsic volcanism between 17.9 and 16.2 Ma in the southwestern Japan Sea region.

However, the volcanism is considered to have changed from felsic volcanism that formed granitic rocks to mafic volcanism, based on the presence of dolerite cutting the Taishu Group and acidic rocks [17–19,31]. Basaltic rocks formed by mafic volcanism between 16 Ma and 15 Ma have been reported from Iki Island and the Yobuko intrusive rocks distributed on the Higashi-Matsuura Peninsula [13,53,54].

In contrast, dacite and rhyolite from the same period are known from the Tsushima and Hirado districts. The age of the Hirado Dacite is between 15.5 and 14.5 Ma [13], and the youngest age of rhyolite and/or dacite on Tsushima Island is 14.1 Ma [45]. Therefore, the distinct mafic volcanism is considered to have started from ca. 15 Ma, but bimodal volcanism occurred between 15.5 and 14.1 Ma in the southwestern Japan Sea.

As a result of the termination of asthenospheric injection, the felsic magma began to cool and become exposed at the crustal surface as granite rhyolite on Tsushima Island. Consequently, felsic volcanism related to the opening of the Japan Sea is considered to have terminated at ca. 14 Ma. There is a high probability that the Tsushima district and its surrounding area changed to a compression-dominated regime, resulting in ascension of the granitic rocks, because of uplift associated with collision and shortening deformation caused by commencement of subduction of the Philippine Sea Plate, which occurred between 15.3 and 12.3 Ma [55]. The sedimentary environment also changed to upward shallowing because of the change to compression, based on the finding that shallowmarine rocks are contained in the Upper Unit of the Taishu Group and its equivalent [5,56]. The small-scale dolerites are considered to be the intrusive rocks from 13.6 Ma onward, because they cut the folding structures formed from 13.6 Ma onward [44]. Therefore, the change from felsic volcanism to mafic volcanism occurred from 14 Ma onward. Based on the geochemistry of the volcanic rocks from 15.3 Ma onward in the Chugoku region, these volcanic rocks are heterogeneous and originated from melting of subarc lithospheric mantle that had been metasomatized by subduction-related fluids [57]. Thus, the dolerites in the Tsushima district may have originated from melting of subarc lithospheric mantle metasomatized by subduction-related fluids as well as volcanic rocks in the Chugoku region. Southwestern Japan, including the Tsushima district, existed in a compression field because of the commencement of subduction of the Philippine Sea Plate [55]. Therefore, lithospheric mantle may have actively moved upward, independent of surface tectonism, at ca. 14 Ma, in addition to the basaltic activity that started from ca. 9 Ma in northern Kyushu (e.g., [13]). The volcanism in the southwestern Japan Sea has been proposed to have been dominated by mafic volcanism between ca. 14 Ma and 4.3 Ma, because the initial age of the Iki Volcano Group, mainly composed of basalt accompanied by andesite and rhyolite, is considered to be 4.3 Ma [54]. This volcanism change is important to understand the volcanism in the southwestern Japan Sea.

6. Conclusions

- (1) The Tsushima Lapilli Tuff was affected by thermal annealing, which caused rejuvenation of the FT age on some grains after deposition.
- (2) The age of the Tsushima Lapilli Tuff was determined as the mean of 16.2 ± 0.6 Ma.

- (3) Rapid subsidence occurred between 17.9 Ma and 16.2 Ma in the southwestern Japan Sea and was accompanied by active felsic volcanism.
- (4) The volcanism in the southwestern Japan Sea is considered to have changed from felsic volcanism originating from melting old continental crust in the opening stage of the Japan Sea to mafic volcanism originating from small-scale lithospheric mantle upwelling at ca. 14 Ma.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10 .3390/geosciences11090363/s1, Table S1: LA-ICP-MS setup and operating conditions for LA-ICP-MS dating of zircon, Table S2: Results of LA-ICP-MS FT datings of TSU-Mg, Table S3: Results of LA-ICP-MS FT datings of TAT-2-058. Table S4: grain data of LA-ICP-MS FT dating for sample TSU-Mg and the Tardree Rhyolite standard, Table S5: grain data of LA-ICP-MS FT dating for sample TAT-2-058, Table S6: result of U–Pb dating of TSU-Mg, Table S7: result of U–Pb dating of TAT-2-058.

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