

**Reconstruction of the eruptive history of Usu volcano,  
Hokkaido, Japan, inferred from petrological correlation  
between tephras and dome lavas**

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## **Abstract**

Usu volcano has erupted nine times since 1663. Most eruptive events started with an explosive eruption, which was followed by the formation of lava domes. However, the ages of several summit lava domes and craters remain uncertain. The petrological features of tephra deposits erupted from 1663 to 1853 are known to change systematically. In this study, we correlated lavas with tephtras under the assumption that lavas and tephra samples from the same event would have similar petrological features. Although the initial explosive eruption in 1663 was not accompanied by lava effusion, lava dome or cryptodome formation was associated with subsequent explosive eruptions. We inferred the location of the vent associated with each event from the location of the associated lava dome and the pyroclastic flow deposit distribution and found that the position of the active vent within the summit caldera differed for each eruption from the late 17th through the 19th century. Moreover, we identified a previously unrecognized lava dome produced by a late 17th century eruption; this dome was largely destroyed by an explosive eruption in 1822 and was replaced by a new lava dome during a later stage of the 1822 event at nearly the same place as the destroyed dome. This new interpretation of the sequence of events is consistent with historical sketches and documents. Our results show that petrological correlation, together with geological

evidence, is useful not only for reconstructing volcanic eruption sequences but also for gaining insight into future potential disasters.

**KEYWORDS:** eruption sequence, lava dome, tephra-lava correlation, Usu volcano, whole-rock composition

## 1. INTRODUCTION

The sequence of an eruptive episode can provide important information on the structure of the magma system (e.g. Pallister, Hoblitt, Meeker, Knight, & Siems, 1996; Takahashi & Nakagawa, 2015) and its eruptive mechanism(s) (e.g. Gurioli, Houghton, Cashman, & Cioni, 2005; Pioli et al., 2008). In addition, detailed reconstruction of previous eruptive sequences of a volcano provides useful information for forecasting future events and mitigating volcanic hazards (e.g. Andreastuti, Alloway, & Smith, 2000; Andronico & Cioni, 2002; Orsi, Di Vito, & Isaia, 2004). For example, explosive (e.g. Plinian) volcanic eruptions are often accompanied by pyroclastic flows and the extrusion of lava domes (e.g. Mt. St Helens 1980 eruption, Washington, Christiansen & Peterson, 1981; Mt. Vesuvius AD 79 eruption, Italy, Lirer, Munno, Petrosino, & Vinci, 1993; Mt. Pinatubo 1991 eruption, Philippines, Wolfe & Hoblitt, 1996). The sequence of explosive eruptions can be reconstructed by investigating medial to distal tephra layers (e.g. Hildreth, Lanphere, & Fierstein, 2003; Fierstein, Hildreth, & Calvert, 2011). However, it can be difficult to correlate proximal deposits, such as lava domes, flows, and agglutinates, with tephra layers because the proximal products are usually covered by younger eruptive materials and are often modified by subsequent explosive events

(e.g. Soufriere Hills volcano, Montserrat, Young et al., 1998; Bezymianny volcano, Kamchatka, Girina, 2013).

Several recent studies have shown that petrological features differ among independent eruptive events, that is, events that are separated by considerable periods of dormancy (e.g. Mt. Somma-Vesuvius, Italy, Santacroce et al., 2008; Tarawera volcano, New Zealand, Shane, Smith, & Nairn, 2008; Mt. Etna, Italy, Viccaro & Cristofolini, 2008; Tarumai volcano, Japan, Nakagawa, Hiraga, & Furukawa, 2011; Yotei volcano, Japan, Uesawa, Nakagawa, & Umetsu, 2016). Thus, it should be possible to use the distinctive petrological features of each eruptive event to establish correlations between not only proximal and distal eruptive products but also explosive and effusive products.

Usu volcano (Figures 1 and 2) has erupted nine times since 1663 (Nakagawa, Matsumoto, Tajika, Hirose, & Ohtsu, 2005). Each eruption produced tephra deposits, including pumice falls and pyroclastic flows. However, the sequence of activity during each eruptive event has not yet been clearly established because the many lava domes and craters present on the volcanic edifice have not been correlated with specific distal or medial tephra layers. Because a large population resides in the area around the volcano, it is important to reconstruct the detailed sequence of each historical eruptive event and clarify the similarities and differences among them to mitigate volcanic

hazards of the next eruption.

We have previously shown that the petrological features of the eruptive materials, mainly tephra, are distinctive among the historical eruptions of Usu volcano (Matsumoto & Nakagawa, 2010) (Table 1). In this study, we established correlations between tephra layers and the lava domes that compose the volcanic edifice. Then, on the basis of our petrological correlations and other geological evidence, we reconstructed the eruptive history of Usu volcano, including the sequence and mode of each eruption.

## **2. GEOLOGY AND ERUPTIVE HISTORY OF USU VOLCANO**

Usu volcano is a post-caldera volcano of Toya caldera (*ca* 110–120 ka; Okumura & Sangawa, 1984). The edifice comprises a small stratovolcano with a summit caldera (*ca* 1.8 km in diameter) and many lava domes and cryptodomes (Figures 1 and 2). The stratovolcano edifice was formed by eruptions of basaltic and basaltic andesitic magmas during *ca* 20–10 ka (Soya, Katsui, Niida, Sakai, & Tomiya, 2007). Following a sector collapse and formation of a summit caldera *ca* 7 ka, the volcano was dormant until 1663, when the volcano resumed activity with an explosive eruption of silicic material. The

subsequent eruptive history of Usu volcano, which has been reconstructed using records in historical documents as well as by geological investigations (Oba, 1966; Yokoyama, Katsui, Oba, & Ehara, 1973; Nakagawa et al., 2005; Nakamura, Matsumoto, & Nakagawa, 2005; Soya et al., 2007), shows that it has erupted eight more times since 1663; four times during the 17th–19th centuries and four times in the 20th century, with the latest eruption in 2000 (Table 1).

Four domes have been recognized in the summit caldera: Kousu lava dome (557 m a.s.l.), Usu-shinzan cryptodome (669 m a.s.l.), Ogariyama cryptodome (672 m a.s.l.), and Ousu lava dome (733 m a.s.l.) (Soya et al., 2007) (Figures 1 and 2b). The Showa-shinzan lava dome and many cryptodomes (e.g. Nishiyama, Kompira Yama, Meiji-shinzan, and Higashi-maruyama cryptodomes) are located on the flanks of the volcano and at its foot (Figures 1 and 2a). A small crater (*ca* 1 km in diameter) is situated in the southwestern area within the summit caldera in addition to craters formed by the 1977–1978 eruptive events (Figure 1).

The sequence of the 20th century eruptions is known: each event began with an explosive phase and was followed by the formation of one or more lava domes or cryptodomes (Hokkaido Government, 1918; Yokoyama et al., 1973; Katsui et al., 1978; Katsui, Yokoyama, & Murozumi, 1981; Kadomura, Okada, & Araya, 1988; Soya et al.,

2007; Yokoyama & Matsushima, 2018). With regard to the pre-20th century historical eruptions, several historical documents and geological investigations suggest that the eruptive events usually began with a Plinian eruption accompanied by pyroclastic flows or surges (Hokkaido Government, 1918; Yokoyama et al., 1973; Soya et al., 2007). However, information about the active areas and the formation of lava domes is scant. During the 1853 eruption, however, the explosion and extrusion of the Ousu lava dome are recorded to have occurred within the summit caldera on the east side.

### **3. PREVIOUS INTERPRETATIONS OF THE FORMATION AGES OF SUMMIT LAVA DOMES AND CRATERS**

Kousu, Ousu, and Ogariyama domes were previously attributed to particular eruptions during the 17th–19th centuries (Yokoyama et al., 1973; Soya et al., 2007) (Table 1). Several historical accounts indicate that Ousu lava dome formed in 1853, after an explosive eruption (e.g. Hokkaido Government, 1918). However, the formation ages of the other two lava domes are still unclear. Two historical sketches of Usu volcano (Kono, 1918) (Figure 3) have been used previously to estimate the formation ages of lava domes. In an 1855 sketch of the volcano viewed from its southern foot, two lava domes

are shown on the summit (Figure 3a). Yokoyama et al. (1973) interpreted the eastern (right) and western (left) domes in the sketch as Ousu and Kousu lava domes, respectively. In contrast, in the sketch drawn in 1799 only one lava dome is shown on the western summit; this dome was thought to be the smaller of the two domes in the 1855 sketch (Figure 3b) and was therefore interpreted as Kousu dome. On the basis of these sketches, therefore, Kousu lava dome was inferred to have been extruded in either 1663 or 1769. In addition, some studies have suggested on the basis of its petrographical features that Kousu dome may have formed during the 1769 eruption (Tomiya & Takahashi, 2005; Soya et al., 2007; Tomiya, 2008). The formation age of Ogariyama cryptodome is not well established. In the historical record, a “small hill” was reported near the newly formed Ousu lava dome by a climber several decades after the 1853 eruption (Ishikawa, 1890). Soya et al. (2007) suggested that Ogariyama was a cryptodome formed during the 1822 eruption because it is partly covered by the 1853 Ousu lava dome (Table 1).

In addition, Kato (1910) identified a crater formed by the 1853 eruption, which he called the “Tachiiwa explosion crater”, on Ousu lava dome. He and other researchers also recognized the small crater in the southwestern area of the summit caldera (Kato, 1910; Yokoyama et al., 1973; Soya et al., 2007). Yokoyama et al. (1973) speculated that

this southwestern crater was formed by the 1663 eruption, because Kousu lava dome, which they interpreted as a product of the 1663 eruptive event, covered it.

#### **4. PETROLOGICAL FEATURES OF JUVENILE MATERIALS**

The juvenile materials erupted from Usu volcano during the historical period (1663–2000) consist of low-K rhyolitic and dacitic rocks (Table 2). The compositional variation of each eruption is narrow; SiO<sub>2</sub> contents vary by less than 2 wt% in a single eruptive event. However, there exist systematic compositional differences between the eruptions, and the silica content in particular decreases over time (Nakagawa et al., 2005; Figure 4). Mineral compositions of the 1663 juvenile products show a bimodal variation, and the compositions of juvenile samples from the pre-1769 (around AD 1700) and later eruptions become wide and are mainly intermediate between the two compositional modes of the 1663 juvenile materials (Tomiya & Takahashi, 1995, 2005; Matsumoto, Nakamura, & Nakagawa, 2005; Tomiya, Takahashi, Furukawa, & Suzuki, 2010; Matsumoto & Nakagawa, 2010).

On the basis of these features, magma mixing has been interpreted as the main magmatic process during Usu historical activity, and the compositions of the juvenile

materials of the pre-1769 and later eruptions have been inferred to reflect the mixing of two end-member magmas (e.g. Tomiya & Takahashi, 1995, 2005). We recently showed, however, that the petrological features (phenocryst content, mineral assemblage, and geochemistry) of juvenile materials collected from tephra deposits of each historical eruption are distinctive (Matsumoto & Nakagawa 2010) (Figure 4; Table 2). Moreover, we divided the juvenile materials of the eruptions into three magmatic groups, each characterized by a distinctive silicic magma, as follows: 1663 eruption (Group 1), nearly aphyric, hornblende-augite-bearing hypersthene rhyolite; pre-1769 to 1853 eruptions (Group 2), porphyritic, augite-hornblende-quartz-bearing hypersthene rhyodacite; and 1943–1945 to 2000 eruptions (Group 3), slightly porphyritic, augite-bearing hypersthene dacite. The whole-rock chemistry also differs among these three magmatic groups, with each group showing a different linear trend in Harker variation diagrams (Figure 4). We interpreted these features to indicate that the materials from the Usu historical eruptions were indeed produced by the mixing of two magmas but that the end-member magmas differed among the three magmatic groups.

## **5. MATERIALS AND METHODS**

To determine the formation sequences of the craters and lava domes of Usu volcano, we re-examined the topographic features of the summit area using topographic maps and aerial photos from the Geospatial Information Authority of Japan (GSI) and a Red Relief Image Map (Asia Air Survey Co. Ltd.; Chiba, 2011) (Figure 5). In addition, we used an aerial photo taken in 1967 and a topographic map produced in 1975 (Figure 6) to identify the topographical features before the 1977–1978 eruptive activity, which caused significant topographic changes, including the formation of craters and cryptodomes within the summit caldera.

We systematically collected 50 samples from the lava domes within the summit caldera for petrological analysis and correlation with tephra (Figure 5b). In addition, we used data of tephra samples and the 1943–1945 dome lava samples reported by Matsumoto & Nakagawa (2010). We made thin sections of all 50 samples for microscopic observation. Representative samples were used to calculate phenocryst modes by point counting, at least 3000 points per thin section. The tephra data from eruptive activity during the 18th–19th centuries were previously obtained by point counting on a vesicle-free basis (Matsumoto & Nakagawa, 2010). For less porphyritic rocks (1663, pre-1769, 1977, and 2000 pumices), we first separated phenocrysts from crushed samples, then weighed them and used mineral density information to convert

the weights to volume (Smyth & McCormick, 1995). Whole-rock compositions were determined by X-ray fluorescence (XRF) using Philips PW1404 and Spectris MagiX PRO systems with a Rh tube. Major elements were determined by analyzing fused glass beads, which were produced using a 1:2 glass to flux ratio. The detailed XRF analysis method and the results of replicate analysis is summarized in Supplementary data (Doc. S1 and Table S1). Representative modal and whole-rock compositions of the dome lavas are given in Table 2 along with data on the erupted tephras (Matsumoto & Nakagawa, 2010).

## **6. RESULTS**

### **6.1 Topography of the summit area**

As previously recognized, Kousu lava dome, Usu-shinzan cryptodome, Ogariyama cryptodome, and Ousu lava dome are in the summit caldera (Soya et al., 2007) (Figures 2b and 5). In addition, small craters that formed during the 1977–1978 eruptive activity have been identified in the north-central and south-central areas of the summit caldera (Soya et al., 2007) (Figure 5). A relatively large, previously identified (Kato, 1910;

Yokoyama et al., 1973; Soya et al., 2007) crater in the southwestern area (SW crater) is partly covered by Kousu lava dome. By examining the Red Relief Image Map (Chiba, 2011), we newly recognized a small crater in the southeastern area (SE crater), which is partly covered by Ousu lava dome (Figure 5a). This crater might be the “Tachiiwa explosion crater” described by Kato (1910).

By examining the 1967 aerial photo and the 1975 topographic map, we investigated the topography in the summit caldera before the formation of the Usu-shinzan cryptodome during the 1977–1978 activity (Figure 6). Soya et al. (2007) inferred that Ogariyama dome was exposed by the 1977–1978 eruptive activity, but the dome is clearly visible, though partly covered by Ousu lava dome, in the central part of the summit caldera both in the photo and on the map. We speculate, therefore, that Ogariyama dome, hitherto considered a cryptodome, might have been extruded partially.

In addition, we recognized a small crater-like depression in the central area (called "C crater" hereafter, Figure 6b) that cuts both the SE crater and Ogariyama dome and is partly covered by Ousu lava dome. Thus, three obvious craters were present before the 1977–1978 eruption, but their formation ages are as yet unknown.

## **6.2 Petrography and geochemistry of dome lavas and their correlations with tephras**

As described in section 2, records show that, at least since the 1853 eruption, each explosive eruption of Usu volcano was accompanied by the formation of a lava dome or cryptodome (Table 1). The presence of lava domes that formed before the 1853 eruption strongly suggests that the earlier explosive eruptions may also have been followed by lava effusion. In addition, the petrological features of tephras from each historical eruption and those of the 1943–1945 dome lava are distinctive (Matsumoto & Nakagawa, 2010). This fact suggests the possibility that the petrological features of dome lavas may resemble those of tephra from the same eruption and that the juvenile materials of each independent eruption may be distinctive. Therefore, under the assumption that petrographical and geochemical features of tephras and related dome lavas are similar, we attempted to correlate each lava dome in the summit caldera with a historical eruption.

Many historical explosive volcanic eruptions were caused by magma injection into a pre-existing chamber, and the produced materials reflect the mingling or mixing of two magmas: for example, Vesuvius (Italy) (Cioni et al., 1995), Lassen Peak (United

States) (Clynne, 1999), and Merapi (Indonesia) (Costa, Andreastuti, de Maisonneuve, & Pallister, 2013) volcanoes. Because of this mixing, juvenile materials of explosive eruptions can exhibit widely varying petrographical and geochemical features. However, dome lavas produced after explosive eruptions are usually more homogeneous, indicating that magma was input into the chamber just once, before the eruption or during its early stage, and that homogenization of the two magmas continued throughout each eruptive episode (e.g. Redoubt, Alaska, 1989–1990 eruption, Wolf & Eichelberger, 1997; Shinmoe-dake, Japan, 2011 eruption, Nakada, Nagai, Kaneko, Suzuki, & Maeno, 2013; Suzuki et al., 2013). This situation might make it difficult to correlate pyroclastic deposits with dome lavas from the same eruptive episode. In the case of Usu volcano, however, tephra samples from a single deposit are fairly homogeneous, with less than 2 wt% variation in SiO<sub>2</sub> (Matsumoto & Nakagawa, 2010) (Figure 4; Table 2). These findings suggest that Usu historical magmas were already well mixed before each eruption began, and it is therefore plausible that the petrographic and geochemical features of dome lavas and related tephtras might be similar.

The phenocryst contents and mineral assemblages of the dome lavas from the summit caldera mostly agree with those of tephra samples from the 18th and 19th

century eruptions (Figure 7; Table 2). In addition, the whole-rock chemistry of the dome lavas is consistent with the compositional trends of tephra erupted during the 18th and 19th centuries (Figure 8). Thus, on the basis of their whole-rock chemistry and petrographical features, we correlated each dome lava with a tephra from a single eruption, as described below (Table 3).

### **6.2.1 Ousu dome lava**

The Ousu dome lava samples are porphyritic (13.0–19.1 vol% phenocrysts) (Figure 7a; Table 3). Phenocrystic minerals are plagioclase, orthopyroxene, and Fe-Ti oxides, and minor phenocrystic phases are absent. The phenocrysts sometimes form large crystal clots 1–2 cm long (Oba, 1989; Tomiya & Takahashi, 1995). Comparison with temporal changes in the whole-rock SiO<sub>2</sub> compositions of tephra samples shows that the SiO<sub>2</sub> contents of Ousu lava (70.8–71.1 wt%) are within the ranges of tephra samples from the 1822 and 1853 eruptions (Figure 8). The TiO<sub>2</sub> and P<sub>2</sub>O<sub>5</sub> contents of Ousu lava samples seem to be slightly higher than those of other dome lavas, whereas Ousu lava samples overlap those of central Kousu (see below) with respect to several elements. In addition, they resemble the tephra samples from the 1853 eruption more than those from the 1822 event (Figure 9). It is also clear from historical records that Ousu dome was formed

during the 1853 eruptive activity (Hokkaido Government, 1918; Yokoyama et al., 1973). These findings suggest that minor but clear differences such as these might also distinguish other historical eruptive events of Usu volcano.

### **6.2.2 Ogariyama dome lava**

The Ogariyama lava samples are porphyritic (13.0–15.8 vol% phenocrysts) (Figure 7b; Table 3). This feature suggests that Ogariyama dome was not formed during the 1663 eruption. All of the samples have plagioclase, orthopyroxene, and Fe-Ti oxides as the dominant phenocrystic phases, and they lack minor phenocryst phases such as hornblende and quartz, unlike tephra samples from eruptions during the 18th and 19th centuries (Table 3). As in the case of Ousu dome lava, large crystal clots are often found (Figure 7b). In the whole-rock chemistry of the Ogariyama lava samples, the silica content (71.6–72.0 wt%) is consistent with that of tephra samples from the 1769 event (Figure 8). Ogariyama lava can be distinguished from lavas of the other domes on plots of various elements against SiO<sub>2</sub>, and they are similar in composition to the 1769 tephra samples, although their P<sub>2</sub>O<sub>5</sub> content is slightly lower than that of the tephra samples (Figure 9). Considering these features, we suggest that Ogariyama dome was extruded during the 1769 event.

### 6.2.3 Kousu dome lava

Although the topography of Kousu lava dome is simple and it has been considered to have formed during a single historical eruption, lava samples from the northeastern part of the dome are less porphyritic (10.8–13.0 vol%) than samples from the central part (14.9–22.4 vol%) (Figures 7c, d; Table 3). In addition, although the major phenocrystic minerals are plagioclase, orthopyroxene, and Fe-Ti oxides in all samples, the northeastern and central lavas differ with respect to their minor phenocrystic minerals. Hornblende with a reaction rim, clinopyroxene, and quartz are found in the northeastern lavas, whereas only quartz is present in the central lavas (Figures 7e, f). Thus, on the basis of the minor phenocryst assemblages, we correlate the northeastern lava with the pre-1769 or 1769 tephra and the central lava with the 1822 or 1853 tephra (Table 3).

The northeastern and central lavas of Kousu lava dome can also be distinguished on the basis of their whole-rock chemistry. The SiO<sub>2</sub> content of the central lavas (70.5–71.6 wt%) is similar to that of the 1822 and 1853 tephra samples, whereas the northeastern lavas have a higher SiO<sub>2</sub> content (72.4–72.7 wt%), which corresponds to that of the pre-1769 and 1769 tephra samples (Figure 8). On variation plots of most elements against SiO<sub>2</sub> (Figure 9), the central lavas resemble the 1822 or 1853 tephra samples,

whereas the northeastern lavas are associated with pre-1769 or 1769 tephra samples. Under the assumption that only one lava dome was built during each eruptive event, we propose the following interpretation. Taking into account the minor phenocryst assemblage and the lower phenocryst content of the northeastern lava samples (Figures 7d, f; Table 3), we suggest that they are pre-1769 rather than 1769 juvenile materials (Figure S1). In addition, the central lavas are distinguishable from those of Ousu dome, which was extruded during the 1853 eruptive event. Therefore, we conclude that Kousu lava dome is composed of two distinct dome lavas, one extruded during the pre-1769 eruption and the other during the 1822 eruption.

## **7. DISCUSSION**

### **7.1 Finding a destroyed lava dome: Old Kousu lava dome**

As described in section 6.2.3, northeastern Kousu lava dome samples share petrological features with the pre-1769 tephra, whereas the samples from the central part of this dome resemble those of the 1822 tephra. This result suggests that Kousu dome did not form, as inferred previously, during either the 1663 (Yokoyama et al., 1973) or 1769

(Soya et al., 2007; Tomiya, 2008) events. The Usu-shinzan cryptodome, which formed in the central area of the summit during 1977–1978 (Kadomura et al., 1988), cuts Kousu lava dome (Figures 1–3). In addition, in the 1967 aerial photo and 1975 topographic map (Figure 6), lava is visible on the northeastern side of Kousu lava dome and forms the lower part of the dome. This lava may correspond to lava extruded during the pre-1769 event and subsequently covered by lava with petrological features similar to those of the 1822 tephra. Thus, we speculate that Kousu lava dome is a composite lava dome that was modified by the uplifting of a cryptodome (Usu-shinzan dome) during the eruptive activity of 1977–1978.

On the basis of the results of our petrological investigation and topographic evidence, we propose that the Kousu lava dome actually consists of two domes, which we call Old Kousu and New Kousu lava domes, and that these domes formed during the pre-1769 and the 1822 activity, respectively. This interpretation is consistent with the historical record. The two lava domes on the sketch drawn in 1855 (Figure 3a) shows two lava domes, which were previously interpreted as the Ousu (on the right) and Kousu (left) domes. Because Ousu dome formed during the 1853 event, in the 1855 sketch, it shows vigorous fumarole activity. However, the dome on the left, which has been presumed to be Kousu lava dome, is also drawn as emitting fumarolic gases. If as

previously supposed, Kousu lava dome formed in 1663 (Yokoyama et al., 1973) or 1769 (Soya et al., 2007; Tomiya, 2008), the sketch implies that fumarole activity continued for 192 or 86 years, respectively. Observations made in the 20th century indicate that Ousu lava dome, which was extruded during the 1853 eruptive event, had almost ceased fumarole activity by 1972 (Yokoyama et al., 1973). In addition, a fumarole with a height of several hundred meters was observed on the 1943–1945 Showa-shinzan lava dome in 1971 (Yokoyama et al., 1973), but since then fumarole activity on that dome has become weak. Considering these observations, a period of fumarole activity as long as 192 or 86 years might not be credible for Usu volcano. In our new interpretation, the left dome must be the New Kousu dome, which formed during the 1822 event; thus, the weak fumarole activity depicted from the left dome in the sketch is plausible, because in 1855 the New Kousu dome would have been just 33 years old.

The 1799 sketch depicts only one lava dome, without fumarole activity (Figure 3b). The position of this dome is similar to that of the New Kousu dome in the 1855 sketch. Thus, it is possible to interpret this dome as the Old Kousu dome, which we believe formed during the pre-1769 event. Nakagawa et al. (2005) estimated that the pre-1769 eruption occurred around AD 1700. Thus, it is reasonable that when the dome was drawn in 1799, no fumarole activity was seen. On the other hand, no dome

recognizable as Ogariyama dome is seen in the 1799 sketch. On the 1975 topographic map, Ogariyama dome is quite small (Figure 6b). Thus, it is likely that the person who drew the sketch in 1799 could not see Ogariyama dome from the southeastern flank of the volcano.

We interpret the petrological features and the two sketches from 1799 and 1855 to indicate that Old Kousu dome, which was extruded during the pre-1769 eruption, was replaced almost entirely by New Kousu lava dome at nearly the same place in 1822. Thus, Old Kousu dome was destroyed by the 1822 eruption.

## **7.2 Re-examination of the five eruptions during the 17th–19th centuries**

Considering together the petrological correlations, historical records, and lava dome structures, we inferred that four lava domes were formed by the eruptions during the 18th and 19th centuries (Table 1). Taking into account the revised correlations of the lava domes, the topography within the summit caldera, and the distribution of pyroclastic flows, we re-examined the eruption sequence of each eruptive event during the 17th-19th centuries (Figure 10). In addition, from the distributions of pyroclastic flows and surge deposits (Yokoyama et al., 1973; Katsui et al., 1981, Katsui, Kouchi, &

Niida, 1988; Soya et al., 2007; our unpublished data), we inferred the locations of the craters from which they originated.

### **7.2.1 1663 eruption**

The 1663 eruption, the largest from Usu volcano during the historical period, began as a phreatomagmatic eruption, and a small pyroclastic surge flowed eastward down the edifice (Nakamura et al., 2005). This activity was immediately followed by a Plinian eruption. After the climactic Plinian eruption, a voluminous base surge flowed out all around the volcano (Nakamura et al., 2005). Considering this eruptive history and the volume of the erupted material, it is reasonable to attribute the formation of the summit caldera to this event (Yokoyama et al., 1973; Soya et al., 2007). Although thick base surge deposits are found all around the volcano (Figure 10a), in our petrological analysis, we recognized no lava dome associated with this event. Information about the active crater during this eruption is poor. However, the summit caldera is closed, though debris avalanche deposits are distributed around the southern foot, indicating that the summit caldera was once open to the south. Therefore, the southern wall of the summit caldera might have been formed during the 1663 eruptive event.

### **7.2.2 pre-1769 eruption**

The pre-1769 eruption was much smaller than any of the other 17th to 19th century events. The eruptive activity started with a sub-Plinian eruption, which was followed by a pyroclastic surge (Nakagawa et al., 2005). Today, the surge deposit is found in six outcrops near the foot of the volcano on the northwestern side; no other outcrops in other locations have been reported (Nakagawa et al., 2005) (Figure 10b). Therefore, the pre-1769 pyroclastic surge must have flowed mainly northwestward from the active crater. Our petrological analysis showed that Old Kousu lava dome formed during this event at nearly the same location as New Kousu dome would later form. Although we cannot find topographic evidence for the pre-1769 vent, we speculate that it was in the northwestern area of the summit caldera because the surge deposits are only identified on the volcano's northwestern flank.

### **7.2.3 1769 eruption**

Previous studies showed that the 1769 eruption began as a sub-Plinian eruption, which was followed by pyroclastic flows (Yokoyama et al., 1973; Soya et al., 2007) (Table 1). Our petrological analysis indicated that Ogariyama dome formed during the 1769 activity. The location of this lava dome suggests that the active vent must have located

in the central or eastern part of the summit caldera. Although deposits of the 1769 pyroclastic flow are found all around the volcano, the deposits are thicker (up to ~50 cm) on the eastern side than on the southwestern side (~30 cm) according to our unpublished data (Figure 10c). In addition, Osarubetsu village, which was located at the foot of the volcano on the southeastern side, suffered severe damage in 1769 (Yokoyama et al., 1973) (Figure 10c). These data suggest that the pyroclastic flow emplacement direction in 1769 was mainly southeastward from the summit caldera. Thus, the active vent may have formed in the southeastern area of the summit caldera, and was subsequently destroyed or buried by the 1822 and 1853 eruptions.

#### **7.2.4 1822 eruption**

Pyroclastic flows during the 1822 event killed 103 people in Abuta town, on the southwestern flank of the volcano (Hokkaido Government, 1918; Yokoyama et al., 1973). Considering the direction of flows from the summit, it is plausible that the SW crater, which can be seen on the 1975 topographic map (Figure 6), was the source of these destructive pyroclastic flows (Figure 10d). This crater is the largest of those formed after the 1663 eruption. Our petrological investigation indicated that New Kousu dome was formed during this eruptive activity. New Kousu dome partly covers

the SW crater, so this interpretation is consistent with the topographic relationship between the two features (Figure 10d). In addition, the explosive eruption that formed the SW crater may have destroyed Old Kousu dome.

Historical records indicate that the 1822 event lasted for at least four months (Soya et al., 2007; Tomiya, 2008), so it is possible that two craters (or vents) might have formed during the eruptive event. We speculate that the central crater (C crater), which is visible in the 1967 aerial photo (Figure 6a), formed during the later period of the 1822 event, because C crater cuts both Ogariyama dome (1769) and the SW crater.

In summary, the explosions that formed first the SW crater and then the C crater mostly destroyed Old Kousu dome. Then dome lava was extruded near the site of Old Kousu lava dome, forming New Kousu lava dome. We speculate that contemporary witnesses did not notice that the lava dome had been rebuilt because the old and new domes were in almost the same location.

### **7.2.5 1853 eruption**

Ousu dome is widely recognized to have formed during the 1853 eruption (Hokkaido Government, 1918; Yokoyama et al., 1973). We newly recognized a small SE crater that is partly covered by Ousu dome (Figure 10e). Moreover, the pyroclastic flows

associated with this event are distributed mainly on the eastern flank of the volcano (Katsui et al., 1988). The eruptive sequence reported in the historical record (Hokkaido Government, 1918; Yokoyama et al., 1973) indicates that an explosion occurred in the eastern area of the summit caldera, and pyroclastic materials flowed eastward. Subsequently, a new lava dome, Ousu dome, formed. The historical record is thus consistent not only with the locations of the dome and the SE crater but also with the distribution of the pyroclastic flow deposits.

### **7.3 Summary of eruptive sequence of Usu volcano and its interpretation**

As a result of our examinations, the whole eruptive history of Usu volcano, including the eruptive activity before the 19th century, has been revealed. We can summarize the eruptive activity of Usu volcano as follows. (1) The eruption in 1663 produced the southern rim of summit caldera. The activity started with a Plinian eruption, followed by vigorous phreatomagmatic eruptions with base surges. No lava dome was extruded in this eruption. (2) The pre-1769 to 1853 eruptions occurred within the summit caldera. In each case, an explosive eruption accompanied by pumice fallout occurred first, and was followed by a pyroclastic flow and surge. The locations of the active vent(s) or

crater(s) that formed differed among these eruptions. During the later period of each eruptive event, a lava dome was built. (3) The eruptive activity during the 20th century, from 1910 to 2000, was characterized mainly by phreatic and phreatomagmatic eruptions on the volcano's flank. Only the 1977–1978 eruptive event occurred at the summit. The active areas were large, and many vents and craters were formed. One or more cryptodomes also formed during the 20th century eruptions; lava was extruded only during the late period of the 1943–1945 eruption. Interestingly, this grouping is consistent with the magmatic groups of Matsumoto & Nakagawa (2010) (Table 1). Thus, the eruption style might have changed as a result of the replacement of silicic magma. These relationships have important implications for the evaluation of the future activity and eruption hazards of Usu volcano as well as for understanding eruption processes more generally.

## **8. CONCLUDING REMARKS**

We were able to correlate dome lavas erupted from Usu volcano, Japan, during and after explosive eruptive events with pyroclasts on the basis of their whole-rock chemistry and phenocryst contents. By combining precise petrological measurements with topographic

analyses, we determined not only the formation age of several lava domes but also the locations of some associated craters. In addition, we were able to reinterpret the historical record and the distributions of eruptive materials during each event. We confirmed that during each eruptive event of Usu volcano, except that in 1663, the eruptive activity consisted of an explosive eruption followed by the formation of a lava dome or cryptodome. Moreover, petrological analyses allowed us to infer the existence of a hitherto unrecognized lava dome, Old Kousu lava dome, which was mostly destroyed by subsequent events. The formation ages of some cryptodomes on the edifice, such as Nishiyama, Kompira Yama, and Higashi-maruyama cryptodomes (Figures 1 and 2), which were already present before the 20th century (Yokoyama et al., 1973), are still unknown. In the future, the formation ages of these cryptodomes should be determined by a similar petrological analysis of samples from the domes. We emphasize that this petrological method can be used not only to improve our understanding of the overall eruptive history of the volcano but also to determine the detailed eruption sequence of each individual event.

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### Figure legends

**Figure 1.** (a) Locality map of Usu volcano. (b) Simplified geological map of Usu volcano showing the many lava domes and cryptodomes in the summit area and on the flanks of the volcano (modified from Soya et al., 2007).

**Figure 2.** Photographs of Usu volcano. (a) Photo taken looking south from Lake Toya. (b) Aerial photo looking northward. Abbreviations are defined in Figure 1. Many cryptodomes are found around the flanks and at the foot of the volcano, and three lava domes and one cryptodome are located within the summit caldera.

**Figure 3.** Sketches of Usu volcano drawn looking north from the southern foot of the mountain in (a) 1855 and (b) 1799 (Kono, 1918). In the 1855 sketch, two lava domes are shown, whereas only one is represented in the 1799 sketch. The two domes in the 1855 sketch were previously identified as Kousu (left) and Ousu (right) lava domes, and the dome in the 1799 sketch was assumed to be the same as the smaller dome in the 1855 sketch (i.e. Kousu dome).

**Figure 4.** Representative SiO<sub>2</sub> variation diagrams of whole-rock chemistry of historical

Usu tephra samples (Matsumoto & Nakagawa, 2010). Only the 1943–1945 samples were dome lavas (asterisk). Rocks from historical Usu eruptions exhibit three linear trends according to their eruptive age: 1663 (short-dash line), pre-1769 to 1853 (solid line), and 1943–1945 to 2000 (long-dash line). No juvenile tephra from the 1910 phreatic explosions have been identified.

**Figure 5.** (a) Red relief image map of Usu volcano taken in May 2005 (Chiba, 2011) showing the detailed topography. Crater walls (solid white lines) can be seen near the southeastern and southwestern rims of the summit caldera. SE, southeastern crater; SW, southwestern crater. Other abbreviations are defined in Figure 1. (b) Topographic map of the summit area (based on a 1:25,000 map in 2006 by the Geospatial Information Authority of Japan; GSI) showing the sampling points of this study.

**Figure 6.** (a) Aerial photo of the summit of Usu volcano taken in 1967 (from GSI). Abbreviations are defined in Figures 1 and 5. In 1967, Ogariyama dome (OGL) seemed to be exposed at the surface, suggesting that it might have been extruded partially. (b) Topographic map of Usu volcano made in 1975 (1:25,000 map by GSI). The small crater in the central area (C) cuts both the southwestern crater (SW) and Ogariyama lava

dome (OGL) and is partly covered by Ousu lava dome (OSL).

**Figure 7.** Photomicrographs of representative samples from dome lavas collected from the summit area. (a) Ousu dome lava (hypersthene dacite), (b) Ogariyama dome lava (hypersthene dacite), (c) central Kousu (quartz-bearing hypersthene dacite) and (d) northeastern Kousu dome lavas (hornblende-augite-quartz-bearing hypersthene dacite), (e) quartz phenocryst in central Kousu lava, and (f) hornblende phenocryst with a reaction rim in northeastern Kousu lava.

**Figure 8.** SiO<sub>2</sub> content in the whole-rock chemistry of dome lava samples collected at the summit in this study (left side) compared with that of tephra and lava (the 1943–1945 event only, marked by an asterisk) samples from each historical event (right side; data from Matsumoto & Nakagawa, 2010). There is no data for the 1910 eruption because of phreatic eruption. The SiO<sub>2</sub> contents of the Ousu and Ogariyama lavas are consistent with those of the 1853 and 1769 tephtras, respectively. The Kousu lava samples consist of high-SiO<sub>2</sub> lava from the northeastern part of the dome, similar in SiO<sub>2</sub> content to the pre-1769 and 1769 tephtras, and low-SiO<sub>2</sub> lava from the central part, similar to the 1822 and 1853 tephra samples.

**Figure 9.** SiO<sub>2</sub> variation diagrams based on the whole-rock chemistry of dome lava samples from the summit. The areas surrounded by solid or broken lines are the compositional ranges of tephra from the eruptions in the 18th and 19th centuries. Ousu, Ogariyama, northeastern Kousu, and central Kousu dome lavas are distinguished, especially by their TiO<sub>2</sub> content. The composition of the Ogariyama lava samples is consistent with that of the 1769 tephra; the northeastern Kousu lavas match the pre-1769 and 1769 tephra samples; and the central Kousu lavas are similar to the 1822 and 1853 tephra samples.

**Figure 10.** Explanations of the relationships among the lava domes, vent and crater locations, and pyroclastic flow distributions associated with the 17th to 19th century eruptions of Usu volcano: (a) 1663, (b) pre-1769 (around AD 1700), (c) 1769, (d) 1822, and (e) 1853. Pyroclastic flow distributions during the 17th–19th centuries (Katsui et al., 1988), along with additional geological data obtained herein, are also shown. Dots indicate outcrops of pyroclastic flows, and the accompanying number indicates the pyroclastic flow thickness in centimeters at each site. Stars show the vent location of each eruption, and triangles indicate lava domes present before the eruption. Arrows

indicate the main pyroclastic flow direction as inferred from historical records (Yokoyama et al., 1973) and geological observations. For all eruptions during the 17th–19th centuries, except the 1663 eruption, the vent locations are consistent with the main pyroclastic flow and surge directions.

### **Supporting Information**

**Figure S1.** Photomicrographs of representative tephra samples from eruptions during the 18th and 19th centuries: (a) 1853, (b) 1822, (c) 1769, and (d) pre-1769 (around AD 1700). Quartz phenocrysts are observed in tephra samples from the (e) 1853 and (f) 1822 eruptions, and hornblende phenocrysts in tephra samples from the (g) 1769 and (h) pre-1769 eruptions.

**Doc S1.** Sample preparation and analytical details for whole-rock chemistry. The results of replicate analysis are also listed in Table S1.

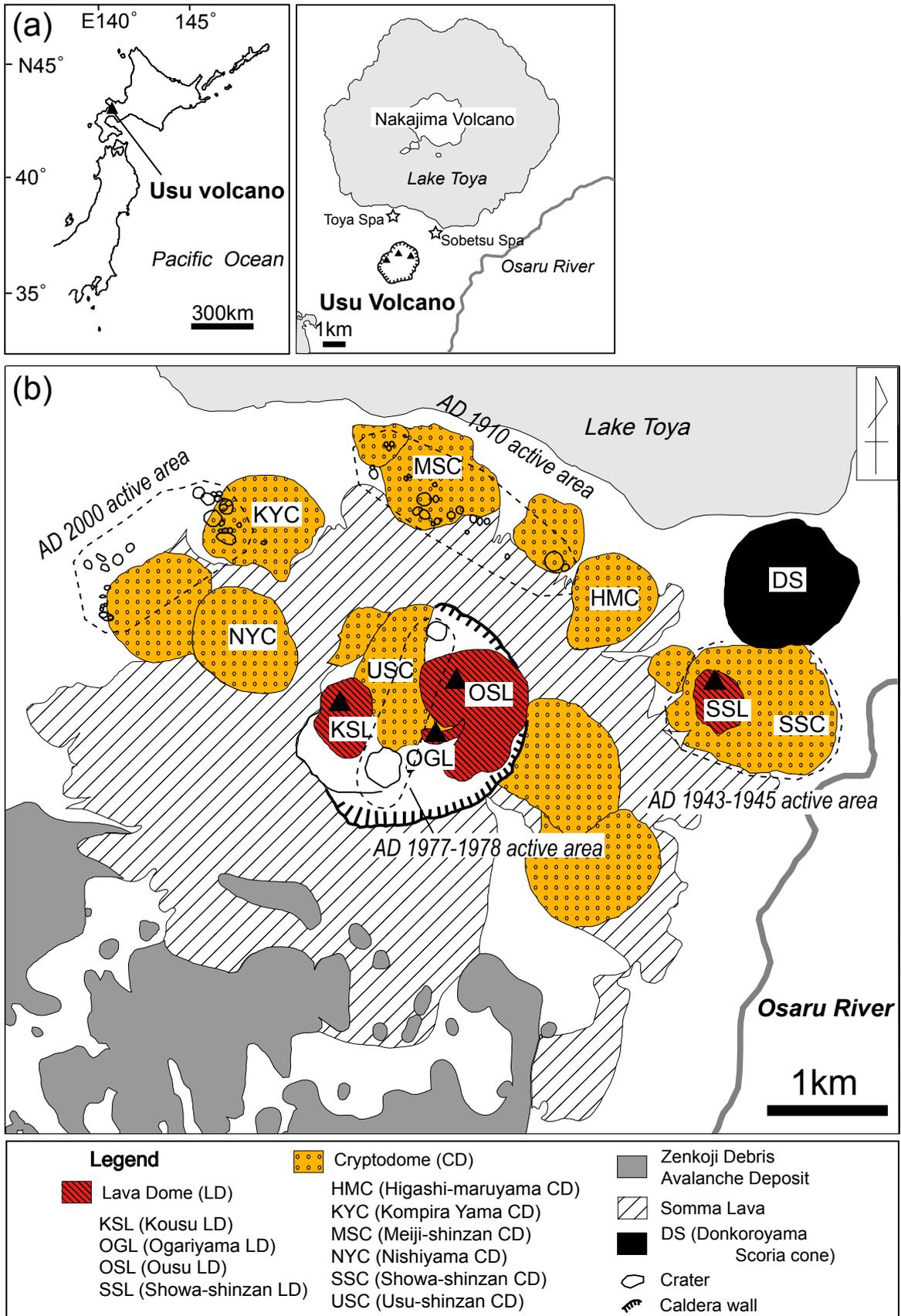


Fig.1

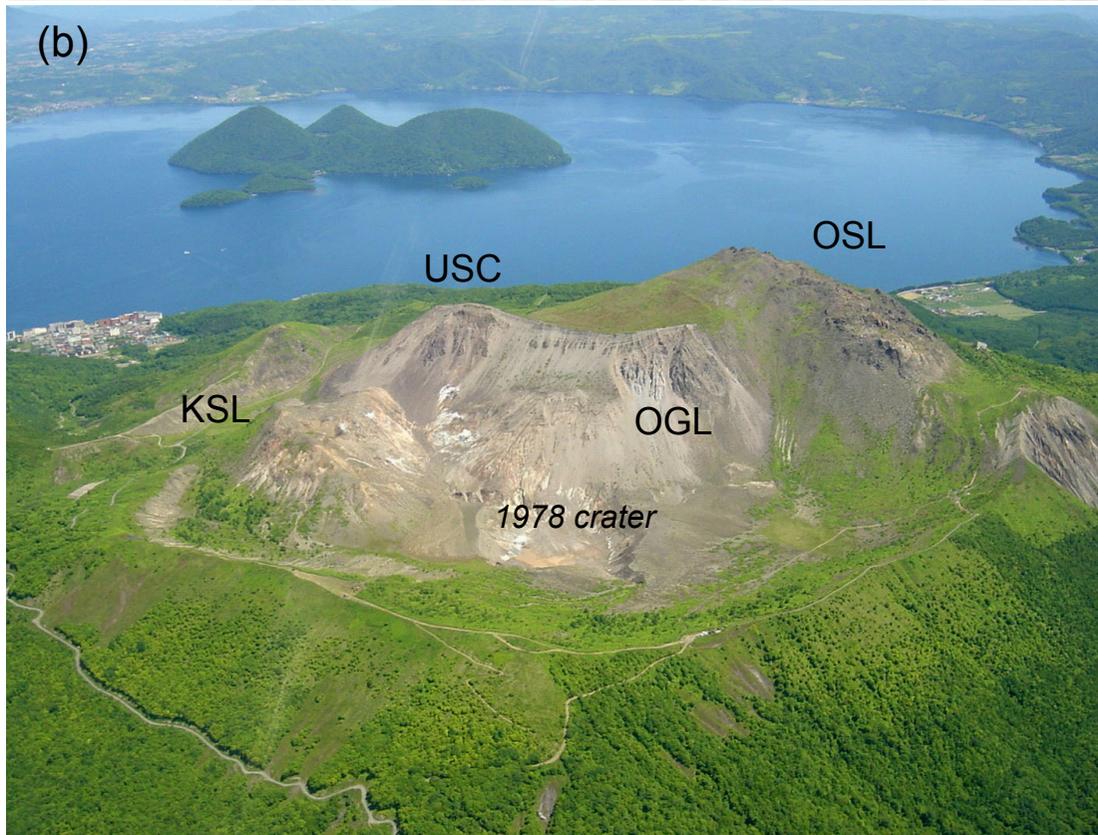
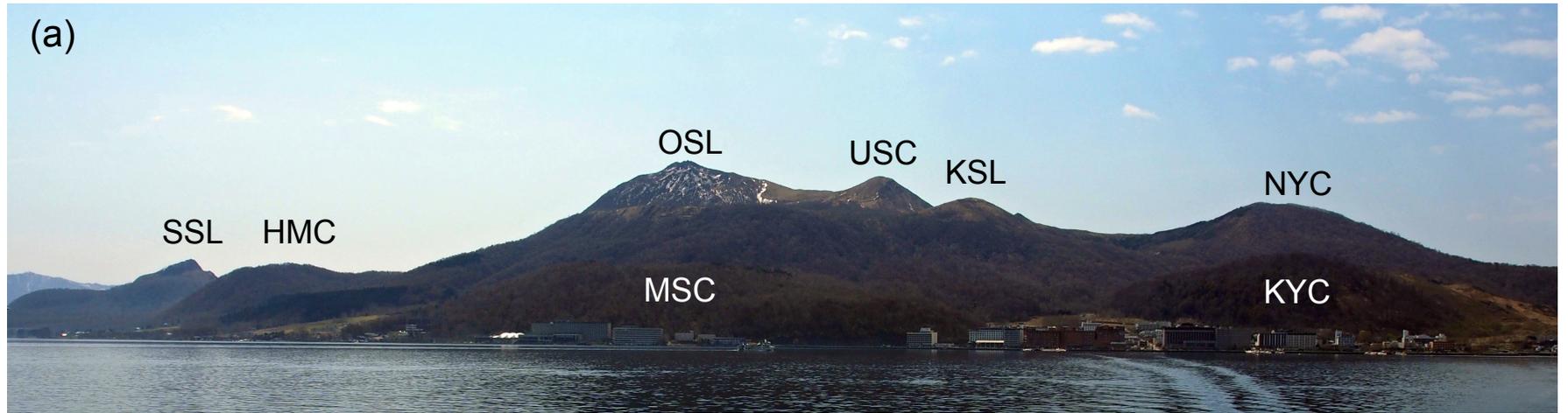
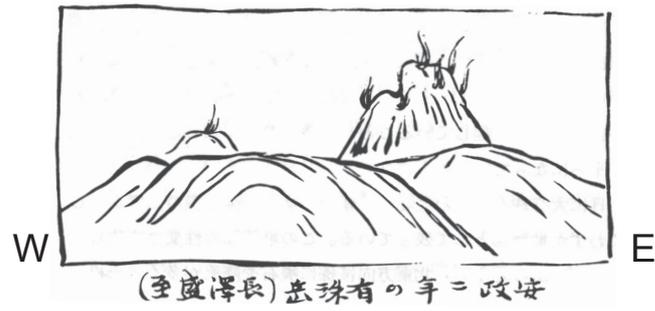


Fig.2

(a) AD 1855



(b) AD 1799



Fig.3

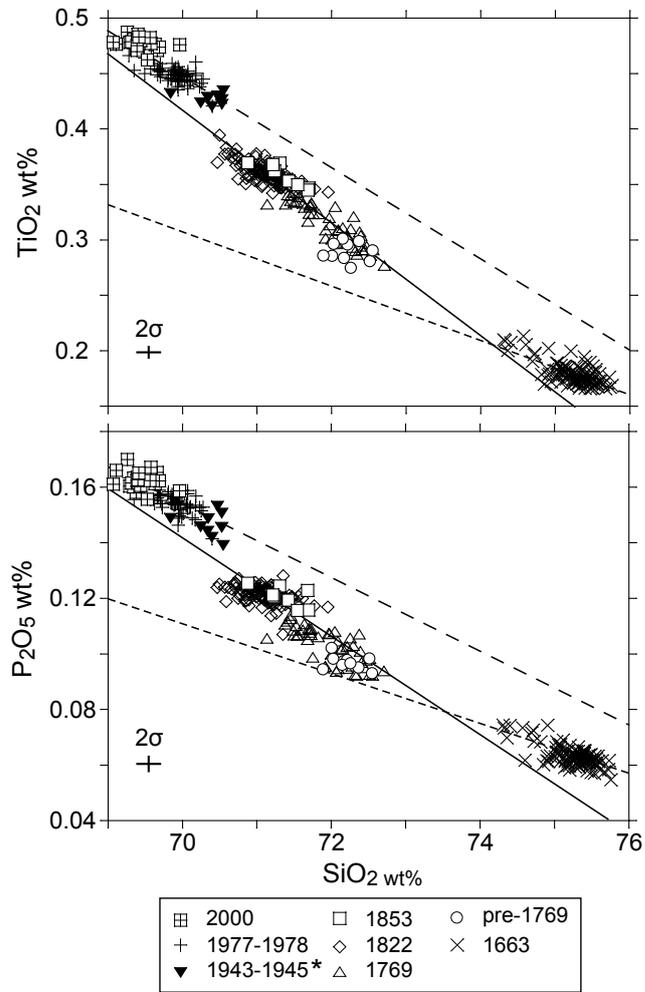


Fig.4

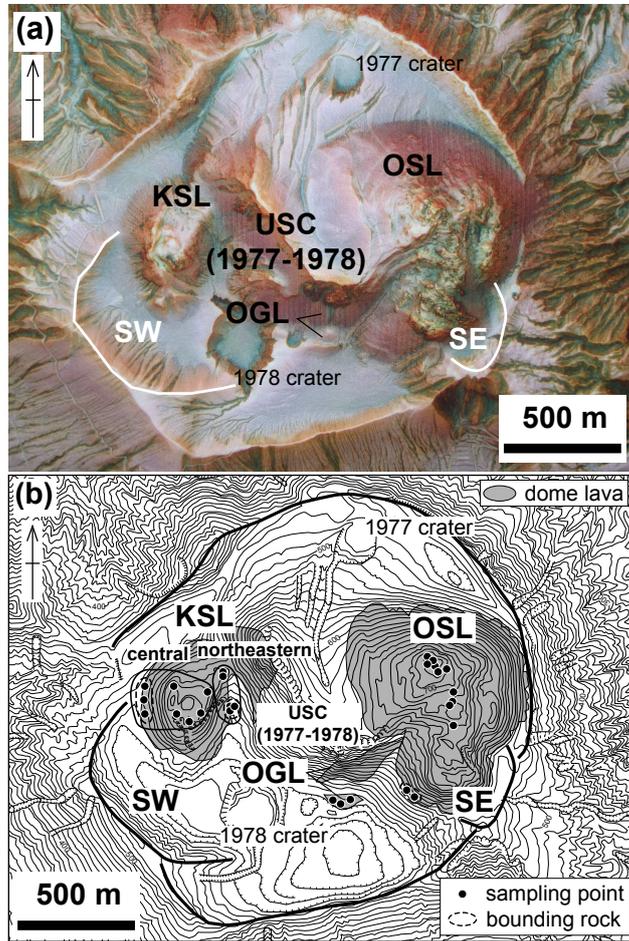


Fig.5

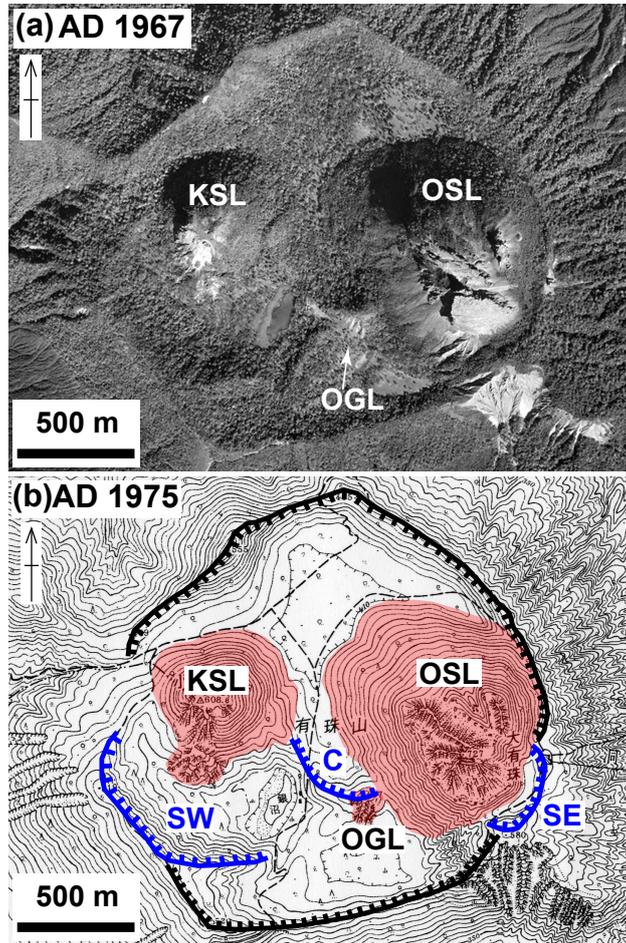


Fig.6

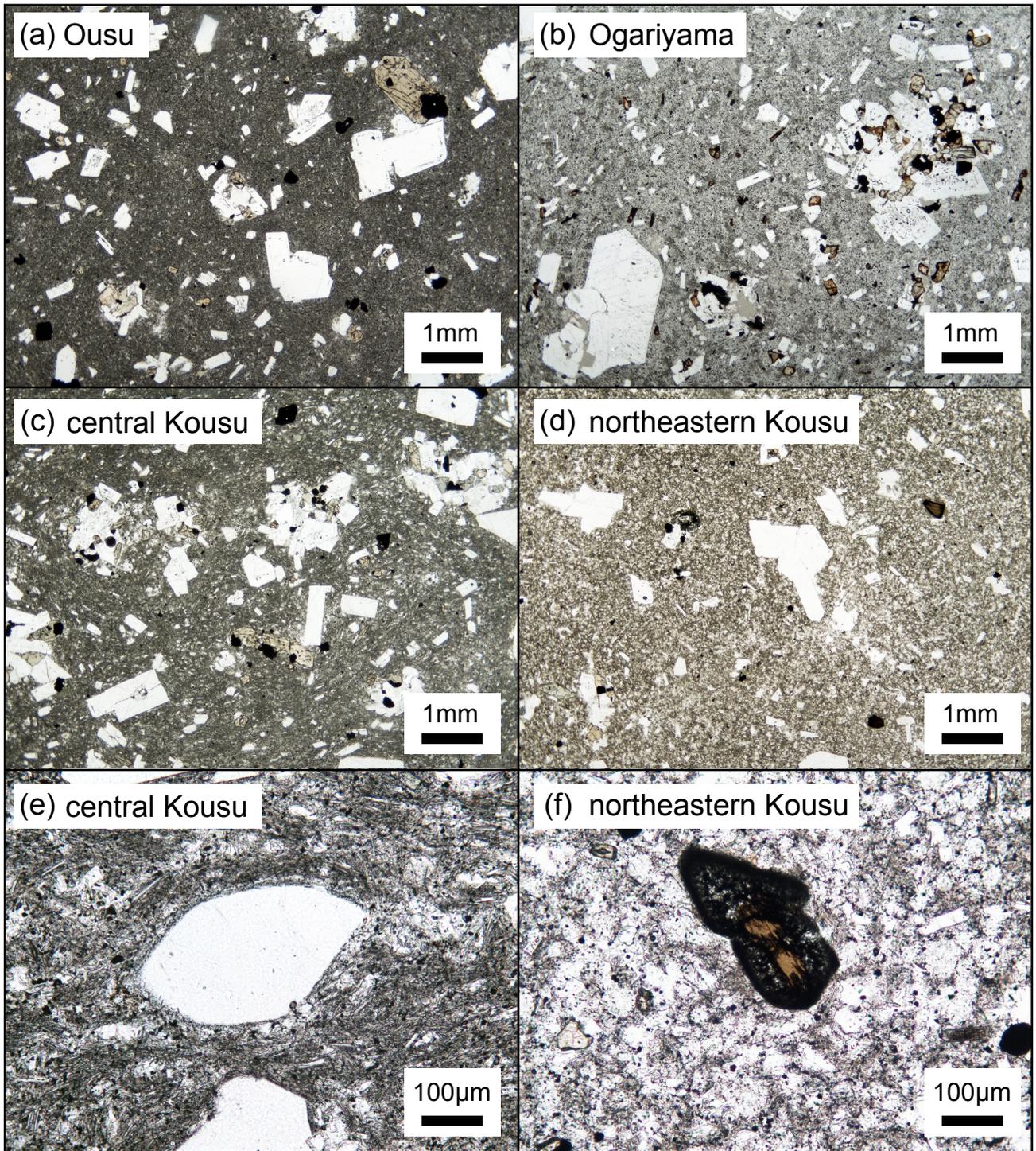


Fig.7

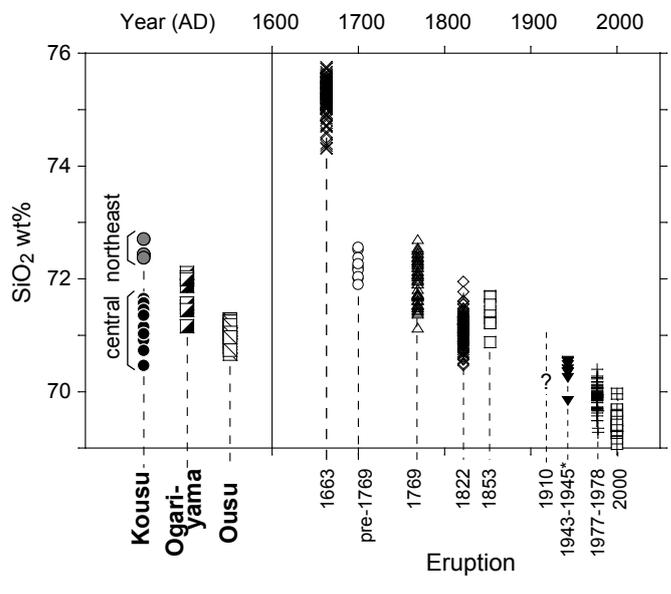


Fig.8

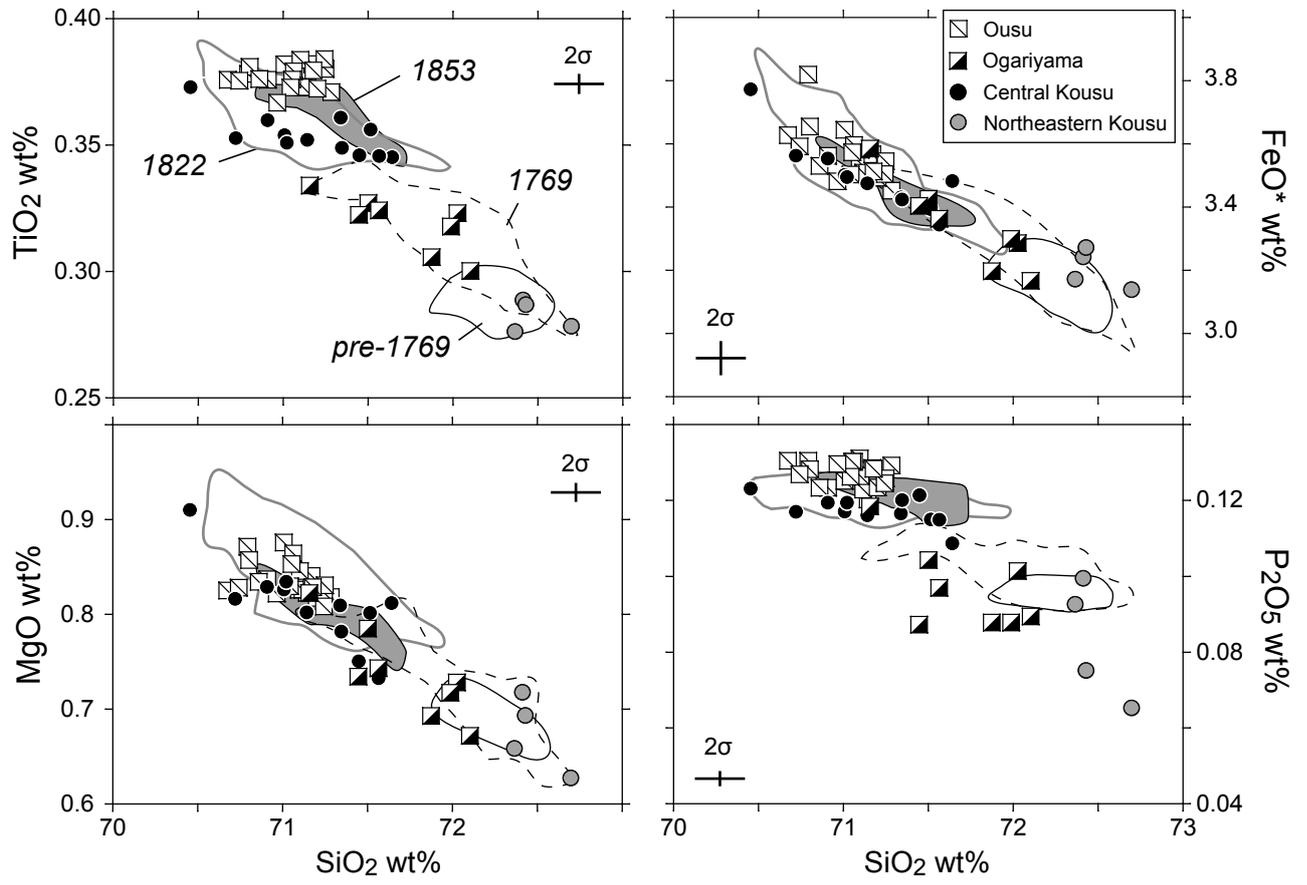


Fig.9

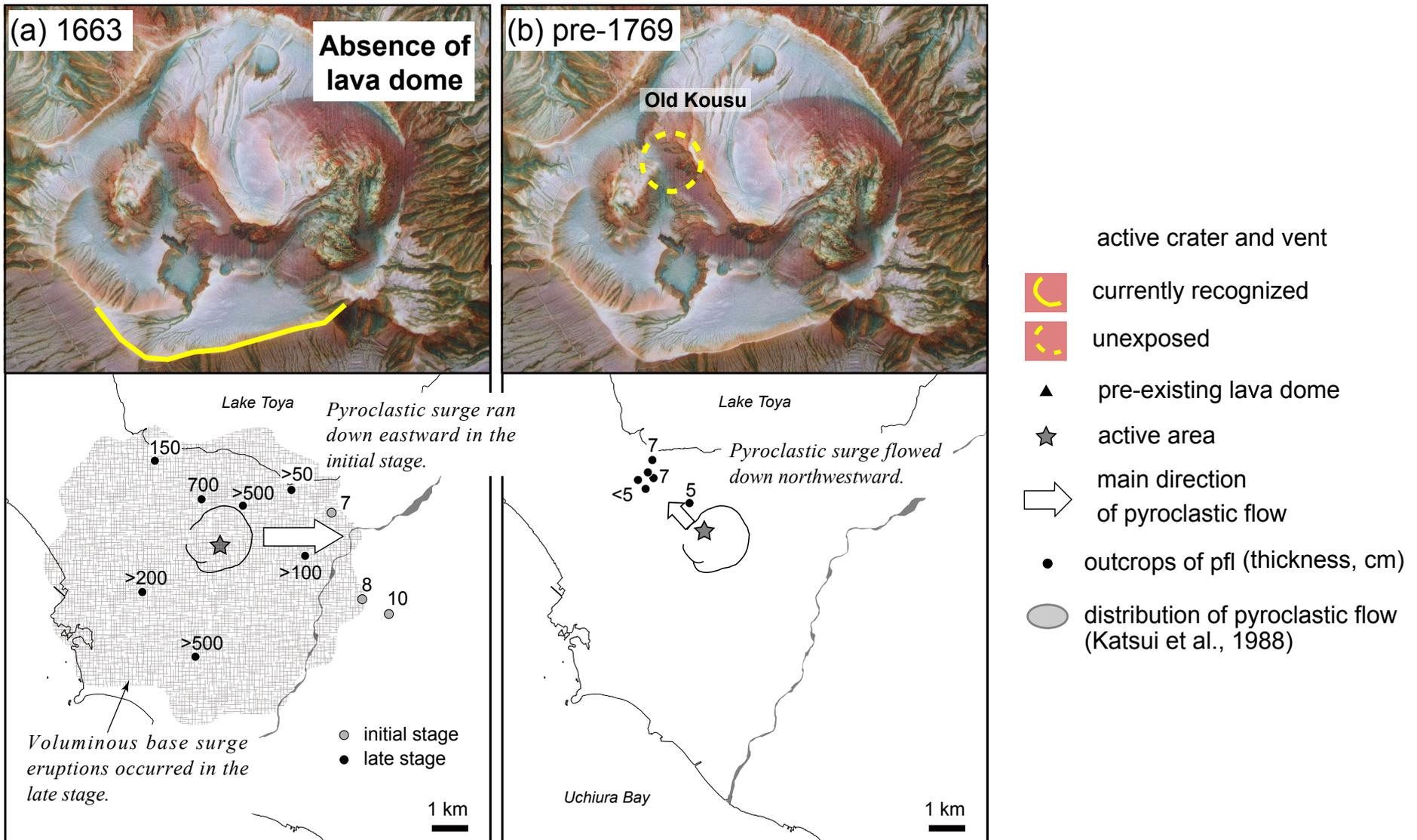


Fig.10

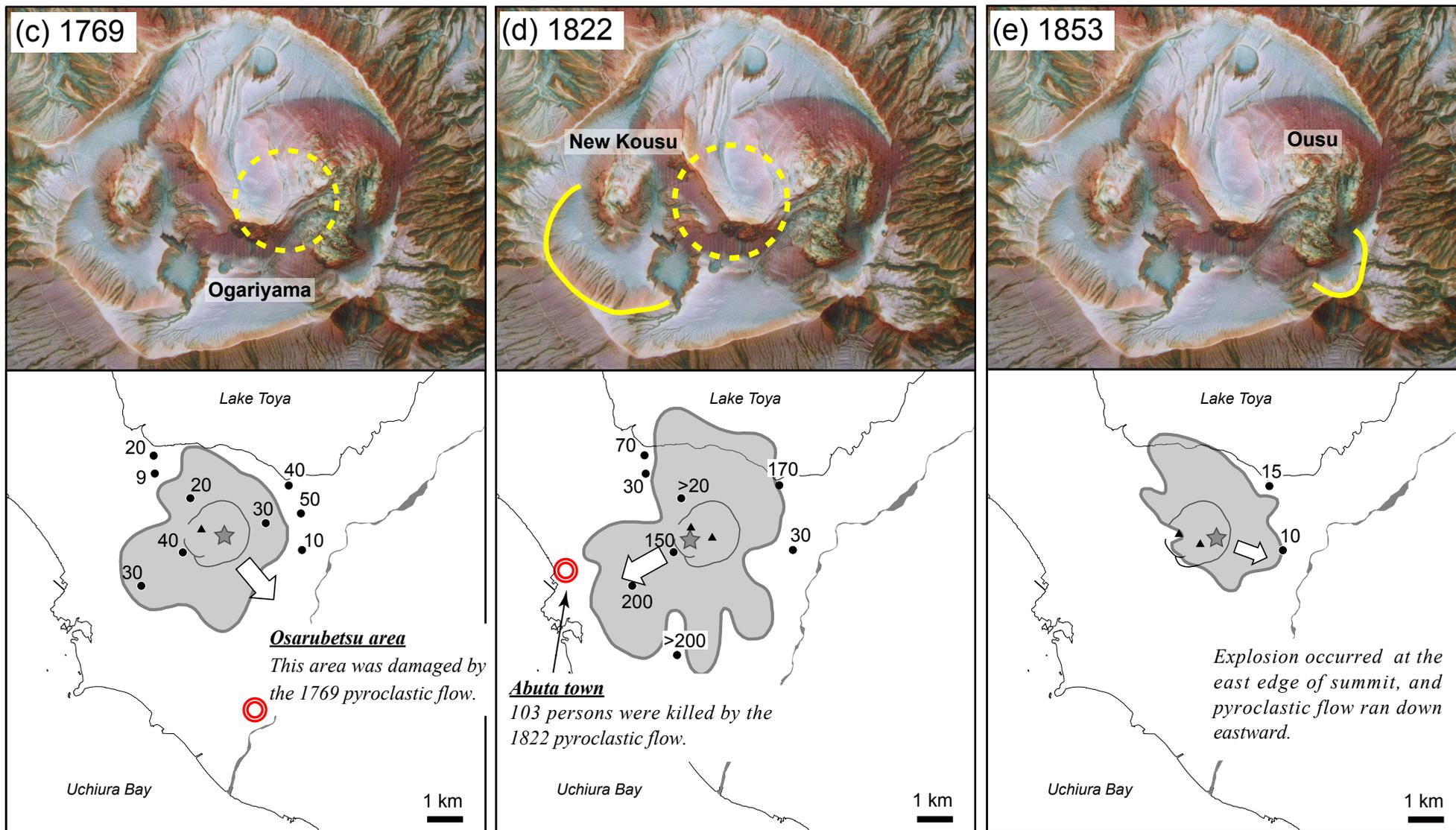


Fig.10 continued

Table 1. Historical eruptive activity of Usu volcano, modified from Nakagawa et al. (2005) and Soya et al. (2007). Bold texts show the revised points by this study.

Magmatic group *	Eruption	Tephra (volume: km <sup>3</sup> **)	Formation of dome		Vent location	
			Previous	This study	Previous	This study
3	2000	ash and pumice fall (0.001)	CD	CD	NW flank	NW flank
	1977-1978	pumice and ash fall (0.0905)	Usu-shinzan CD ***	Usu-shinzan CD ***	Summit	Summit
	1943-1945	ash fall(phreatic?) (0.004)	Showa-shinzan LD (spine)	Showa-shinzan LD (spine)	E flank	E flank
	1910	ash fall(phreatic?) (0.004)	Meiji-shinzan CD	Meiji-shinzan CD	N flank	N flank
2	1853	pyroclastic flow (0.01) pumice and ash fall (0.35)	Ousu LD	Ousu LD	Summit	<b>SE summit</b>
	1822	pyroclastic flow (0.09) pumice and ash fall (0.28)	Ogariyama CD (?)	<b>New Kousu LD</b>	Summit	<b>SW and C summit</b>
	1769	pyroclastic flow (0.03) pumice and ash fall (0.11)	Kousu LD (?)	<b>Ogariyama CD****</b>	Summit	<b>C or SE summit</b>
	pre-1769	pyroclastic surge (nd) pumice and ash fall (nd)	?	<b>Old Kousu LD</b>	Summit	<b>NW summit</b>
1	1663	base surge (0.6) pumice and ash fall (1.85) pyroclastic surge and pumice fall (nd)	?	<b>None</b>	Summit	<b>Summit</b>

nd, no data

CD: cryptodome; LD: lava dome; NW: northwestern; E: eastern; N: northern; SE: southeastern; SW: southwestern; C: central.

\* The grouping is after Matsumoto & Nakagawa (2010).

\*\* Data are from Katsui et al. (1981), Katsui et al. (1988) and Kadomura et al. (1988).

\*\*\* The Growth of dome had continued until 1984 (Kadomura et al., 1988).

\*\*\*\* This cryptodome might extrude partially.

Table 2. Representative phenocryst modes and whole-rock compositions of Usu dome lavas from the summit. The data of tephra from the historical eruptions are also shown (Matsumoto & Nakagawa, 2010).

Lava dome	Ousu	Ousu	Ogari-yama	Ogari-yama	Northeastern Kousu	Northeastern Kousu	Central Kousu	Central Kousu	Tephra					
	No. / Eruption	Os-16	Os-3	Og-5	Og-6	Ks-22	Ks-29	Ksc-2	Ks-26	1663	pre-1769	1769	1822	1853
<b>Phenocryst mode (vol%)</b>									<i>Average</i>					
plg.	16.5	13.5	12.9	13.1	11.1	8.6	12.7	17.5	2.0	5.2	14.3	11.8	13.2	9.0
opx	2.0	1.5	1.3	2.3	1.3	1.4	1.7	2.0	0.5	1.3	2.3	1.6	1.9	1.4
opq	0.7	0.5	0.3	0.4	0.3	0.7	0.5	0.7	0.3	0.4	0.8	0.8	0.7	0.4
cpx	-	-	-	-	tr	0.2	-	-	0.1	tr	tr	tr	tr	tr
hbl	-	-	-	-	0.2	tr	-	-	0.1	tr	0.1	tr	tr	-
qtz	-	-	-	-	tr	tr	tr	tr	-	tr	0.3	0.5	0.8	-
pheno.	19.1	15.5	14.5	15.8	13.0	10.8	14.9	20.2	3.0	7.2	17.9	14.4	16.7	10.9
gms.	80.9	84.5	85.5	84.2	87.0	89.2	85.1	79.8	97.0	92.8	82.1	85.6	83.3	89.1
<b>Whole-rock (wt%)</b>														
SiO <sub>2</sub>	71.6	73.0	70.9	71.4	73.6	73.3	69.9	73.2						
TiO <sub>2</sub>	0.37	0.37	0.32	0.32	0.28	0.27	0.37	0.36						
Al <sub>2</sub> O <sub>3</sub>	14.9	15.2	14.6	14.4	14.8	14.7	14.9	15.2						
FeO*	3.5	3.4	3.3	3.3	3.1	3.0	3.7	3.3						
MnO	0.16	0.16	0.17	0.17	0.16	0.16	0.17	0.16						
MgO	0.84	0.82	0.74	0.71	0.69	0.65	0.90	0.80						
CaO	3.5	3.5	3.2	3.1	3.0	3.0	3.7	3.4						
Na <sub>2</sub> O	4.5	4.5	4.7	4.7	4.7	4.8	4.6	4.5						
K <sub>2</sub> O	0.95	0.95	0.96	0.97	1.02	1.03	0.92	0.94						
P <sub>2</sub> O <sub>5</sub>	0.13	0.13	0.10	0.09	0.08	0.10	0.12	0.12						
total	100.4	102.0	99.0	99.1	101.4	101.0	99.3	101.9						
<b>Corrected value (100%-normalized)**</b>									<i>Range</i>					
SiO <sub>2</sub>	70.8	71.1	71.6	72.0	72.4	72.4	70.5	71.3	74.3-75.8	71.9-72.6	71.1-72.7	70.5-72.0	70.9-71.7	69.1-70.6
TiO <sub>2</sub>	0.38	0.37	0.32	0.32	0.29	0.28	0.37	0.36	0.17-0.21	0.27-0.30	0.28-0.34	0.34-0.39	0.35-0.37	0.42-0.49
Al <sub>2</sub> O <sub>3</sub>	14.9	14.9	14.8	14.5	14.5	14.5	15.0	14.9	13.5-14.2	14.5-14.7	14.5-14.8	14.6-15.0	14.7-14.9	14.9-15.4
FeO*	3.7	3.5	3.4	3.30	3.3	3.2	3.8	3.4	2.1-2.8	3.0-3.3	2.9-3.5	3.3-3.9	3.3-3.6	3.7-4.2
MnO	0.17	0.16	0.17	0.17	0.17	0.17	0.17	0.16	0.15-0.17	0.16-0.17	0.16-0.17	0.16-0.18	0.16-0.17	0.16-0.18
MgO	0.86	0.82	0.74	0.72	0.69	0.66	0.91	0.81	0.22-0.46	0.65-0.73	0.63-0.82	0.77-1.02	0.75-0.84	0.86-1.05
CaO	3.5	3.4	3.2	3.2	2.9	3.0	3.7	3.3	1.9-2.5	3.0-3.2	3.0-3.4	3.2-3.7	3.3-3.5	3.6-4.2
Na <sub>2</sub> O	4.7	4.6	4.8	4.7	4.7	4.8	4.67	4.7	4.7-4.9	4.6-4.8	4.6-4.7	4.5-4.8	4.6-4.7	4.5-4.7
K <sub>2</sub> O	0.95	0.93	0.97	0.98	0.98	1.01	0.92	0.93	1.07-1.17	0.98-1.05	0.95-1.05	0.92-1.00	0.96-1.01	0.89-0.97
P <sub>2</sub> O <sub>5</sub>	0.13	0.12	0.10	0.09	0.08	0.09	0.12	0.12	0.05-0.07	0.09-0.10	0.09-0.11	0.11-0.13	0.12-0.13	0.14-0.17
total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0						

\*: the total iron is given as FeO. \*\*: In order to compare the WR data, we corrected the influence of the difference between MagiX PRO and PW1404 systems, using more than 10 samples.

plg: plagioclase; opx: orthopyroxene; opq: opaque mineral; cpx: clinopyroxene; hbl: hornblende; qtz: quartz; pheno.: phenocryst; gms.: groundmass.

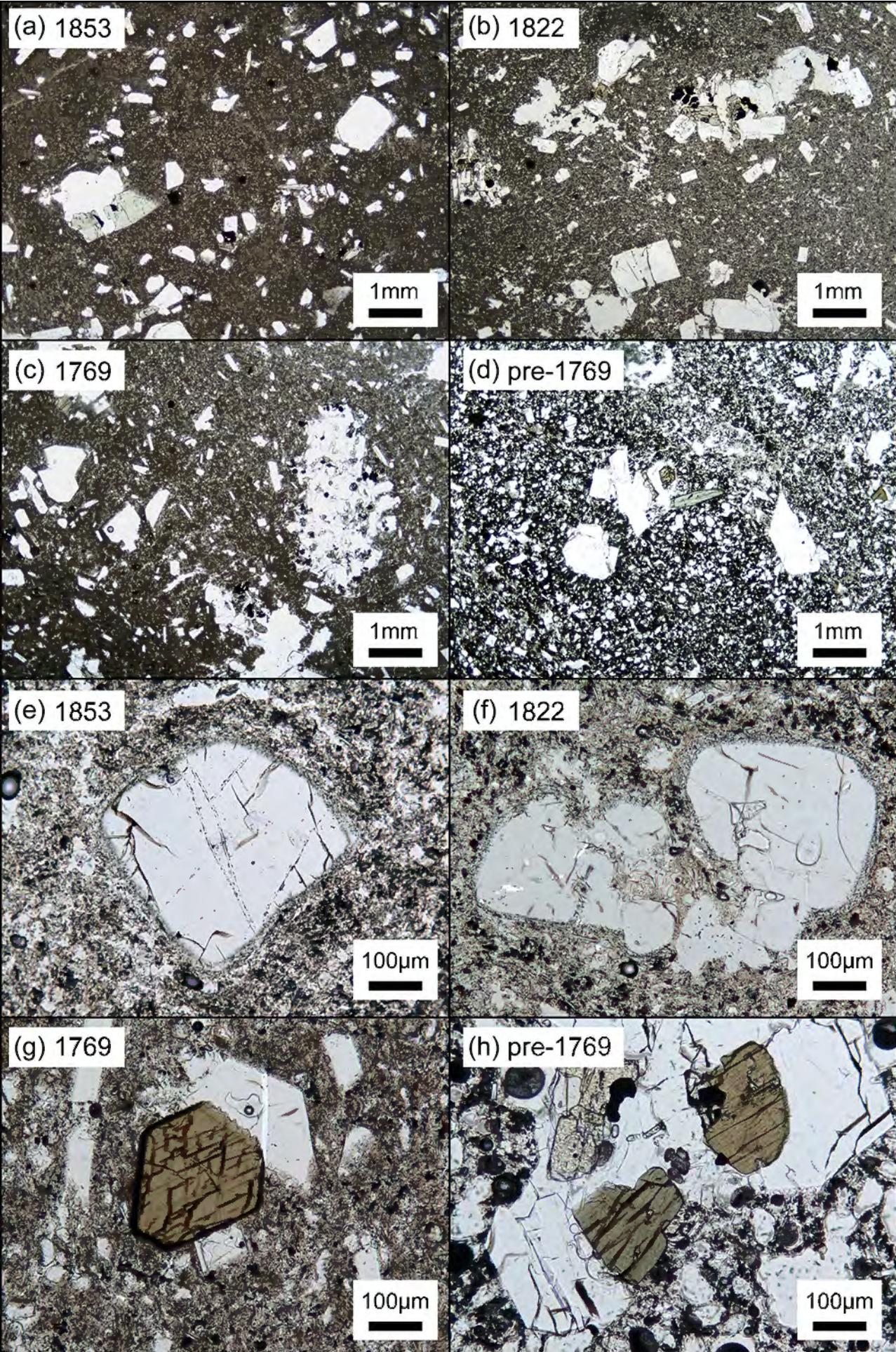
Table 3. Comparison of the petrological features between dome lavas and tephra samples of Usu volcano during the 17<sup>th</sup>-19<sup>th</sup> centuries.

Eruption / Lava dome	Type	Whole-rock SiO <sub>2</sub> wt.%	Whole-rock P <sub>2</sub> O <sub>5</sub> wt%	Phenocryst content vol% (average)	plg	opx	opq	cpx	hbl	qtz
1853	T	70.9-71.7	0.116-0.125	16.7	●	○	++	tr	tr*	++
Ousu	DL	70.8-71.1	0.123-0.131	15.8	●	○	++	-	-	-
1822	T	70.5-72.0	0.107-0.128	14.4	●	○	++	tr	tr*	+
C Kousu	DL	70.5-71.3	0.105-0.123	19.6	●	○	++	-	-	tr
1769	T	71.1-72.7	0.092-0.114	17.9	●	○	++	tr	+	+
Ogariyama	DL	71.6-72.0	0.087-0.118	14.5	●	○	+	-	-	-
pre-1769	T	71.9-72.6	0.093-0.102	7.2	●	○	+	tr	tr	tr
NE Kousu	DL	72.4-72.4	0.067-0.099	12.0	●	○	++	+	+	tr
1663	T	74.3-75.8	0.055-0.074	3.0	○	++	+	+	+	-

T: tephra; DL: dome lava. C: central; NE: northeastern. plg: plagioclase; opx: orthopyroxene; opq: opaque mineral; cpx: clinopyroxene; hbl: hornblende; qtz: quartz.

●: >5.0; ○: >1.0; ++: >0.5; +: >0.1; tr: <0.1; -: absent

tr\*: We found a few grains in more than 5 samples.



**Figure S1.** Photomicrographs of representative samples of the tephra samples from eruptions during the 18th and 19th centuries: (a) 1853, (b) 1822, (c) 1769, and (d) pre-1769 (around AD 1700). Quartz phenocrysts in tephra samples from the (e) 1853 and (f) 1822 eruptions, and hornblende phenocrysts in tephra samples from the (g) 1769 and (h) pre-1769 eruptions.

## Supporting information Doc S1. Sample preparation and analytical details for whole-rock chemistry

All the preparation and analyses were carried out at Hokkaido University. We made a few thin slabs (5 mm to 1 cm in thickness, >10 g in total) from each sample, and washed them using the ultrasonic cleaner. After drying at 100 °C for two days, we crushed them to 1 cm particles by iron mortar and made their powders using agate ball mill. We measured 1.5 g of powder from each sample, and baked at 800 °C for 3 hours. After cooling, we made their glass beads by fusing at 1100 °C for 8 minutes with an alkali flux (lithium tetraborate), which are diluted to 1:2.

Whole-rock compositions were determined by X-ray fluorescence spectrometry (XRF) using Philips PW1404 and Spectris MagiX PRO systems with a Rh tube. The calibration curve method was applied, using GSJ standard rock samples: JB-1a, JB-1b, JB-2, JB-3, JA-1, JA-2, JA-3, JR-1, JR-2, JR-3, JGb-1, JGb-2, JG-1a, JG-2, JG-3, JF-1, JF-2, JSy-1, and JP-1 (<https://gbank.gsj.jp/geostandards/welcome.html>; Imai et al., 1995). Total time of analysis for each sample is *ca* 10 minutes. The results of replicate analysis using the house-standard sample in our laboratory (HR-1: rhyolitic pumice from Shikotsu caldera volcano, Japan) are listed in Table S1.

Table S1. Results of replicate analysis for house-standard sample (HR-1: rhyolitic pumice of Shikotsu caldera volcano, Japan).

wt.%	PW-1404		MagiX PRO	
	Average n=18	2sigma	Average n=15	2sigma
SiO <sub>2</sub>	76.47	0.15	76.52	0.07
TiO <sub>2</sub>	0.19	0.00	0.18	0.01
Al <sub>2</sub> O <sub>3</sub>	13.33	0.07	13.46	0.02
FeO*	1.76	0.05	1.67	0.03
MnO	0.07	0.00	0.07	0.00
MgO	0.21	0.01	0.19	0.00
CaO	1.58	0.03	1.60	0.01
Na <sub>2</sub> O	3.98	0.13	3.87	0.03
K <sub>2</sub> O	2.39	0.05	2.43	0.04
P <sub>2</sub> O <sub>5</sub>	0.02	0.00	0.02	0.00

All the data are normalized to 100% volatile free.

\*: total Fe is given in FeO.

Imai, N., Terashima, S., Itoh, S. and Ando, A. (1995). 1994 compilation values for GSJ reference samples, "Igneous rock series". *Geochemical Journal*, 29, 91-95.