

Accelerator Mass Spectrometric Radiocarbon Chronology during the Last 30,000 Years of the Aira Caldera, Southern Kyushu, Japan

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Abstract

An advanced and sophisticated technique of radiocarbon (^{14}C) dating by accelerator mass spectrometry (AMS) has revealed a detailed eruptive history of the Aira caldera during the last 30,000 years. This caldera is located in the northernmost part of Kagoshima Bay, southern Kyushu, Japan. Sakurajima volcano, one of the most active volcanoes in Japan, is a post-caldera stratovolcano of the caldera.

I analyzed 60 samples in total; 12 charcoal, one wood and 47 paleosol samples in the Aira caldera and its surroundings. Paleosol samples are available more easily and systematically than charcoal or wood samples. In addition, I focused particularly on non-disturbed tephra layers which implied good depositional conditions of the paleosol layers immediately below the tephra layers (*i.e.*, continuity of soil accumulation and non-disturbance of paleosols), because the tephra layers covering the paleosol layers work as a barrier to prevent vertical movement of soil organic matter. Their ^{14}C dates are consistent with stratigraphical relationship among them. Therefore, ^{14}C dates of paleosol samples represent the time when the tephra layer covered the paleosol, namely, its eruption age. C/N ratio of humin fraction from paleosol sample is a promising indicator for detecting possible sample contamination from allochthonous organic materials.

On the basis of these ^{14}C dates, I restored a well-constrained eruptive history of the Aira caldera during the last 30,000 years. Since the Otsuka eruption of 30 ka, at least four major pyroclastic eruptions (0.1 km^3 to 1.5 km^3 in bulk volume for each tephra) occurred intermittently in the eastern part of Aira caldera until 25 ka. At 24.5 ka, a series of large-scale eruptions ($>411 \text{ km}^3$ in bulk volume),

referred to the AT eruption, occurred. Sakurajima volcano started eruption at 22.5 ka. The Old Kita-dake stage of this volcano lasted until 20 ka. During the dormant period between the Old and Young Kita-dake stages, the Takano base surge was erupted from the eastern part of the Aira caldera at 16 ka. After 9000 years of quiescence of Sakurajima volcano, the Young Kita-dake stage started with the Satsuma eruption at 11 ka. This stage continued from 11 ka to 3.5 ka with quasi-periodic activity of an 800–2000 year recurrence interval. Several well-known historical eruptions in the Minami-dake stage followed it. The calendar age ranges calibrated dendro-chronologically from ^{14}C dates for these eruptions agree with their calendar dates, based on the historical documents. Thus, these ^{14}C dates provide chronological constraints for correlating tephra layers with the historical records.

Key words: AMS ^{14}C dating, paleosol, tephra layers, eruptive history, late Quaternary, Aira caldera, Sakurajima volcano

1. Introduction

The timing of past volcanic eruptions is essential to understanding of the temporal behavior of the earth's magmatic system. The radiocarbon (^{14}C) method is very useful for precise dating of recent eruptions. It is generally accepted by many workers that charcoal and wood are the most suitable samples for ^{14}C dating. However, I suspect that this view is not always true for ^{14}C dating of volcanic eruptions, for of the following reasons. Charcoal and wood samples often introduce uncertainties to their ages, sometimes resulting in ^{14}C dates that exceed the expected ages by more than 1 ka (Okuno, 1995). A possible reason for such discrepancy is that charcoal is rather resistant to weathering, remaining intact for some millennia after its production, at which time the old charcoal may be included in younger volcanic deposits. In addition, charcoal and wood samples are commonly collected from pyroclastic flow and surge deposits, but rarely from fallout deposits. The temperature of a fallout deposit is usually not high enough to carbonize plant materials. Charcoal is usually restricted to near-vent deposits. On the other hand, paleosol samples can be collected more easily and systematically than charcoal or wood

samples (Braitseva *et al.*, 1993; Okuno, 1995; Okuno *et al.*, in press), though organic matter in them may be subject to some vertical movement after deposition. Orlova and Panychev (1993) discussed the validity of ^{14}C dates of soil organic matter and concluded that reasonable ^{14}C dates can be obtained for organic fractions of paleosols below alluvial and flood deposits, provided that they accumulated very quickly and that their thickness is sufficient to prevent any penetration of rootlets from more recent vegetation. The same condition seems to apply for paleosol layers which are sandwiched between tephra layers. Accelerator mass spectrometry (AMS) is the most suitable method for paleosol ^{14}C dating because it requires very small amounts of carbon containing material, *i.e.*, about 1 mg of carbon.

Matumoto (1943) recognized three gigantic calderas, the Aira, Ata, and Kikai calderas in southern Kyushu, Japan (Fig. 1). Many tephra layers have been deposited by eruptions of the Aira caldera during the late Quaternary. Aramaki (1969) described the Ito pyroclastic flow deposit and several underlying pyroclastic units around Kagoshima Bay. Aramaki and Ui (1976) correlated pyroclastic flow deposits in southern Kyushu on the basis of the Ca-Mg-Fe ratios of phenocrystic minerals within them. Machida and Arai (1976, 1978, 1983) identified two widespread tephtras for the first time, the Aira-Tn (AT) Tephra and the Kikai-Akahoya (K-Ah) Tephra, which were erupted from the Aira and Kikai calderas, respectively. Nagaoka (1988) established the stratigraphy of fallout tephtras in southern Kyushu. The stratigraphy of tephra layers from Sakurajima volcano, collectively named the Sakurajima (Sz) Tephra Group, has been investigated by Kobayashi (1986a) and Moriwaki (1994). Recently, Kuwahata and Higashi (*in press*) reviewed the stratigraphic relation of prominent tephra layers and archeological remains, such as pottery fragments and stone implements, excavated from archeological sites in southern Kyushu. The Aira caldera with its surroundings is therefore one of the fields suitable for establishing a high-resolution ^{14}C chronology of the major eruptions.

Many ^{14}C dates for charcoal fragment samples from the AT Tephra (*e.g.*, Kigoshi *et al.*, 1972) and the K-Ah Tephra (*e.g.*, Ui and Fukuyama, 1972) have been given by the traditional beta counting method. These dates indicate an approximate eruption age of 21,000–22,000 yr BP for the AT Tephra and of 6300 yr BP for the K-Ah Tephra (Machida and Arai, 1992). AMS ^{14}C dating gives an eruption age of about

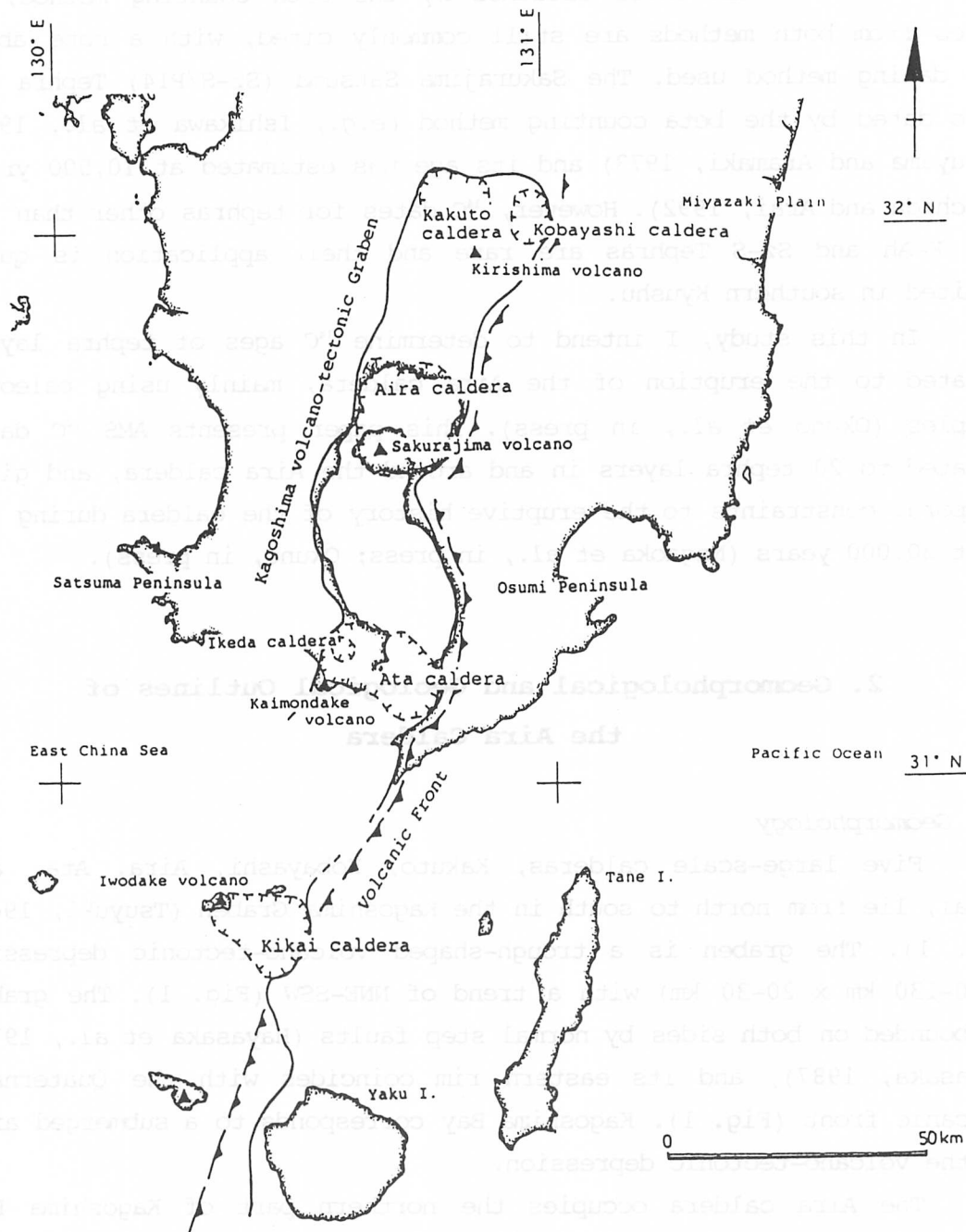


Fig. 1 Index map of Quaternary volcanoes in southern Kyushu (modified from Nagaoka, 1988).

24,500 yr BP for the AT Tephra (Ikeda *et al.*, 1995) and 6750 yr BP for the K-Ah Tephra (Kitagawa *et al.*, 1995). While these AMS ^{14}C dates are somewhat older than those obtained by the beta counting method, ^{14}C dates from both methods are still commonly cited, with a note about the dating method used. The Sakurajima Satsuma (Sz-S/P14) Tephra was also dated by the beta counting method (*e.g.*, Ishikawa *et al.*, 1972; Fukuyama and Aramaki, 1973) and its age was estimated at 10,500 yr BP (Machida and Arai, 1992). However, ^{14}C dates for tephtras other than the AT, K-Ah and Sz-S Tephtras are rare and their application is quite limited in southern Kyushu.

In this study, I intend to determine ^{14}C ages of tephtra layers related to the eruption of the Aira caldera, mainly using paleosol samples (Okuno *et al.*, in press). This paper presents AMS ^{14}C dates related to 20 tephtra layers in and around the Aira caldera, and gives temporal constraints to the eruptive history of the caldera during the last 30,000 years (Nagaoka *et al.*, in press; Okuno, in press).

2. Geomorphological and Geological Outlines of the Aira Caldera

2.1 Geomorphology

Five large-scale calderas, Kakuto, Kobayashi, Aira, Ata, and Kikai, lie from north to south in the Kagoshima Graben (Tsuyuki, 1969: Fig. 1). The graben is a trough-shaped volcano-tectonic depression (120-130 km x 20-30 km) with a trend of NNE-SSW (Fig. 1). The graben is bounded on both sides by normal step faults (Hayasaka *et al.*, 1978; Hayasaka, 1987), and its eastern rim coincides with the Quaternary volcanic front (Fig. 1). Kagoshima Bay corresponds to a submerged area of the volcano-tectonic depression.

The Aira caldera occupies the northern part of Kagoshima Bay (Fig. 1). Figure 2 shows the geomorphological map of the Aira caldera. The caldera is about 20 km in diameter. Its depth is about 130 to 200 m below sea level. The Wakamiko caldera (6.5 km x 5.5 km), named by Kuwashiro (1964), is located in the northeastern part of the submerged Aira caldera. Three subaqueous post-caldera cones, Tagiri, Hirase and

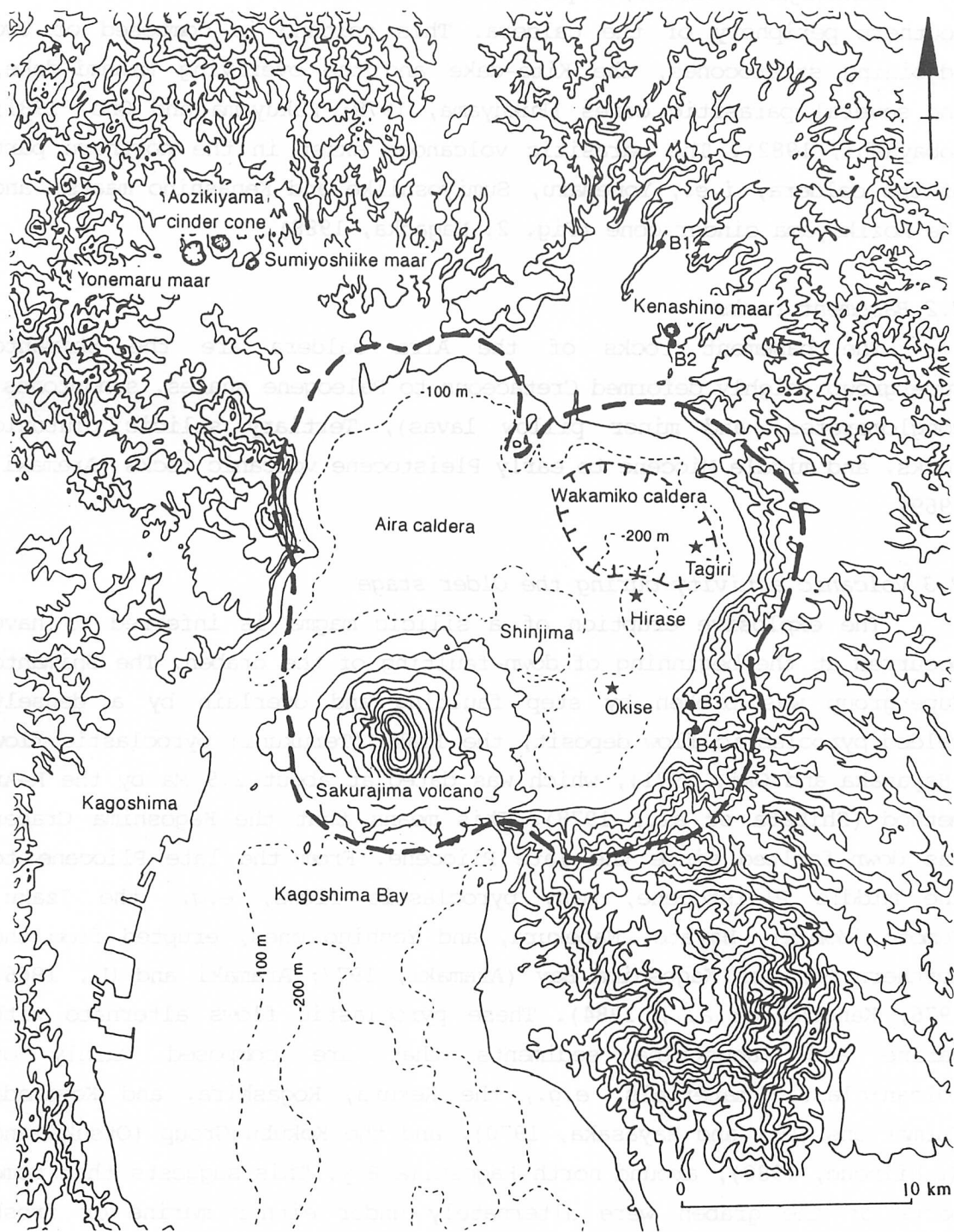


Fig. 2 Geomorphological map of the Aira caldera (modified from Nagaoka, 1988). Contour interval is 100 m. Thick broken line is the rim of the Aira caldera originally proposed by Matumoto (1943). Three submerged post-caldera cones are shown by stars. B1: Tsumaya, B2: Kenashino, B3: Ukitsu, B4: Fukaminato.

Sakurajima volcano, a post-caldera strato-volcano, stands on the southern periphery of the caldera. This volcano is composed of two adjoining stratocones, the Kita-dake and the overlying Minami-dake, and several parasitic cones (Fukuyama, 1978; Fukuyama and Ono, 1981; Kobayashi, 1982). The parasitic volcanoes exist in the northern part of the caldera, *i.e.*, Yonemaru, Sumiyoshiike and Kenashino maars, and the Aozikiyama cinder cone (Fig. 2: Nagaoka, 1988).

2.2 Basement rocks

The basement rocks of the Aira caldera are the Shimanto Supergroup (highly deformed Cretaceous to Paleogene shales, sandstones, conglomerates, and minor pillow lavas), Tertiary silicic plutonic rocks, and middle Miocene to early Pleistocene volcanic rocks (Aramaki, 1969).

2.3 Volcanic activity during the Older stage

The explosive eruption of a silicic magma is inferred to have occurred at the beginning of down-faulting of the graben. The Shimanto Supergroup was broken by step faulting and overlain by a densely welded pyroclastic flow deposit, the Izaku (Terukuni) pyroclastic flow (Hayasaka and Oki, 1971), which was dated at about 2.9 Ma by the K-Ar method (Shibata *et al.*, 1978). This means that the Kagoshima Graben has down-faulted since the late Pliocene. From the late Pliocene to the middle Pleistocene, many pyroclastic flows, *e.g.*, the Izaku, Fumoto, Mobiki, Hayato, Nabekura, and Yoshino ones, erupted from the northern part of Kagoshima Bay (Aramaki, 1977; Aramaki and Ui, 1966, 1976; Kaneoka *et al.*, 1984). These pyroclastic flows alternate with marine and lacustrine sediments that are composed mainly of volcanoclastic materials, *e.g.*, the Kekura, Kogashira, and Koyamada Formations (Oki and Hayasaka, 1970), and the Kokubu Group (Otsuka and Nishiinoue, 1980), around north Kagoshima Bay. This suggests that some parts of the graben were alternately under either marine or fresh water conditions from the early Pliocene to the middle Pleistocene. Thus, it is feasible that the initial structure of the Aira caldera was already established in this period (Nagaoka, 1988).

The period from 0.1 Ma to 0.5 Ma was a relatively quiet period (Nagaoka, 1988). In this period, the Shikine andesitic lava flow (Aramaki, 1969) and the Ushine rhyolitic and basaltic lava flows (Kobayashi *et al.*, 1977; Kaneoka *et al.*, 1984) effused from the

northeastern and southeastern rims of the Aira caldera, respectively. This quiet period separates the eruptive history of the Aira caldera into two active periods, *i.e.*, the Older and Younger stages (Nagaoka, 1988). In the middle Pleistocene, the Kobayashi, Tonohira, and Kakuto pyroclastic flow deposits (Aramaki and Ui, 1976; Tajima and Aramaki, 1980) erupted from the northern most part of the graben, *i.e.*, the Kobayashi and Kakuto calderas (Fig. 1).

2.4 Volcanic activity during the Younger stage

The Younger stage of the Aira caldera is represented by eight tephra layers (Fig. 3: Nagaoka, 1988, 1989; Nagaoka *et al.*, in press). The bulk volume of each tephra is also shown in Fig. 3 (Nagaoka, 1988). Three widespread tephra layers, the Ata, Kikai-Tozurahara (K-Tz), Aso-4 Tephtras can be used as fundamental time-markers in this region. Their ages are estimated at 102 ka, 92 ka, and 87 ka, respectively (Fig. 3), based on their stratigraphic positions (Machida and Arai, 1992, 1994). These tephtras erupted from the Ata, Kikai and Aso calderas in southern and central Kyushu, respectively.

Immediately after the Ata eruption, explosive activities of the Aira caldera restarted with the Hikiyama eruption (Hky: Nagaoka, 1988, 1989). The Aozikiyama cinder cone and the Hikiyama scoria were formed by this eruption. The stratigraphic position of the tephtra is between the Ata and K-Tz Tephtras (Fig. 3). The Kongoji (Kg) Tephtra consists of pyroclastic surge and ash fall deposits (Nagaoka, 1989). The Fukuyama (Fk) Tephtra comprises a single Plinian pumice fall deposit (Nagaoka, 1988). An eruption age for the tephtra is estimated at 88 ka, from its stratigraphic relation to the K-Tz and AT Tephtras in the southern part of Osumi Peninsula (Okuno *et al.*, 1995). During the time from 25 ka to 60 ka, the Iwato (Iwt), Otsuka (Ot), Fukaminato (Fm), Kenashino (Kn), and Arasaki (Ar) Tephtras were intermittently erupted from the eastern half of the Aira caldera (Nagaoka, 1984, 1988). The Iwt eruption is represented by Plinian pumice fall and pyroclastic flow deposits (Nagaoka, 1988). The eruption age of the Iwt Tephtra is estimated as 60 ka based on a stratigraphic horizon described by Nagaoka (1984). Both the Ot and Fm Tephtras are composed of only a Plinian pumice fall deposit. The Kn Tephtra comprises pumice fall, pyroclastic surge and ash fall deposits in ascending order (Nagaoka, 1988). The Ar Tephtra consists of pumice fall and pyroclastic flow deposits. Its stratigraphic relations with the Ot, Fm, and Kn are not yet known

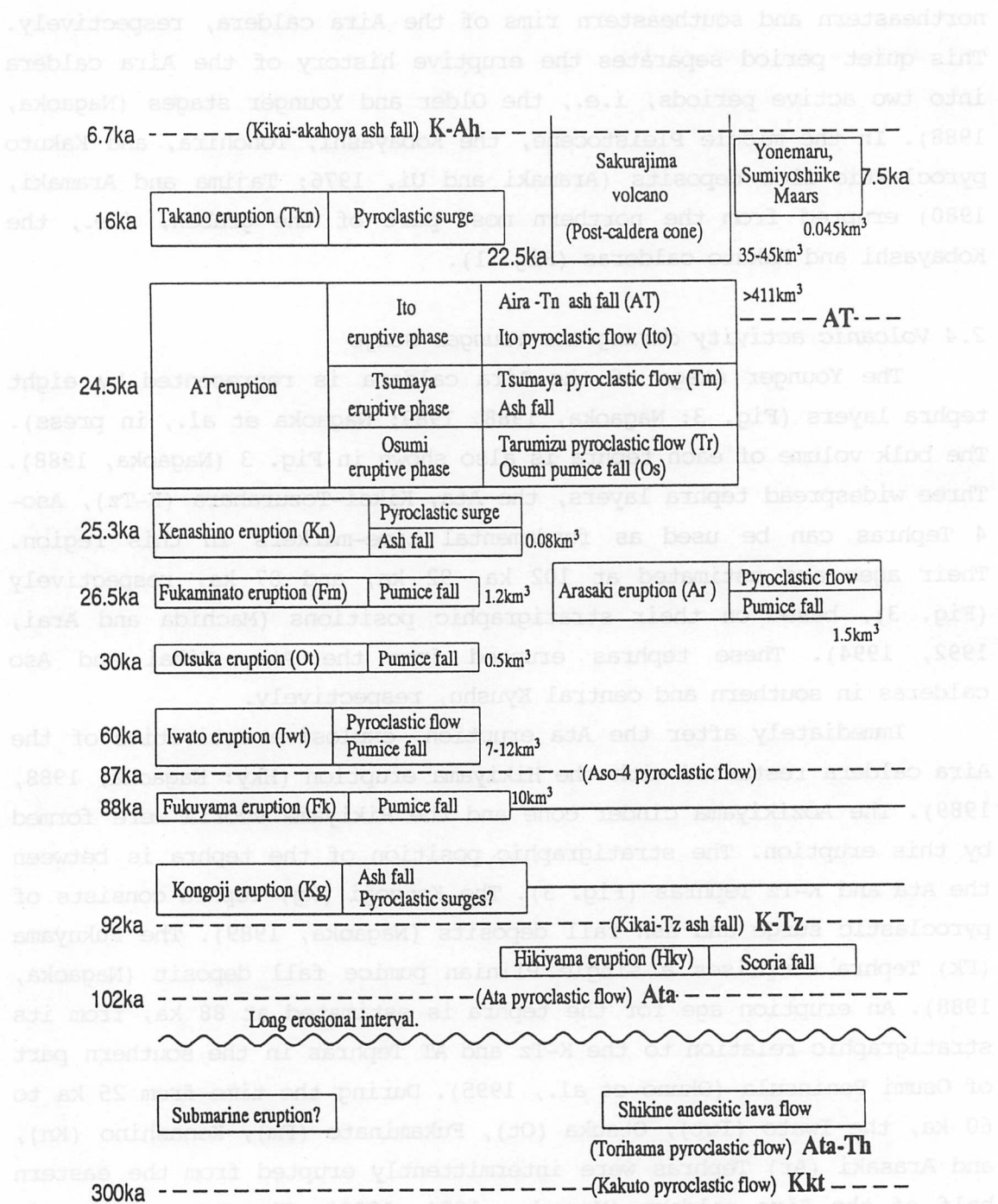


Fig. 3 Diagram showing the eruptive history of the younger stage of the Aira caldera (modified from Nagaoka, 1988; Nagaoka et al., 1997).

(Nagaoka et al., in press). These explosive eruptions were followed by the AT eruption, the largest one in the late Quaternary history of the Aira caldera. AT is a collective name for the products of this eruption (Machida and Arai, 1992). It consists of four major members; the Osumi pumice fall deposit (Os), Tsumaya pyroclastic flow deposit (Tm), Ito pyroclastic flow deposit (Ito) and co-ignimbrite ash-fall deposit in ascending order (Fig. 3: Aramaki, 1969, 1984; Kobayashi et al., 1983; Nagaoka, 1988). Immediately after the AT eruption, Sakurajima volcano on the southern periphery began eruption (Kobayashi, 1986a; Okuno et al., in press; Okuno, in press), and has continued its activity throughout the historical period.

3. Experimental Method

3.1 Sampling and sample preparation

Charcoal fragment samples in paleosols and tephra layers were collected from various sites, for AMS ^{14}C dating. Paleosol samples were also collected mainly from horizons immediately below and rarely from those above tephra layers (2 cm below and/or above tephras). Any disturbance of the paleosol was checked by careful observation of the boundary between the paleosol and tephra layers, and the paleosol samples were rejected when any trace of disturbance was detected. To obtain organic materials which were appropriate for ^{14}C dating, usually the humin fraction was separated from the paleosol samples, using the following procedures (Fig. 4: Okuno, 1995). The humic acid fraction, another organic fraction contained in paleosols, is not commonly used for ^{14}C dating, because it may contain some allochthonous carbon.

After washing its possibly contaminated surface with distilled water, a lump of paleosol, was dispersed in distilled water using an ultrasonic cleaner. Generally, about 20 g of paleosol sample was used to obtain enough carbon dioxide (CO_2) to produce graphite. Then the samples were wet-sieved using a 106 μm sieve to remove plant rootlets and animal remains. The samples were treated twice with 1.2 N HCl for 2 hours at 80 $^\circ\text{C}$ to remove carbonate contaminants, and then treated with 1.2 N NaOH for 1 hour at 80 $^\circ\text{C}$. They were again treated twice with HCl to remove NaOH completely. Then the treated samples were rinsed with distilled water to get rid of HCl and dried at 85 $^\circ\text{C}$. Carbon and nitrogen content of the humin fraction was measured, on 200 to 1000 mg

of the pretreated samples, using a CN coder (MT-700, Yanaco Ltd.).

In order to check for possible contamination of the paleosol samples by allochthonous carbon, humic acid fractions were separated from some of the samples in the following manner (Fig. 4). An alkaline solution, saved from the NaOH treatment, was filtered through glass, to remove solid materials. The solution was then combined with conc. HCl to make its pH value less than one in order to precipitate the humic acid. The solid residue was recovered by centrifugal separation and decantation, and again dissolved in 1.2 N NaOH solution for refining. The humic acid fraction was refined by repeating the treatments of precipitation with HCl and dissolution by NaOH solution a few times (Ikeda et al., 1995; Okuno et al., 1996a).

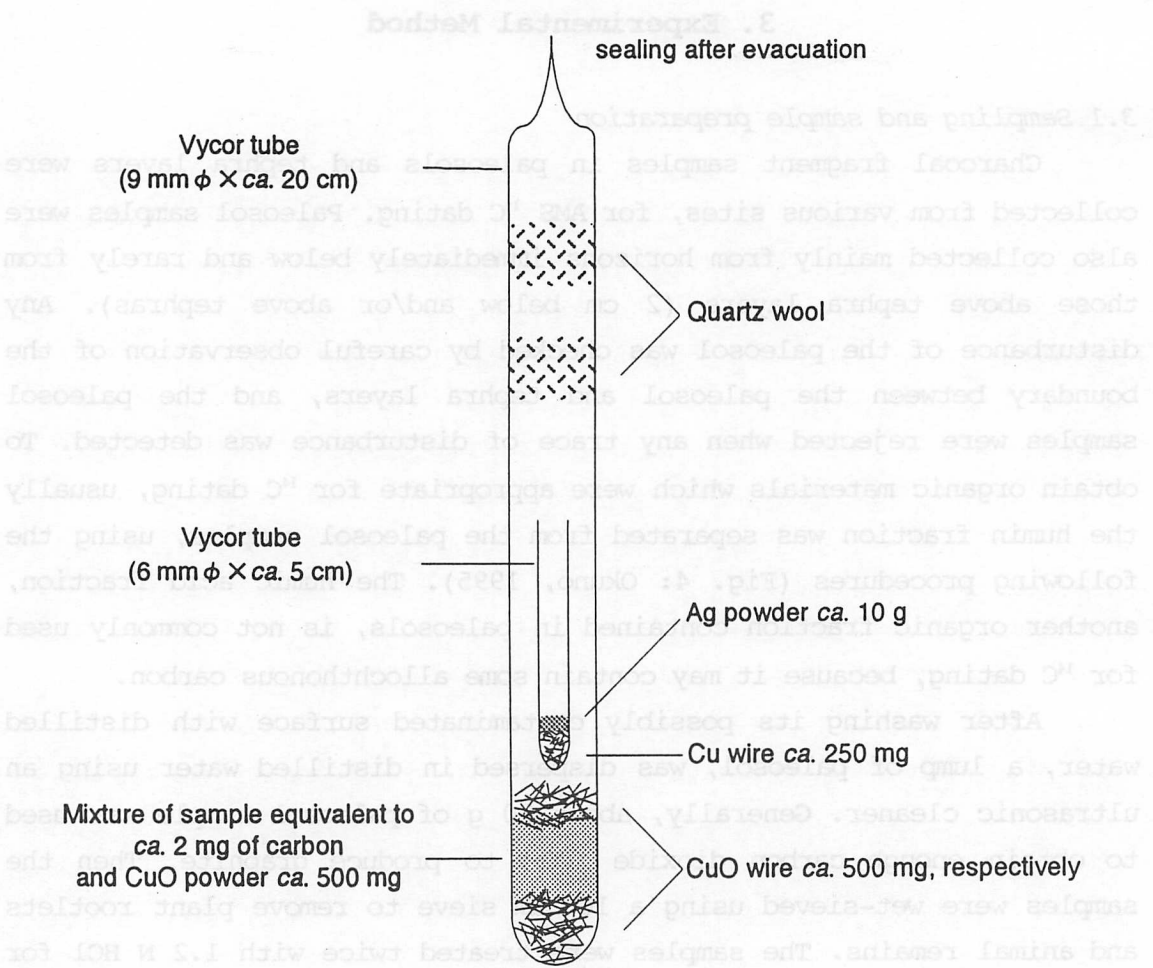


Fig. 5 Schematic diagram of combusting a humin fraction separated from paleosol samples using a Vycor tube.

After physical cleaning and ultrasonic washing charcoal fragments were also purified by a routine acid-alkali-acid (AAA) treatment, in order to remove carbonate and humic acids, which may have affected the charcoal samples while they were in the tephra layers.

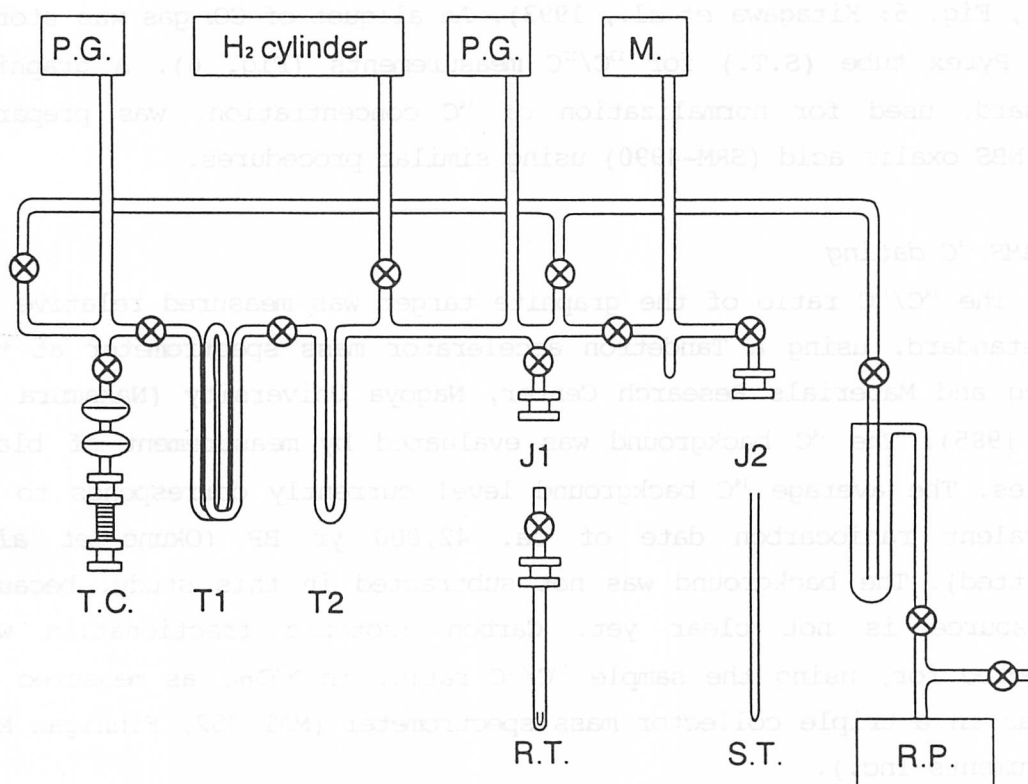


Fig. 6 Vacuum line for the purification and reducing carbon dioxide to graphite catalytically on iron powder with H₂ gas. The heated tube is set at T.C., tube cracking joint. T1 and T2, cryogenic traps. The purified gas for graphitization is sealed in R.T., reduction tube set at J1, cajon o-ring joint. The purified gas for ¹³C/¹²C measurement is sealed in S.T., sealing tube, set at J2, cajon o-ring joint. P.G., pirani gauge. M., manometer. R.P., rotary pump.

A sample of the humin fraction from the paleosol samples, containing about 2 mg of carbon, was oxidized to produce CO₂, by heating at 950 °C for 2 hours in a sealed Vycor tube together with CuO, Cu and Ag (Fig. 5). The other pretreated materials were sealed in a Vycor tube with CuO and oxidized. The resulting CO₂ gas was then purified cryogenically (T1: ethanol slush, -100 °C and T2: normal-pentane slush, -130 °C) in a vacuum line and reduced catalytically to graphite on iron powder, with hydrogen gas in a sealed Vycor tube (R.T., Fig. 6: Kitagawa *et al.*, 1993). An aliquot of CO₂ gas was stored in a Pyrex tube (S.T.) for ¹³C/¹²C measurements (Fig. 6). A graphite standard, used for normalization of ¹⁴C concentration, was prepared from NBS oxalic acid (SRM-4990) using similar procedures.

3.2 AMS ¹⁴C dating

The ¹⁴C/¹³C ratio of the graphite target was measured relative to the standard, using a Tandetron accelerator mass spectrometer at the Dating and Materials Research Center, Nagoya University (Nakamura *et al.*, 1985). The ¹⁴C background was evaluated by measurement of blank samples. The average ¹⁴C background level currently corresponds to an equivalent radiocarbon date of ca. 42,000 yr BP (Okuno *et al.*, submitted). The background was not subtracted in this study, because its source is not clear yet. Carbon isotopic fractionation was corrected for, using the sample ¹³C/¹²C ratio, in δ¹³C_{carb}, as measured on CO₂ gas in a triple collector mass spectrometer (MAT 252, Finnigan Mat Instruments Inc.).

4. Results and Discussion

4.1 Stratigraphy and AMS ¹⁴C dates of the tephra layers in and around the Aira caldera since 30 ka

The stratigraphy and AMS ¹⁴C dates of the tephra layers in and around the Aira caldera since 30 ka are summarized in Fig. 7.

4.1.1 The AT and lower tephra layers

Figure 8 shows representative columnar sections for the Ot, Fm, and Kn Tephtras. The AMS ¹⁴C dates of paleosol samples immediately below these tephtras are shown in Table 1. The relevant ¹⁴C dates of paleosol samples from the Ot and Fm Tephtras, respectively, are scattered a bit

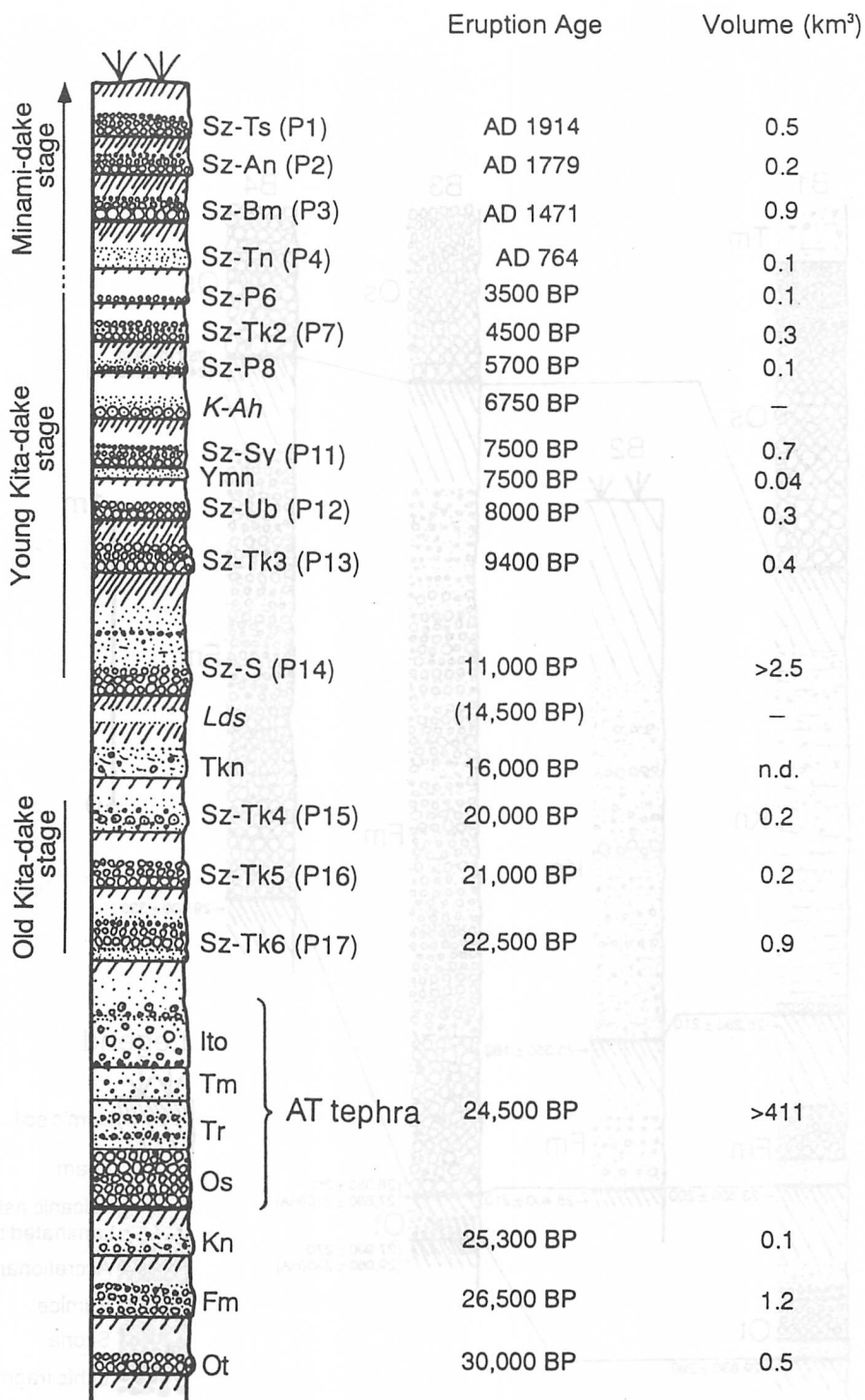


Fig. 7 Summarized columnar section of tephra layers erupted from the Aira caldera during the last 30,000 years (modified from Okuno et al., in press). Eruption age is inferred from ¹⁴C dates as well as the tephra-stratigraphy. Calendar dates are certified by historical documents (Fukuyama, 1978; Kobayashi, 1982). The bulk volume of each tephra is after Nagaoka (1988), Moriwaki (1994) and Kobayashi and Ezaki (unpublished data).

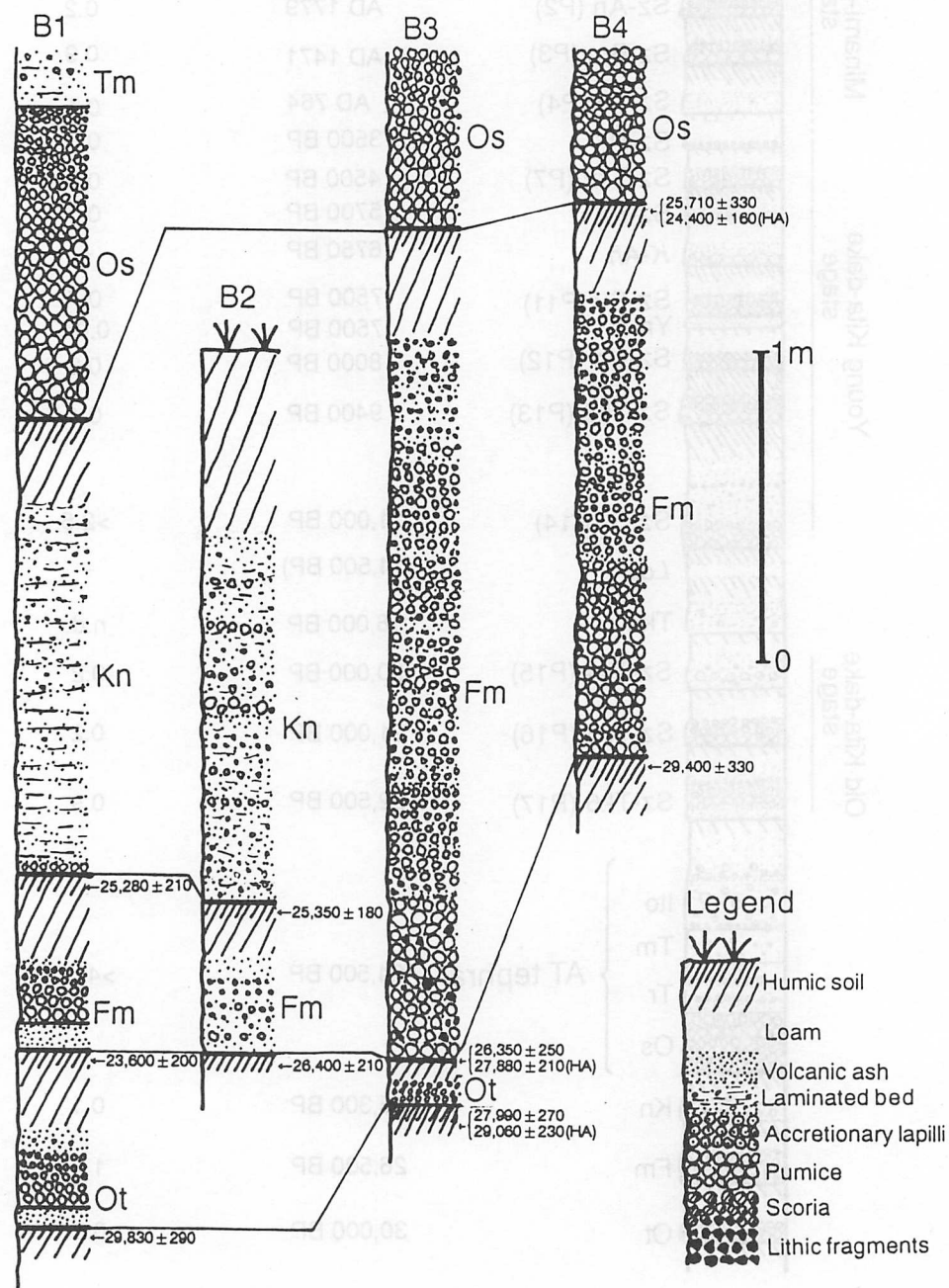


Fig. 8 Representative columnar sections of the Ot, Fm, and Kn Tephra. Locations of sections are shown in Fig. 2. Dates with HA in parentheses are measured for the humic acid fraction.

wider than their one-sigma error ranges. On the other hand, two dates for paleosol samples just below the Kn Tephra show good agreement with each other. However, the dates of the Kn are not completely reliable because their C/N ratios are close to 10, as discussed below (Okuno, 1995; Okuno *et al.*, in press). On the basis of these ^{14}C dates as well as the tephra-stratigraphy, eruption ages are tentatively estimated at 30 ka for the Ot, 26.5 ka for the Fm and 25.3 ka for the Kn Tephras. Nagaoka *et al.* (in press) suggested an eruption age of 25 ka to 30 ka for the Ar Tephra on the basis of its stratigraphic relation with the AT Tephra.

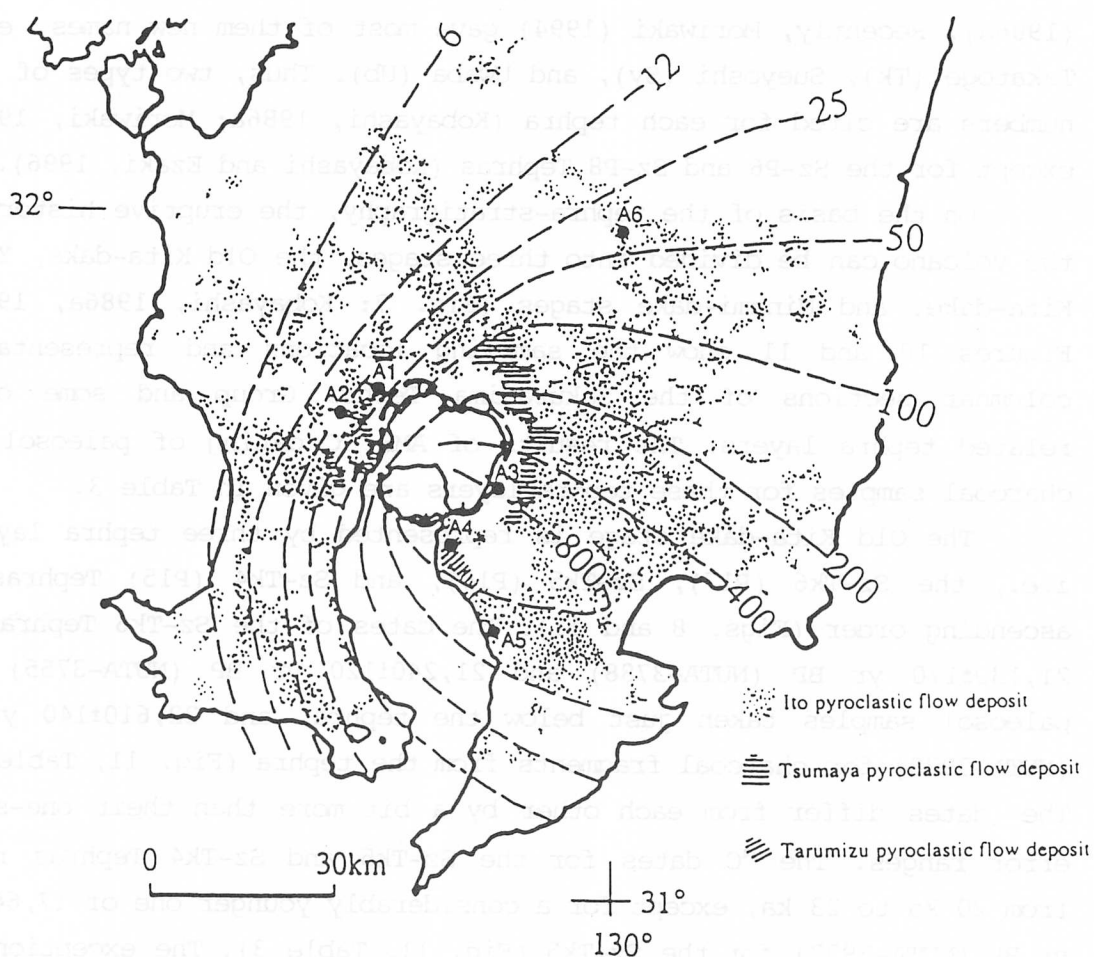


Fig. 9 Isopach map of the Osumi pumice fall deposit (Kobayashi *et al.*, 1983) and distribution of the Tarumizu, Tsumaya, and Ito pyroclastic flow deposits (Aramaki, 1969, 1984; Nagaoka, 1988). Values are in centimeters. A1: Aira IC, A2: Koyamada, A3: Fukaminato, A4: Uratan, A5: Takasu, A6: Sakoma.

Figure 9 shows isopach map of the Osumi pumice fall deposit, and the distribution of the Tarumizu, Tsumaya and Ito pyroclastic flow deposits. Sampling locations for the AT Tephra are also shown in Fig. 9. The AMS ^{14}C dates for the AT Tephra, shown in Table 2, converge on 24.5 ka, with only one exception, *i.e.*, 25,710 \pm 330 yr BP (NUTA-4682). Thus, the eruption age of the AT Tephra is determined to be 24.5 ka.

4.1.2 The Sakurajima Tephra Group

The Sakurajima (Sz) Tephra Group overlies the AT Tephra and intercalates the K-Ah Tephra (Ui and Fukuyama, 1972). The Sakurajima Tephtras are numbered in descending order, from P1 to P17, by Kobayashi (1986a). Recently, Moriwaki (1994) gave most of them new names, *e.g.*, Takatoge (Tk), Sueyoshi (Sy), and Uwaba (Ub). Thus, two types of code numbers are cited for each tephra (Kobayashi, 1986a; Moriwaki, 1994), except for the Sz-P6 and Sz-P8 Tephtras (Kobayashi and Ezaki, 1996).

On the basis of the tephra-stratigraphy, the eruptive history of the volcano can be divided into three stages, the Old Kita-dake, Young Kita-dake, and Minami-dake stages (Fig. 7: Kobayashi, 1986a, 1989). Figures 10 and 11 show the sampling locations and representative columnar sections of the Sakurajima Tephra Group and some other related tephra layers. The results of AMS ^{14}C dating of paleosol and charcoal samples for these tephra layers are given in Table 3.

The Old Kita-dake stage is represented by three tephra layers, *i.e.*, the Sz-Tk6 (P17), Sz-Tk5 (P16), and Sz-Tk4 (P15) Tephtras in ascending order (Figs. 8 and 11). The dates of the Sz-Tk6 Tephtra are 21,130 \pm 170 yr BP (NUTA-3788) and 21,240 \pm 120 yr BP (NUTA-3755) for paleosol samples taken just below the tephra, and 22,610 \pm 140 yr BP (NUTA-3938) for charcoal fragments from the tephra (Fig. 11, Table 3). The dates differ from each other by a bit more than their one-sigma error ranges. The ^{14}C dates for the Sz-Tk5 and Sz-Tk4 Tephtras range from 20 ka to 23 ka, except for a considerably younger one of 17,640 \pm 90 yr BP (NUTA-3937) for the Sz-Tk5 (Fig. 11, Table 3). The exceptionally younger date seems to result from sample contamination by younger carbon. These results indicate that this stage started at about 22.5 ka and ended by 20 ka. Thus the time gap between the Sz-Tk6 and AT eruptions is about 2000 years.

Two geologic units overlie the tephra layers of the Old Kita-dake stage, *i.e.*, the Takano base surge deposit (Tkn) and a loess-derived soil (*Lds*) in ascending order (Figs. 9 and 11). The Tkn Tephtra

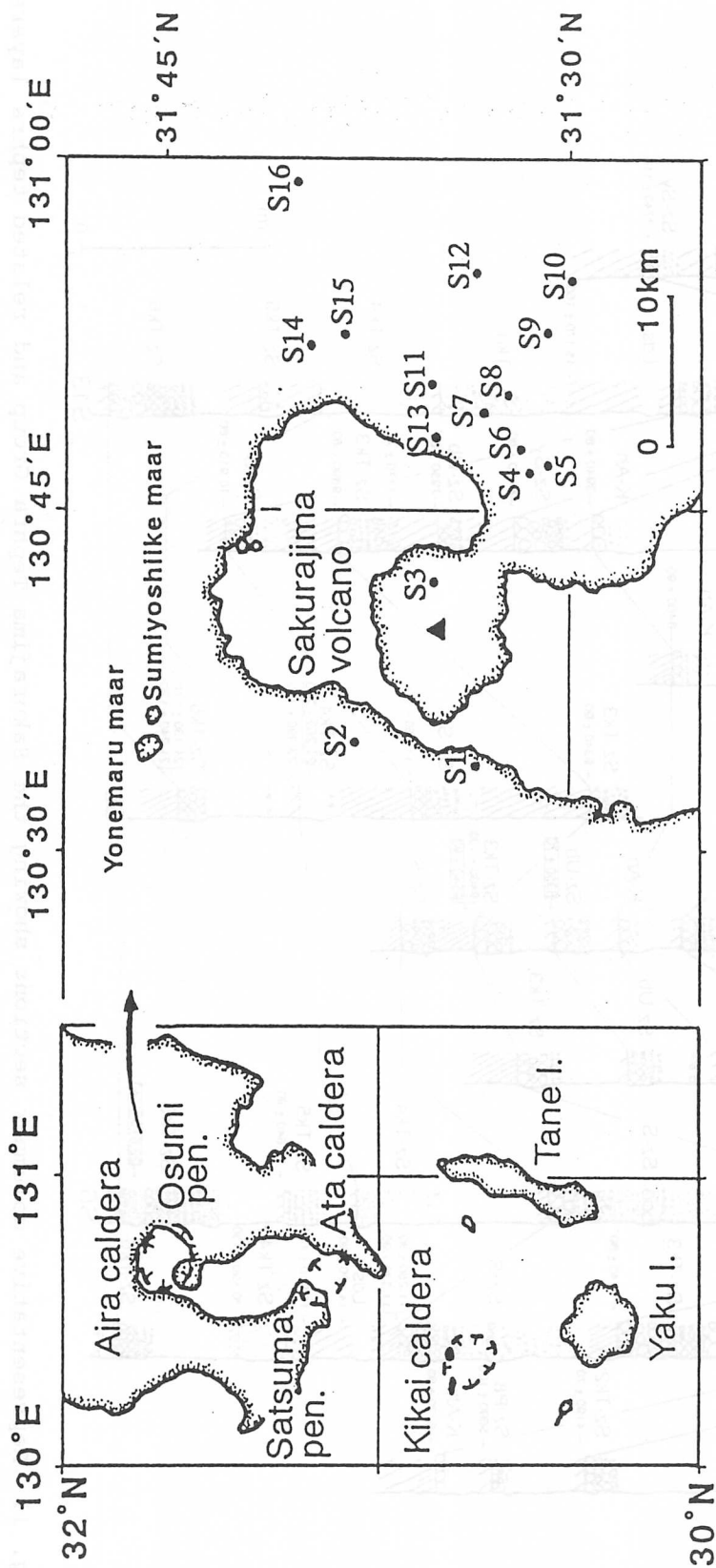


Fig. 10 Sampling locations for the Sakurajima Tephra Group. S1: Shimoarata campus of Kagoshima Univ., S2: Yoshino, S3: Kurokami River, S4: Forestry road of Kagoshima Univ. (Takatoge entrance), S5: Takatoge south, S6: Forestry road of Kagoshima Univ., S7: Dakeno north, S8: Dakeno south, S9: Kamifuruzono, S10: Harabeppu, S11: Uwaba Park, S12: Mae-toko Archaeological Site, S13: Fukaminato, S14: Makinohara, S15: Fujibeizakadan Archaeological Site, S16: Goma

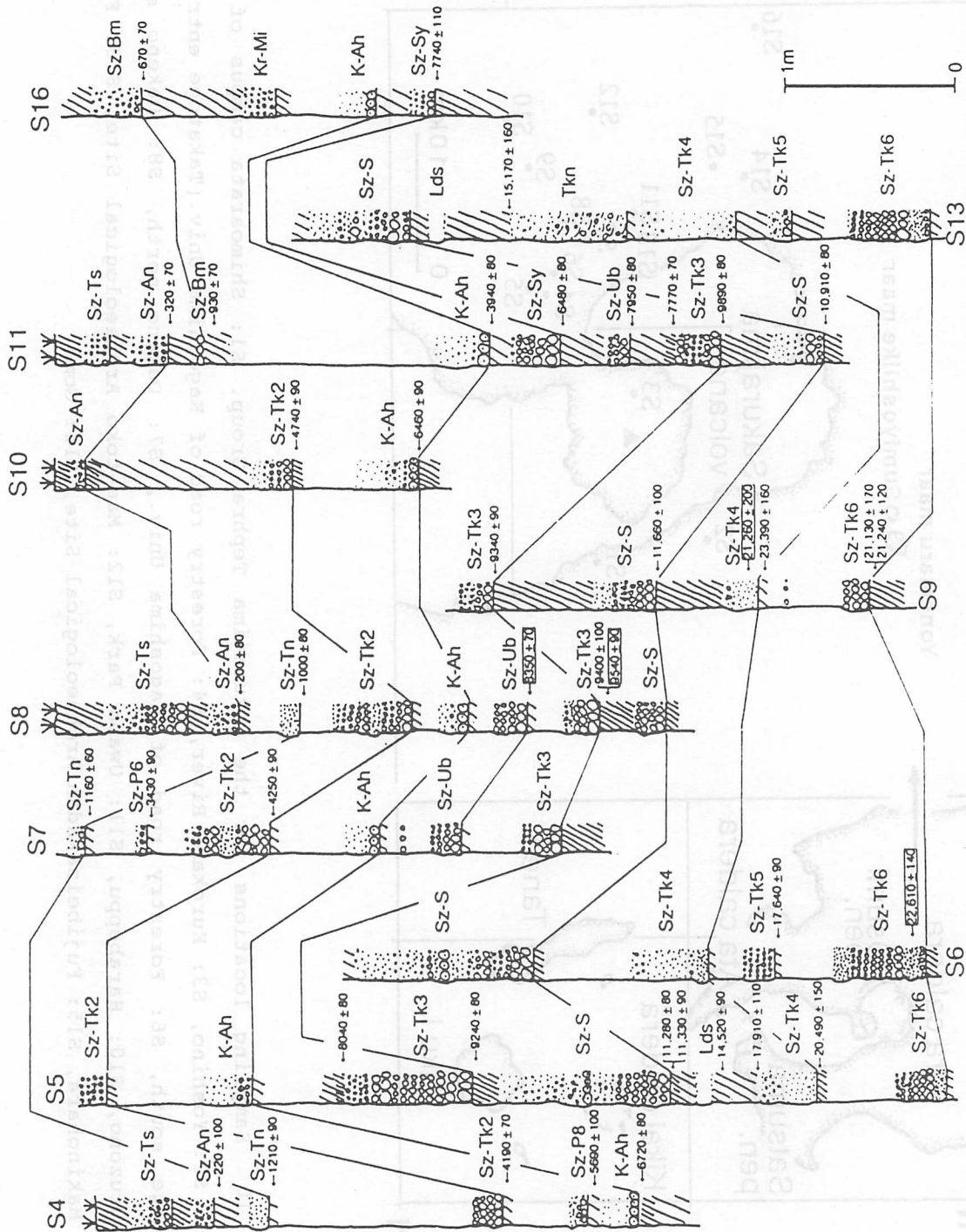


Fig. 11 Representative columnar sections showing the Sakurajima Tephra Group and related tephra layers. Locations of sections are shown in Fig. 10. Framed dates are measured for charcoal samples. Symbols are the same as in Fig. 8.

Table 3 AMS ^{14}C dates of the Sakurajima Tephra Group (modified from Okuno et al., in press)

*Loc.	**Stratigraphic position	Material	C (%)	N (%)	C/N ratio	$\delta^{13}\text{C}_{\text{PDB}}$ (‰)	^{14}C date (yr BP)	Lab no. (NUTA)
S8	Below Sz-An	Paleosol	11.15	0.44	25.2	-17.4	200 ± 70	4072
S4	Below Sz-An	Paleosol	14.84	0.68	21.9	-22.5	220 ± 100	4135
S11	Below Sz-An	Paleosol	9.82	0.35	27.8	-17.2	320 ± 70	3782
S3	In Sz-Bm	Charcoal	—	—	—	-24.4	500 ± 70	4367
S16	Below Sz-Bm	Paleosol	17.71	0.42	42.2	-22.1	670 ± 70	4357
S14	Below Sz-Bm	Paleosol	18.19	0.67	27.2	-16.8	680 ± 70	4136
S11	Below Sz-Bm	Paleosol	12.25	0.41	30.1	-15.2	930 ± 70	4073
S8	Below Sz-Tn	Paleosol	1.61	0.11	14.3	-23.6	1000 ± 80	4079
S7	Below Sz-Tn	Paleosol	1.75	0.09	20.0	-27.4	1160 ± 60	4009
S4	Below Sz-Tn	Paleosol	0.31	n.d.	—	-25.0	1210 ± 90	4148
S7	Below Sz-P6	Paleosol	1.29	0.16	8.2	-28.2	3430 ± 90	4399
S12	Below Sz-Tk2	Charcoal	—	—	—	-29.1	4250 ± 70	4017
S4	Below Sz-Tk2	Paleosol	0.45	n.d.	—	-26.7	4190 ± 70	4124
S7	Below Sz-Tk2	Paleosol	0.35	0.03	11.2	-27.0	4250 ± 70	4008
S10	Below Sz-Tk2	Paleosol	12.64	0.24	52.9	-22.7	4740 ± 90	4318
S4	Below Sz-P8	Paleosol	0.76	0.10	7.4	-26.2	5690 ± 100	4398
S11	Below <i>K-Ah</i>	Paleosol	1.27	0.11	12.0	-24.1	3940 ± 80	4078
S10	Below <i>K-Ah</i>	Paleosol	2.49	n.d.	—	-22.9	6460 ± 90	4333
S4	Below <i>K-Ah</i>	Paleosol	2.34	0.11	21.7	-16.5	6720 ± 80	4150
S11	Below Sz-Sy	Paleosol	0.46	0.04	12.0	-25.3	6480 ± 80	3758
S16	Below Sz-Sy	Paleosol	1.88	n.d.	—	-22.2	7740 ± 110	4237
S15	Below Ynm	Charcoal	—	—	—	-27.0	7480 ± 80	4300
S11	Below Sz-Ub	Paleosol	1.18	0.05	23.8	-20.7	7950 ± 80	3757
S8	Below Sz-Ub	Charcoal	—	—	—	-26.0	8350 ± 70	3943
S11	Above Sz-Tk3	Paleosol	3.05	0.19	16.1	-19.7	7770 ± 70	4080
S5	Above Sz-Tk3	Paleosol	1.66	0.07	23.0	-19.7	8040 ± 80	3940
S5	Below Sz-Tk3	Paleosol	8.07	0.26	31.5	-21.7	9240 ± 80	3875
S9	Below Sz-Tk3	Paleosol	6.02	0.29	20.5	-21.9	9340 ± 90	4036
S8	Below Sz-Tk3	Paleosol	5.99	0.18	32.8	-20.6	9400 ± 100	4235
S8	Below Sz-Tk3	Charcoal	—	—	—	-26.8	9540 ± 90	4035
S11	Below Sz-Tk3	Paleosol	5.08	0.14	36.0	-23.1	9890 ± 80	3756

erupted from Wakamiko caldera in the northeastern part of Aira caldera (Kobayashi, 1986a). A ^{14}C date of 11,800±140 yr BP (Gak-16276), by the beta counting method (Moriwaki, 1994), was given by charcoal collected from the Tkn Tephra. However, this date is much younger than that inferred from its stratigraphic position. An AMS ^{14}C date of 15,170±160 yr BP (NUTA-4689), for the paleosol sample just above the Tkn Tephra,

Table 3 AMS ¹⁴C dates of the Sakurajima Tephra Group (cont.)

*Loc.	**Stratigraphic position	Material	C (%)	N (%)	C/N ratio	* δ ¹³ C _{PDB} (‰)	¹⁴ C date (yr BP)	Lab no. (NUTA)
S2	In Sz-S	Charcoal	—	—	—	-23.8	{ 10,670 ± 100 11,050 ± 120	4634 4642
S11	Below Sz-S	Paleosol	5.52	0.23	24.4	-22.7	10,910 ± 80	3874
S5	Below Sz-S	Paleosol	3.56	0.15	23.1	-21.4	{ 11,280 ± 80 11,330 ± 90	3878 4025
S9	Below Sz-S	Paleosol	2.16	0.10	21.0	-20.7	11,660 ± 100	3868
S1	Below Sz-S	Paleosol	5.16	0.18	29.4	-19.5	11,850 ± 90	3561**
S1	Below Sz-S	Paleosol,HA	—	—	—	-20.4	11,170 ± 80	3548**
S1	1m below Sz-S	Paleosol	5.42	0.20	29.4	-21.3	12,110 ± 90	3595**
S5	Below <i>Lds</i>	Paleosol	1.81	0.14	13.2	-22.2	14,520 ± 90	4356
S13	Above Tkn	Paleosol	0.91	0.07	12.7	-21.6	15,170 ± 160	4689
S5	Above Sz-Tk4	Paleosol	2.65	0.12	22.7	-25.1	17,910 ± 110	4350
S9	In Sz-Tk4	Charcoal	—	—	—	(-25)	21,260 ± 200	4323
S5	Below Sz-Tk4	Paleosol	1.43	0.09	15.7	-23.4	20,490 ± 150	3869
S9	Below Sz-Tk4	Paleosol	0.61	n.d.	—	-21.1	23,390 ± 160	4397
S6	Below Sz-Tk5	Paleosol	0.44	0.05	8.3	-21.1	17,640 ± 90	3937
S6	In Sz-Tk6	Charcoal	—	—	—	-25.3	22,610 ± 140	3938
S9	Below Sz-Tk6	Paleosol	1.97	0.09	22.1	-20.3	{ 21,130 ± 170 21,240 ± 120	3788 3755

* See Fig. 10

** See Figs. 7 and 11

δ ¹³C value in parenthesis is assumed.

Data from Okuno et al. (1996a)

n.d. not detected

is consistent with the dates for the paleosol samples from horizons above and below it (Fig. 11, Table 3). This date should be considered the upper limit of the eruption age range. Consequently, the age of the Tkn Tephra is tentatively estimated at 16 ka (Okuno, in press). The *Lds* layer is regarded as a loess-derived soil by Naruse et al. (1994).

The Young Kita-dake stage is represented by seven tephra layers in Osumi Peninsula, *i.e.*, the Sz-S (P14), Sz-Tk3 (P13), Sz-Ub (P12), Sz-Sy (P11), Sz-P8, Sz-Tk2 (P7), and Sz-P6 Tephtras, in ascending order (Figs. 8 and 11). The Sz-S Tephra, with a bulk volume of more than 2.5 km³, consists of pyroclastic surge and pumice fall deposits (Kobayashi, 1986a; Moriwaki, 1992). The present ¹⁴C dating (Table 3) gives an eruption age of 11 ka for the Sz-S Tephra. A gap of 500 years between

the present AMS ^{14}C age and that (10.5 ka) estimated by Machida and Arai (1992) is probably due to some systematic difference in the method of detecting ^{14}C . Frequent eruptions in this stage have occurred at intervals of 800 to 2000 years, following the eruption of the Sz-S Tephra. The Yonemaru scoria (Ynm), which immediately underlies the Sz-Sy Tephra, erupted from Yonemaru maar (Fig. 2: Moriwaki et al., 1986; Moriwaki, 1994). Hornblende phenocrysts derived from the Ikedako Tephra (Ik: Naruo and Kobayashi, 1980), whose ^{14}C age is dated at 5640 ± 30 yr BP (Okuno et al., 1996b), were detected in the horizon above the Sz-P8 Tephra (Kobayashi and Ezaki, 1996). A ^{14}C date of 5690 ± 100 yr BP (NUTA-4398) for the paleosol just below the Sz-P8 Tephra is consistent with it. The series of eruptions of the Young Kita-dake stage ended with the Sz-P5 Tephra, distributed only on the north slope of Sakurajima volcano (Kobayashi, 1986a). A ^{14}C date of 4840 ± 110 yr BP (Gak-10020) for wood charcoal collected from the Sz-P5 was reported by Kobayashi (1986a). However, this study gave far younger ages for the Sz-P6 and Sz-Tk2 Tephtras (Fig. 7) which underlie the Sz-P5 Tephra, i.e., 3.5 ka and 4.5 ka, respectively. This disagreement may not be caused by the difference in the dating methods, but by misidentification of the tephra layers in Sakurajima and the Osumi Peninsula.

The Minami-dake stage started with intermittent Vulcanian eruptions which formed a volcanic sand layer in the proximal area of the Minami-dake (Kobayashi, 1986b). A ^{14}C date of 4050 ± 120 yr BP (I-15284) was reported for the paleosol just below the sand layer (Kobayashi, 1986b; Kobayashi and Ezaki, 1996). This date cannot be the age of the beginning of the Minami-dake stage because the ^{14}C age of the Sz-P6 Tephra of the Young Kita-dake stage is estimated at 3430 ± 90 yr BP (NUTA-4399). Thus the ^{14}C date for the above mentioned sand layer should be regarded as the lower chronological limit of this layer. The lowest tephra of this stage, observed at Osumi Peninsula, is the Sz-Tn (P4) Tephra. Several large-scale eruptions are recorded in historical documents (Fukuyama, 1978; Kobayashi, 1982). They are the eruptions of AD 764 (Tenpyo era: Tn), AD 1471-1476 (Bunmei era: Bm), AD 1779 (An-ei era: An) and AD 1914 (Taisho era: Ts). The ^{14}C dates for the first three eruptions were calibrated to calendar years using the Calib ETH 1.5b program (Niklaus, 1991; Niklaus et al., 1992) based on dendro-chronological calibration data (Pearson and Qua, 1993), and are summarized in Table 4. Each calibrated age range substantially agrees

Table 4 Calibrated ^{14}C dates for historical eruptions of the Sakurajima volcano (after, Okuno et al., in press)

*Historical date	Material	^{14}C date (yr BP)	Cal range AD/ probability (%)	Lab no. (NUTA)
An-ei (AD1779)	Paleosol	200 ± 70	1652-1687 (25.3) 1732-1827 (74.7)	4072
An-ei (AD1779)	Paleosol	220 ± 100	1522-1553 (12.7) 1638-1689 (27.3) 1725-1841 (60.0)	4135
An-ei (AD1779)	Paleosol	320 ± 70	1481-1567 (59.0) 1578-1606 (17.4) 1620-1656 (23.6)	3782
Bunmei (AD1471)	Charcoal	500 ± 70	1310-1335 (14.4) 1393-1472 (85.6)	4367
Bunmei (AD1471)	Paleosol	670 ± 70	1282-1313 (32.2) 1334-1394 (67.8)	4357
Bunmei (AD1471)	Paleosol	680 ± 70	1275-1310 (36.1) 1335-1393 (63.9)	4136
Bunmei (AD1471)	Paleosol	930 ± 70	1032-1171 (100.0)	4073
Tenpyo (AD764)	Paleosol	1000 ± 80	972-1060 (54.8) 1076-1129 (28.4) 1131-1161 (16.8)	4079
Tenpyo (AD764)	Paleosol	1160 ± 60	821-846 (15.3) 851-967 (84.7)	4009
Tenpyo (AD764)	Paleosol	1210 ± 90	712-749 (16.3) 751-898 (76.0) 917-926 (3.8) 948-957 (3.9)	4148

* Based on documentary records

with its calendar date, based on historical records. The present results suggest that the Sz-Tn (P4) Tephra corresponds to that documented for AD 764 (Okuno, 1996; Okuno et al., in press). The ^{14}C dates seem to provide an important chronological constraint to correlate tephra layers with documented historical eruptions.

4.1.3 Relation between volume and preceding repose period of the eruption in the Aira caldera

It is remarkable that the AT eruption formed more than 97 percent of volume of the tephra which were erupted from the Aira caldera during the last 30,000 years. The lengths of the repose periods before and after the AT eruption are estimated as 800 and 2000 years, respectively. These periods correspond to the repose periods for the Young Kita-dake stage.

Figure 12 shows a cumulative volume plotted against ^{14}C age for the Sakurajima Tephra Group. A straight line best-fitted to the data is given by

$$V = 6.88 - 2.754 \times 10^{-4} \times A,$$

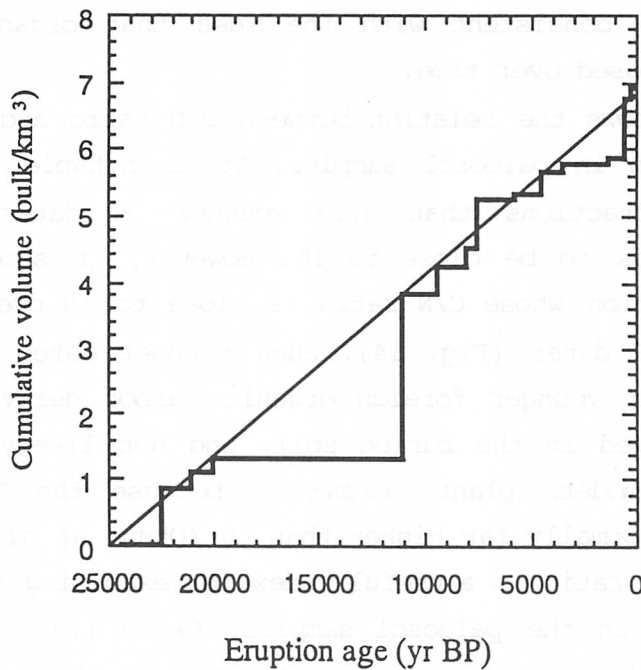


Fig. 12 Cumulative volume plotted against eruption age for the Sakurajima Tephra Group. Eruption age and bulk volume of each tephra are shown in Fig. 7.

where A is the eruption age (yr BP) of the tephra, and V is the cumulative bulk volume (km^3) of the erupted tephras. The cumulative bulk volume of the Sakurajima Tephra Group is related to the length of its preceding repose period (Fig. 12). However, lava flows and small-scale tephras are not taken into account in the above estimation, because of burial and destruction by younger eruptive materials as well as submergence under the sea.

4.2 Geochemical properties of paleosol samples

Geochemical properties of paleosol samples obtained in the course of ^{14}C dating are discussed below. The carbon content of the humin fraction in paleosol samples ranges widely from 0.17 to 18.19 % (Tables 1, 2, and 3). Figure 13 shows the relation between carbon content and ^{14}C date of the humin fraction in paleosol samples. The carbon content ^{14}C dates are younger than 5000 yr BP for those paleosol samples whose humin fraction shows a carbon content greater than 10 %. These results are consistent with the idea that organic fractions become more decomposed over time.

Figure 14 shows the relation between C/N ratio and ^{14}C date for the humin fraction in paleosol samples. It is notable that the C/N ratio of humin fractions that give younger ^{14}C dates than their expected ages tends to be close to 10. However, it should be noted that a humin fraction whose C/N ratio is close to 10 does not always produce younger ^{14}C dates (Fig. 14). Such younger dates are probably due to addition of younger foreign organic carbon derived from soil organisms that lived in the buried soil, and not likely from sample contamination by modern plant fragments, because the C/N ratio of modern plants is normally far higher than 10 (Okuno *et al.*, in press). Therefore, the C/N ratio is a useful index for examining the source of organic materials in the paleosol samples. Okuno (1995) has already observed this tendency, for organic materials in the paleosols just above the Sz-S Tephra (Fig. 15), within the Komoriko Tephra Group (K-Km: Okuno *et al.*, 1994; Okuno, 1996) which is distributed on the northern rim of the Kikai caldera (Fig. 1). The relation between C/N ratio and ^{14}C date for the humin fraction of paleosol and charcoal samples immediately below the Kirishima Miike Tephra (Kr-Mi in Fig. 11) is shown in Fig. 16 (Okuno *et al.*, 1996c). It seems that the ^{14}C dates whose C/N ratio is as small as 10 are sometimes younger than the expected ages. In addition, the older ^{14}C date (NUTA-4238) measured for

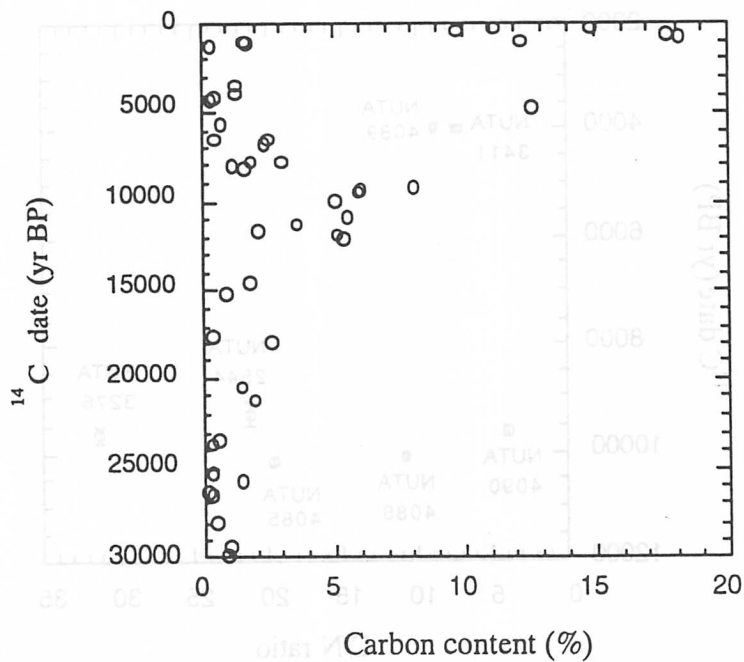


Fig. 13 Relation between carbon content and ^{14}C date of the humin fraction in paleosol samples.

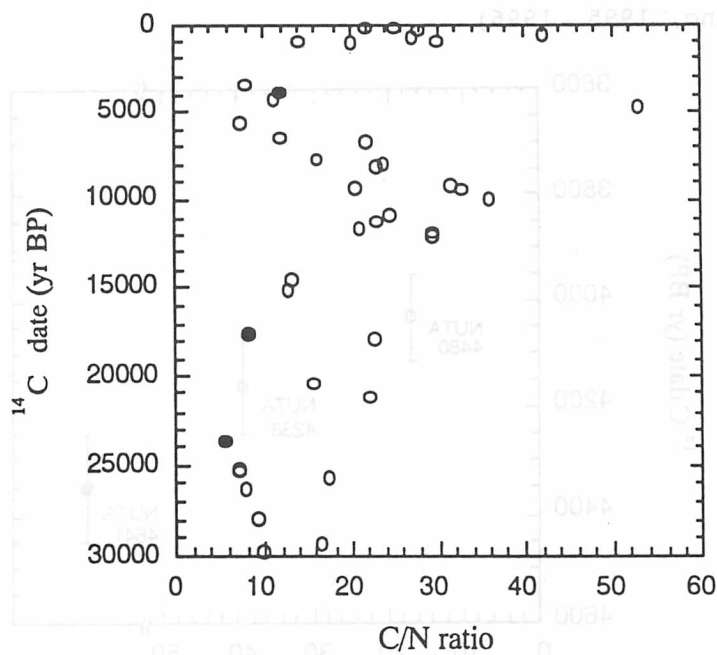


Fig. 14 Relation between C/N ratio and ^{14}C date of the humin fraction in paleosol samples. Solid circles indicate those ^{14}C dates that are considerably younger than ages inferred from the stratigraphic position.

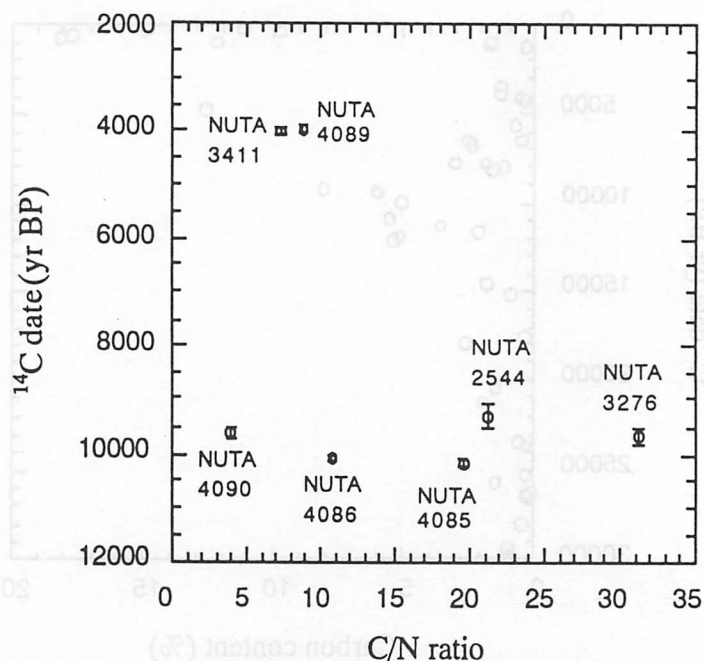


Fig. 15 Relation between C/N ratio and ¹⁴C date of the humin fraction in paleosol samples just above the Sz-S Tephra intercalated in the Kikai-Komoriko Tephra Group (K-km: data from Okuno, 1995, 1996)

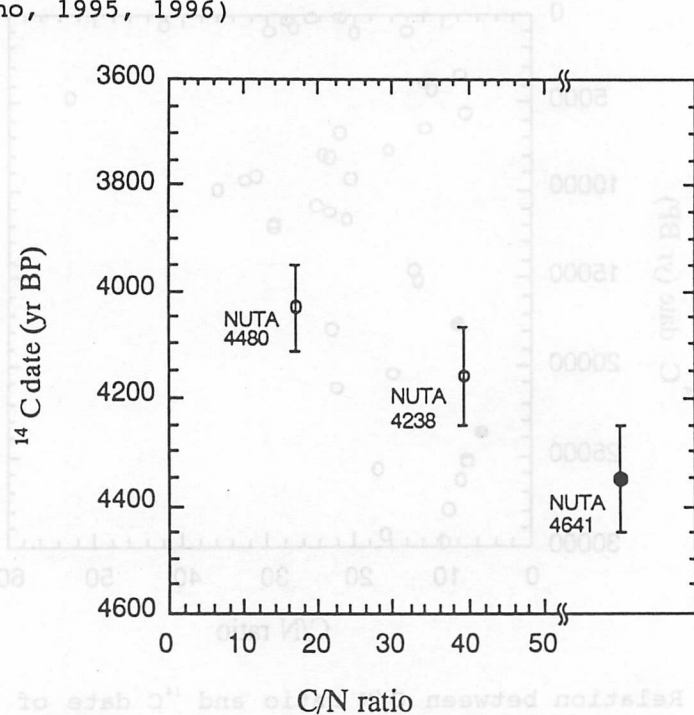


Fig. 16 Relation between C/N ratio and ¹⁴C date for the humin fraction (○) in paleosol and charcoal (●) samples immediately below the Kr-Mi Tephra (data from Okuno et al., 1996c).

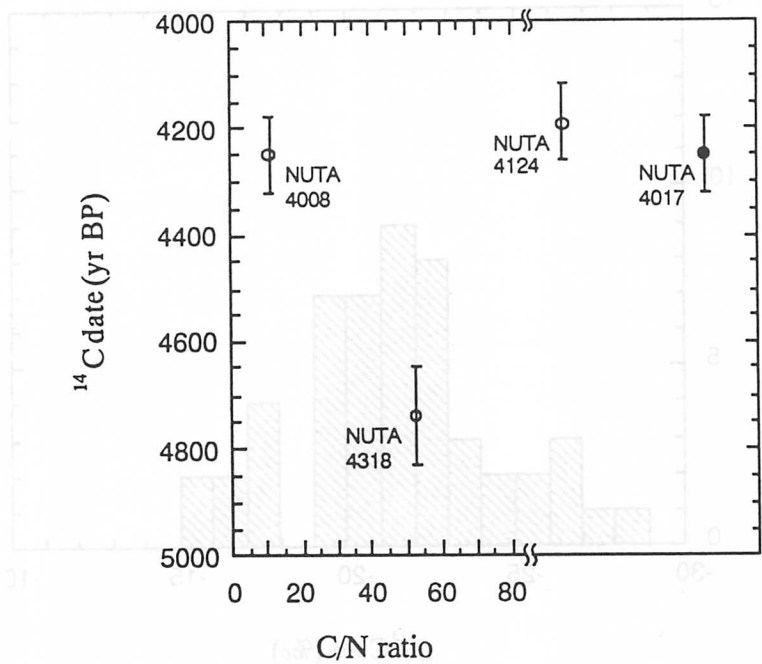


Fig. 17 Relation between C/N ratio and ¹⁴C date for the humin fraction (○) in paleosol and charcoal (●) samples immediately below the Sz-Tk2 Tephra.

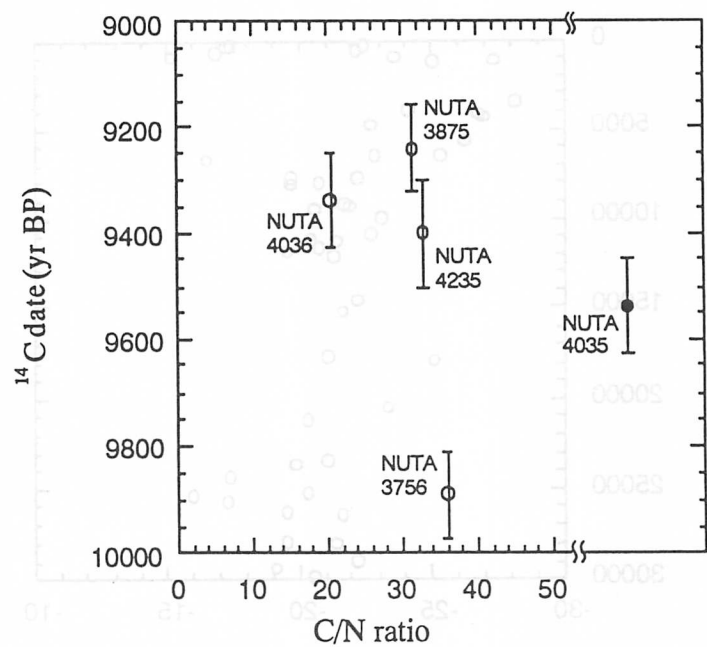


Fig. 18 Relation between C/N ratio and ¹⁴C date for the humin fraction (○) in paleosol and charcoal (●) samples immediately below the Sz-Tk3 Tephra.

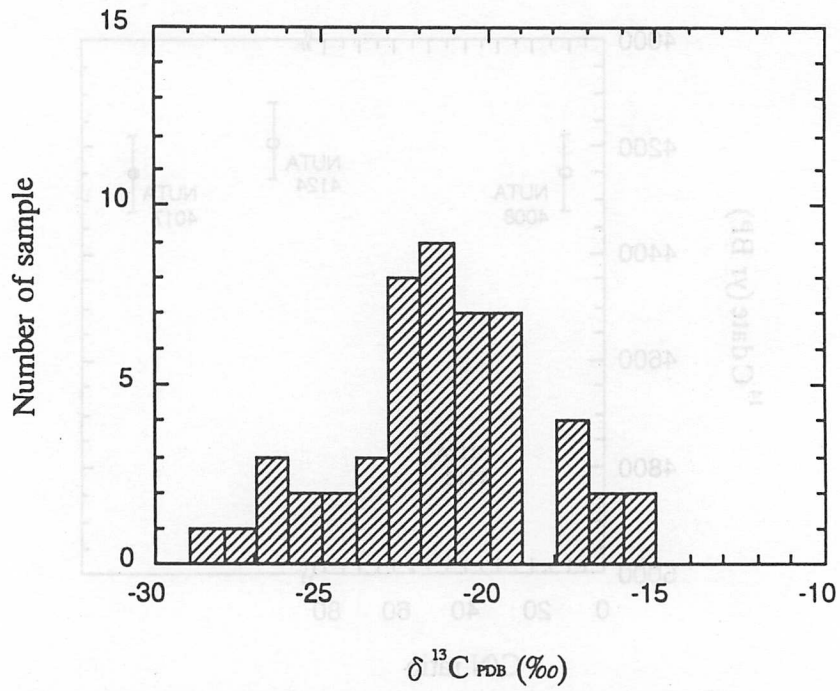


Fig. 19 Frequency histogram of the $\delta^{13}\text{C}$ value for paleosol samples.

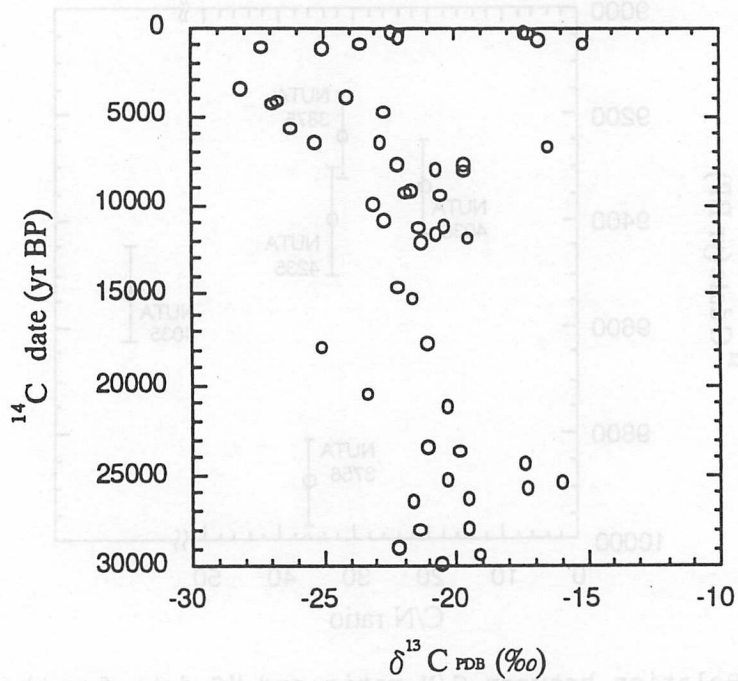


Fig. 20 Relation between the $\delta^{13}\text{C}$ value and ^{14}C date for the paleosol samples.

the paleosol sample just below the tephra layer agrees with the ^{14}C date (NUTA-4641) for charcoal, within one-sigma errors (Fig. 16). For the paleosol samples just below the Sz-Tk2 and Sz-Tk3 Tephtras, one ^{14}C date is rather older than the others as shown in Figs. 17 and 18, respectively. The C/N ratios of the humin fraction of these paleosol samples for the Sz-Tk2 and Sz-Tk3 are far larger than 10, indicating no younger-carbon contamination by soil organisms as discussed above. The older ^{14}C dates probably result from contamination by soil organic materials in the lower horizons. The same explanation may be applied to the case in which the calibrated age range of cal AD 1032-1171 (NUTA-4073) for the Sz-Bm (P3) Tephra is notably older than its historical date (Table 4).

The stable carbon isotopic ratio, $\delta^{13}\text{C}_{\text{PDB}}$, of paleosol samples varies widely from -28.2 to -15.2 ‰ (Tables 1, 2, and 3). Figure 19 shows the frequency histogram for $\delta^{13}\text{C}$ value of the humin and humic acid fractions from paleosol samples. This shows a peak at -21 ‰, which is higher by a few permil than the typical value for C3 plants. Figure 20 shows the relation between $\delta^{13}\text{C}$ value and ^{14}C date of paleosol samples. It appears that the values of $\delta^{13}\text{C}$ converge to -21 ‰ with increasing ^{14}C age.

4.3 Geological interpretation of paleosol ^{14}C dates

Humin and humic acid fractions in the paleosol samples are dated separately in this study (see Chapter 3). There is a difference greater than their one-sigma errors between dates for the two fractions of each sample (Fig. 21), e.g., 26,350±250 yr BP (NUTA-4837) and 27,880±250 yr BP (NUTA-4830), 27,990±270 yr BP (NUTA-4836) and 29,060±230 yr BP (NUTA-4829), 25,710±330 yr BP (NUTA-4682) and 24,400±160 yr BP (NUTA-4828), 11,850±90 yr BP (NUTA-3561) and 11,170±80 yr BP (NUTA-3548). However, there is no systematic tendency in ^{14}C dates between the two fractions. This implies that the humic acid fraction contains some allochthonous carbon.

The ^{14}C dates, 7950±80 yr BP (NUTA-3757) and 7770±70 yr BP (NUTA-4080), of two paleosol samples from different horizons within a single layer, which is interbedded by the Sz-Tk3 and Sz-Ub Tephtras at Loc. S11 (Fig. 10), agree within two-sigma errors, although the two sampled horizons are separated vertically by 40 cm (Fig. 11). This agreement of the two ^{14}C dates indicates that the organic matter in both samples was put simultaneously in a closed system. On the other hand, a date

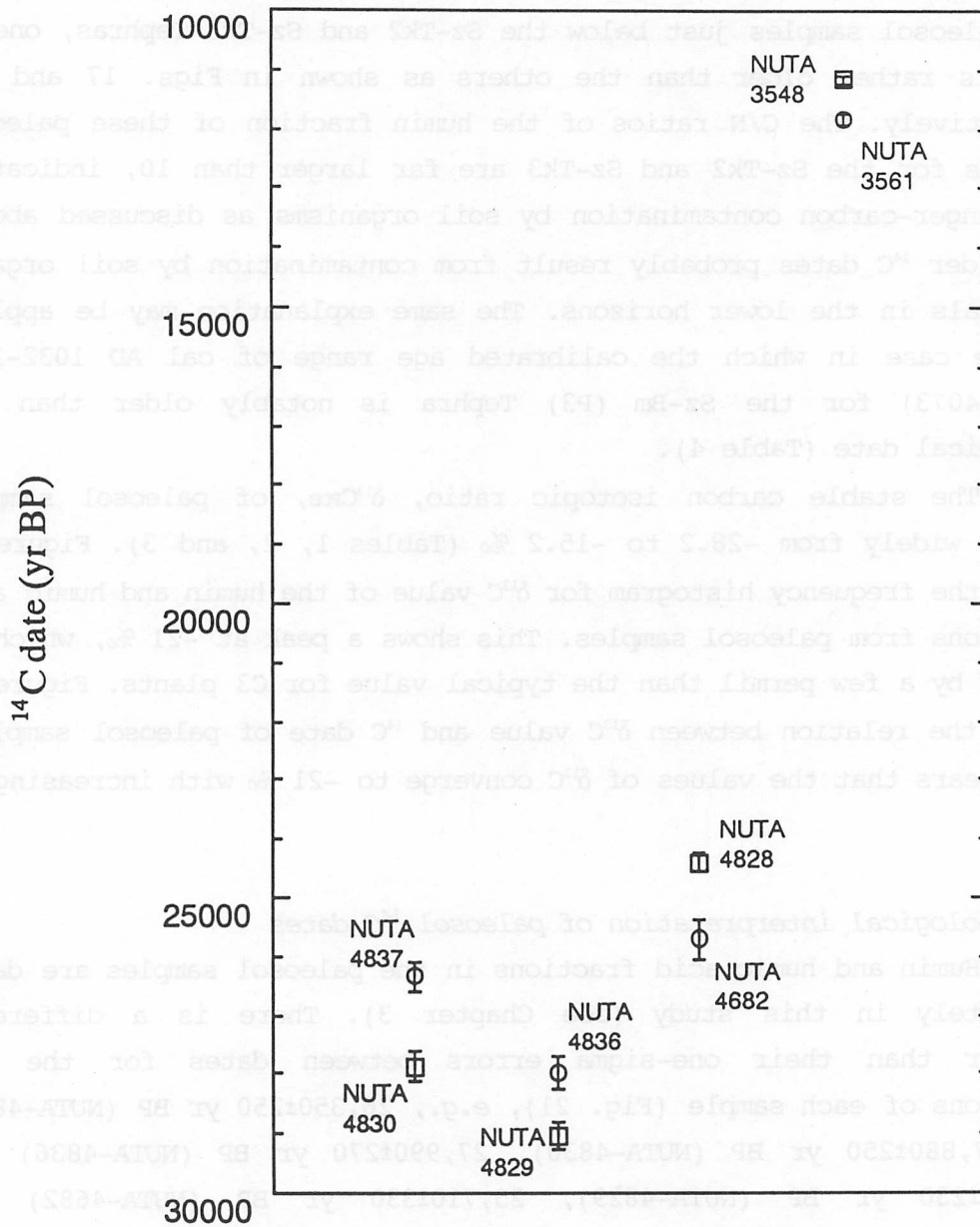


Fig. 21 Comparison of ¹⁴C dates between the humin (○) and the humic acid (□) fractions separated from each paleosol sample.

of 9890 ± 80 yr BP (NUTA-3756) for the paleosol sample immediately below the Sz-Tk3 Tephra at Loc. S11 (Fig. 10) is significantly older than those mentioned above. There is a similar difference in dates for paleosols above and below the Sz-Tk3 Tephra at Loc. S5 (Fig. 10), *i.e.*, 8040 ± 80 yr BP (NUTA-3940) for the paleosol just above the Sz-Tk3 Tephra and 9240 ± 80 yr BP (NUTA-3875) for the paleosol just below the Sz-Tk3 Tephra (Fig. 11). These facts suggest that the tephra layer serves as a strong cover, preventing the vertical movement of organic matter to the underlying paleosol layer. The ^{14}C dates of paleosol samples from different localities in the underlying paleosol layers of these tephtras are concentrated in narrow time intervals, *e.g.*, from 10,910 to 11,850 yr BP for the Sz-S Tephra and from 9240 to 9890 yr BP for the Sz-Tk3 Tephra (Fig. 17) *etc.*, and are also consistent with the ^{14}C dates of charcoal and the historical records (Tables 3 and 4). Therefore, ^{14}C dates of paleosol samples represent the time when the tephra layer covered the paleosol, namely, its eruption age. It may be said that more frequent tephra deposition gives higher time resolution. There should be some cases in which more recent original carbon contaminates paleosol samples, as mentioned above. However, old-carbon contamination can be detected by comparing the ^{14}C date with the tephra-stratigraphy. Therefore, we can estimate a reasonable eruption age for every tephra layer from the ^{14}C dates of its underlying paleosol layer.

5. Conclusion

This study shows that a paleosol is very useful for obtaining a high-resolution ^{14}C chronology of major volcanic eruptions. The advantages of using paleosol samples for ^{14}C dating are summarized as follows. First, paleosol samples are collected easily and systematically. This implies that the eruption age of every tephra layer can be dated, provided that a soil was formed in repose interval between two successive eruptions. Second, ^{14}C dates for paleosol samples have a smaller chronological uncertainty than those for charcoal samples which are usually used as samples for ^{14}C dating. Some charcoal fragments are so resistant to weathering that charcoal fragments carbonized by earlier volcanic deposits could be included and preserved in younger volcanic deposits. The ^{14}C dates of the humin

fraction from paleosol samples just below tephra layers represent the time when the tephras covered the paleosols, namely, their eruption ages. A tephra layer is a useful marker for detecting bio-turbation in the paleosol layers, as well as a good cover which prevents vertical movement of soil organic matter. Better time resolution is expected in cases of frequent tephra deposition, from the viewpoint of ^{14}C dating of paleosol samples. C/N ratios serve as an indicator of the source of contaminants of the humin fraction in paleosol samples.

A high-resolution ^{14}C chronology of the last 30,000 years (Fig. 7) is compiled for the Aira caldera, on the basis of ^{14}C dates of the tephra layers. Since the Otsuka eruption of 30 ka, at least four pyroclastic eruptions occurred intermittently in the eastern part of the Aira caldera. The AT eruption occurred at about 24.5 ka.

Following the AT eruption, the Sz-Tk6 Tephra erupted from Sakurajima volcano at 22.5 ka. The time gap between the Sz-Tk6 and AT eruptions is 2000 years. The Old Kita-dake stage ended at 20 ka. At 16 ka, the Tkn Tephra was erupted from the northeastern part of the Aira caldera. After a 9000 year break for Sakurajima volcano, the Young Kita-dake stage began with the Satsuma eruption at 11 ka. This stage continued until 3.5 ka with eruptions at an 800 to 2000 year recurrence interval. For historical eruptions in the Minami-dake stage, the calibrated age ranges are consistent with calendar dates, based on the historical records, and provide an important chronological constraint when correlating tephra layers with documentary records.

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