# A significant middle Pleistocene tephra deposit preserved in the caves of Mulu, Borneo

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A B S T R A C T

A distinctive white sediment in the caves of Mulu, Sarawak, Borneo is a well-preserved tephra, representing a fluvially transported surface air-fall deposit, re-deposited inside the caves. We show that the tephra is not the Younger Toba Tephra, formerly considered as most likely. The shards are rod-shaped with elongate tubular vesicles; the largest grains ~170 μm in length; of rhyolitic composition; and $^{87}Sr/^{86}Sr$ ratio of $0.70426 \pm 0.00001$. U–Th dating of associated calcites suggest that the tephra was deposited before $125 \pm 4$ ka, and probably before $156 \pm 2$ ka. Grain size and distance from closest potential source suggests an eruption of VEI 7. Prevailing winds, grain size, thickness of deposit, location of potential sources, and Sr isotopic ratio limit the source to the Philippines. Comparisons with the literature give the best match geochemically with layer 1822 from Ku et al. (2009a), dated by ocean core stratigraphy to 189 ka. This tephra represents a rare terrestrial repository indicating a very substantial Plinian/Ultra-Plinian eruption that covered the Mulu region of Borneo with ash, a region that rarely receives tephra from even the largest known eruptions in the vicinity. It likely will be a valuable chronostratigraphic marker for sedimentary, palaeontological and archaeological studies.

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## Introduction

Gunung Mulu National Park (GMNP) is located in equatorial northern Sarawak, Malaysia (island of Borneo) at 4 02′N, 114 48′E. The Park was established in 1975 in recognition of its spectacular karst geomorphology, and, in particular, its extensive cave systems (Fig. 1). The region was first scientifically explored by British cavers and academics in the 1960s–80s (e.g., Wilford, 1964; Wilford and Wall, 1965; Brook and Waltham, 1979; Laverty, 1980; Sweeting, 1980; Eavis, 1981) and is the focus of continued fruitful exploration today (http://www.mulucaves.org/wordpress/). The caves are amongst the largest in the world (Gillieson and Clark, 2010) and in many the sedimentary fill is substantial, some 20 m or more thick. A distinctive white sediment was noted in the early reports (Bull and Laverty, 1981; Laverty, 1982) and, on the basis of a positive test for allophane (a typical product of tephra breakdown) in one sample from the Clearwater system, identified as volcanic ash. However, this study was never followed up.

The ash layer is significant because it is so well-preserved, and accessible, in the Mulu cave sediments in contrast to the surrounding alluviated and vegetated landscapes. It is also significant because the centers of Quaternary volcanic activity are quite distant (e.g., the

Philippine arc is ~1200 km distant, and the Sunda Islands ~1400 km); hence, it is reasonable to assume that it originated in an eruption of unusual size and violence that probably caused significant ecosystem changes. This well-preserved terrestrial record of a significant ultra-plinian volcanic event represents a potentially important chronostratigraphic marker for the region.

Here we report the physical and geochemical nature of this deposit, and assess the most likely age and provenance of the source eruption. Considerations of general location, along with approximate elevation in the cave in relation to down-cutting rate (the ash in Lagang Cave is 12 m above base level: using the rate of base-level lowering of 0.2 m/ka from Farrant et al., 1995, yields ~60 ka), led us to the hypothesis that the ash layer most likely represented the Younger Toba "super-eruption" at ~74 ka, originating in northern Sumatra almost 1800 km to the west (Smith et al., 2011). Work by Bühring and Sarnthein (2000) indicates that heavy ash fall, identified as Younger Toba Tephra (YTT) from matching of rhyolitic glass shard chemistry, took place across the South China Sea, some 600 km to the NW of Mulu. On the southern Indian subcontinent, the Toba ash averages 15 cm thick, with local deposits exceeding 6 m (Westgate et al., 1998; Petraglia et al., 2007), while parts of Malaysia were buried as deeply as 9 m (Acharyya and Basu, 1993). Song et al. (2000), having found YTT much further to the east than expected, suggest that the tephra may have been dispersed by northeastward winds (as well as the prevailing westward winds). However, we also noted that the largest eruption in recent history, the 1815 eruption of Tambora,

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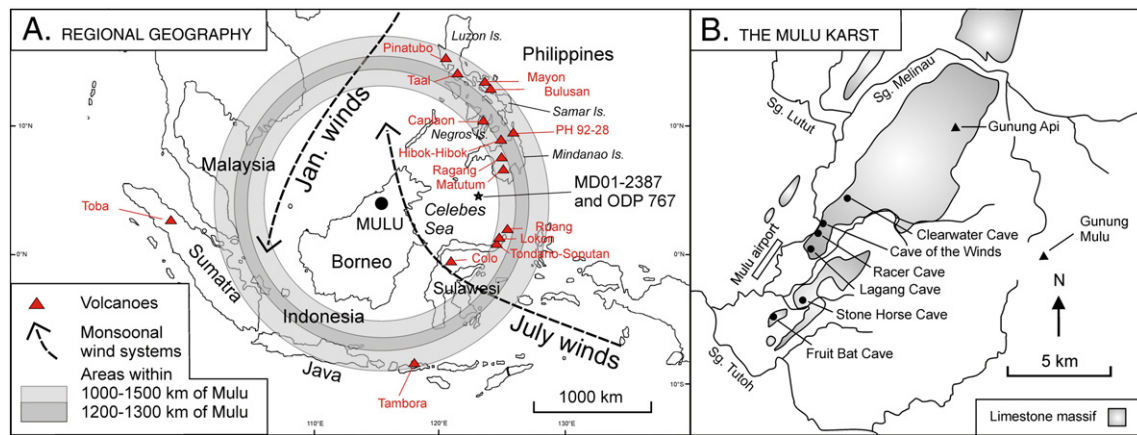
**Figure 1.** A. Geography of SE Asia, and region enclosed by 1200–1300 km (dark grey shading) and 1000–1500 km (light grey shading) distance from Mulu (see text). Dominant monsoon wind systems are shown with dashed-line arrows. Major volcanoes of potential relevance are shown with red triangles. The location of the ocean core MD01-2387/ODP 767 (Ku et al., 2009a) is shown with the star. B. The Mulu karst and the caves studied.

lying at approximately the same distance from Mulu as Toba, placed only a few centimeters of ash on extreme southwest tip of Borneo (Oppenheimer, 2003) and that deposition fell short of Mulu by some 800 km. Therefore, we could not rule out possible sources from the Philippines or elsewhere in Indonesia.

## Background

Gunung Mulu National Park is dominated by 2376-m high Gunung Mulu to the east. The Melinau Karst forms a series of three massifs trending NNE–SSW with steep relief rising from the Melinau floodplain at ~30 m asl to 1700 m at Gunung Api. The three primary geological formations include the basal Setap Shale of Miocene age; the overlying ~2100 m of very pure, massively bedded Melinau Limestone Formation of Lower Miocene to Upper Eocene age (Wannier, 2009); and the upper, slightly metamorphosed, Paleocene and Eocene shales and sandstones of the Mulu Formation (Gillieson, 2005; Hutchison, 2005; Gillieson and Clark, 2010). The highest summits in the Park, around Gunung Mulu to the east of the caves, act as catchment for drainage and thus the source of fluvial sediments in the caves, as well as the presumed original site of ash air-fall deposition.

The climate is governed by the Indo-Australian monsoon system, with the NE monsoon from December to March, the SW monsoon from May to October, and variable winds in the transition periods. Precipitation is ~5000 mm/yr in the lowlands, rising to 6000 mm/yr at 1500 m but falling off at higher altitudes to 4330 mm/yr at the summit of Gunung Mulu. Mean annual temperature is approximately 27°C in the lowlands (Proctor et al., 1983).

The basic sedimentary sequence was first reported from the Clearwater Cave complex (Bull and Laverty, 1981). The basal exposed sediments are gravels with poorly sorted sandy matrix, at least 15 m thick. The basal gravels are capped by as much as 5 m of silt, termed the "Cricket Muds," which in turn are locally capped by a thin flowstone floor and speleothem, many still active. In Clearwater Cave itself, the Cricket Muds are bisected by a white layer of partially decomposed volcanic ash (Laverty, 1982). The ages of the sediments are not well constrained, but the presence of upper-level paleomagnetically reversed sediments (Farrant et al., 1995) suggests that the main body of sediments is younger than 780 ka.

This distinctive white layer appears to be rather widespread regionally. A gray literature report (Laverty, 1983) noted the presence of a presumed-ash layer in the Penrissen karst of southern Sarawak. Harrisson (1961) also noted a presumed-ash layer in archaeological excavations at Niah, northern Sarawak. None of these has been further studied, nor confirmed as tephra.

The ash deposit in the caves is obviously not a primary air-fall deposit but rather fluvially reworked from the original surface deposit. Nonetheless the limited weathering and high purity (at least of the main deposit in Cave of the Winds — see below) suggests that it was fresh at the time of deposition and that the time of deposition in the cave was quite soon after the eruption.

## Methods

In the field we mapped the distribution and elevation of the white layer; sampled from a variety of locations; and searched for dateable speleothem in association with the layer. In the lab we identified the nature of the material through Scanning Electron Microscopy (SEM) and Energy Dispersive Spectral (EDS) analysis, X-ray Diffraction (XRD), Electron Probe Micro-analysis (EPMA) on polished thin section, and Inductively-coupled Plasma Mass Spectrometry (ICPMS). U, Th and Sr isotopic analyses were done by Thermal Ionization Mass Spectrometry (TIMS). The initial focus was on the comparison of this tephra with the Younger Toba Tephra; in addition to a literature search, we obtained a sample of YTT from Michael Petraglia, Research Lab for Archaeology and the History of Art (RLAHA), Oxford, for direct comparison. U–Th dating of speleothem samples was done by TIMS. Technical details are documented in on-line supplementary materials.

To minimize the inclusion of non-shard material in chemical analyses the material was washed in ultrapure water and filtered through acid-cleaned nylon meshes. For subsequent analyses, we did not use the coarsest fraction (>~63 μm) or the finest fraction (<~20 μm).

## Results

The fine grain size precluded differentiation of the material in the field with a binocular microscope, and post-field studies with a light microscope were of limited value. The tephra shards and the degree of weathering/alteration became clear only with SEM. In the field all the white layers appear to be quite similar; however, SEM results indicate that only some of the beds from the Clay Hall site (Cave of the Winds) are close-to-pure tephra.

### Sedimentary sequence

The deposits of relevance to this report are those in Lagang Cave (Overhang Site), and in Clay Hall, Cave of the Winds (Fig. 2). The
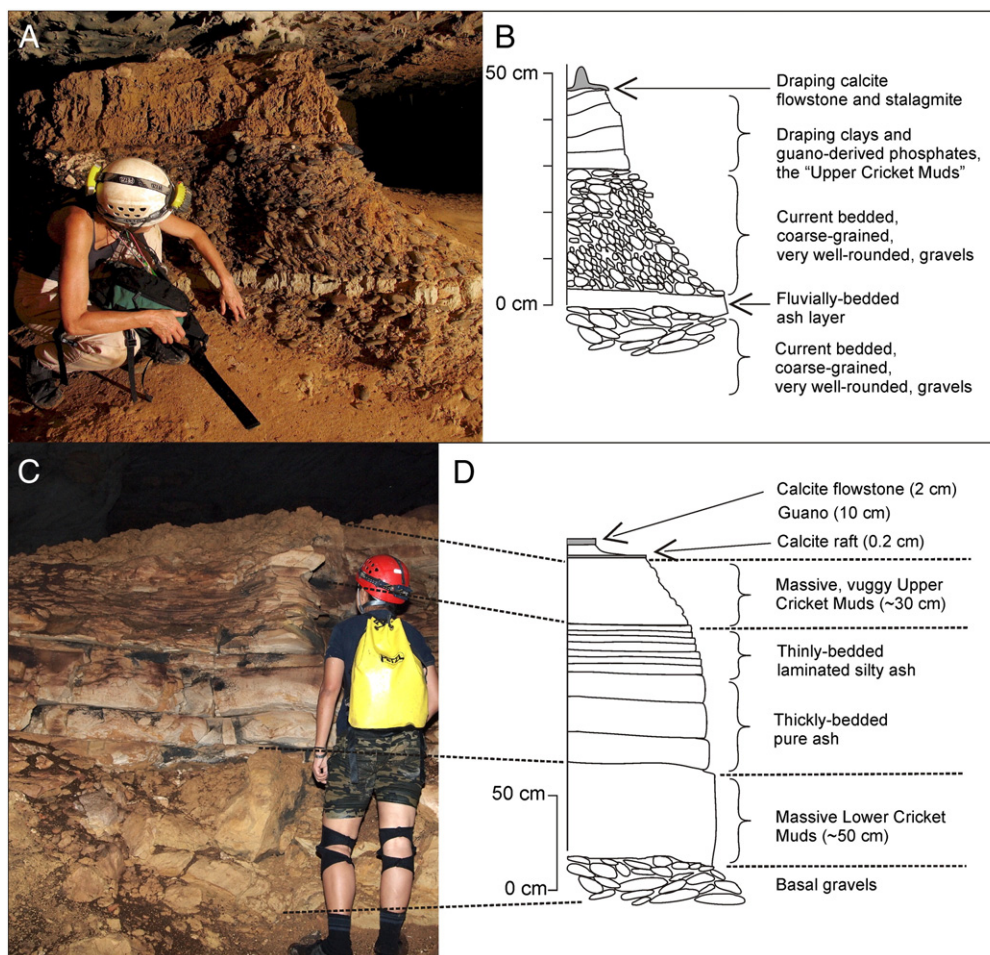
**Figure 2.** A. The thin white ash layer in Lagang Cave, Overhang site, apparent under the left hand of the person. B. Sedimentary sequence diagram. C. The ~2 m-thick ash layer in Clay Hall, Cave of the Winds, overlying basal massive Lower Cricket Muds, and capped by massive, vuggy Upper Cricket Muds. D. Sedimentary sequence diagram.

somewhat weathered ash layer in Lagang Cave is only about 7 cm thick but quite distinct. It is overlain by ~30 cm of coarse gravels, well rounded and current bedded; ~15 cm of horizontally bedded silts/clays of the Upper Cricket Muds (Bull and Laverty, 1981); and capped by a ~1-cm-thick layer of calcite flowstone (Fig. 2A, B). The depositional sequence in Clay Hall is much thicker and more complicated (Fig. 2C, D), consisting of basal current-bedded gravels, ~50 cm of tan-coloured Lower Cricket Muds, ~ 1.5 m of ash (grading upward from more massive pure white ash, from which our sample was taken, to more laminated white-brown silty ash with brown silt partings), ~20 cm of tan-coloured Upper Cricket Muds, capped by 2 mm of calcite raft, ~10 cm of guano, and 2 cm of calcite flowstone.

Since none of the cave sites is open to surface winds, the tephra originally had to be an air-fall deposit on the land surface. The fine-grained sediments, including the ash layer, show fine laminations, small-scale cross-bedding, and fining-up sequences that are typical of fluvial deposition, indicating that the ash was transported from the Gunung Mulu catchment area to the east into the caves. The va-dose river has since downcut by many metres (Farrant, et al., 1995), and flood deposits no longer reach these elevations. The simple interpretation is that the depositional fabric of the ash and associated sediments is indicative of a series of flood-ebb sequences, the Clay Hall sediments signifying largely slack-water conditions. However, the juxta-position of coarse gravels and fine ash-silt in Lagang Cave suggests that the through-flow route of the passage may have been temporarily plugged (presumably by ash). The depositional environment of the ash and other sediments is the subject of a companion paper in preparation.

*Physical structure of the tephra*

The grains are light in colour, varying from clear to pale cream. The physical structure of the shards is shown in Figure 3A, B, and C. They are elongate to equant, the majority having elongate, parallel, tubular vesicles, and only a few having spherical bubbles. In this respect the Mulu tephra differs from the Younger Toba Tephra which has more bubble-wall morphology shards (see also Song et al., 2000), fewer vesicles, and more equant shapes (Fig. 3D). We assessed the shard characteristics from the SEM images by assigning every shard to the categories "rod-like," "platy, or "irregular," and the vesicles to the categories "elongate" or "spherical" (following Ku et al., 2009a). Of the 130 shards measured, 51% were clearly rod-shaped with elongate vesicles, 21% irregular-shaped with elongate vesicles (these look like broken rods), 15% irregular-shaped with spherical vesicles, and 13% irregular-shaped with unclear vesicles.

Many studies of tephra measure the proportion of small pumice shards, pure solid glass fragments, mineral crystals and fragments, and fine lithic fragments (e.g., Sarna-Wojcicki, 2000). We did not do this because the mineral and lithic fragments are more likely from contaminant fluvial material than from the tephra.

Grain size was assessed by wet sieving and sedimentation rate. All the Clay Hall material went through the smallest of our sieves, ~63 μm, indicating that the minimum Feret diameter (Bowen, 2002) must be smaller than this — obviously the elongate shards can be longer than 60 μm as they pass through lengthwise. The median Feret diameter of the ten largest shards (calculated using ImageJ: National Institutes of Health, 2009) from Clay Hall is 63 μm (~4 phi
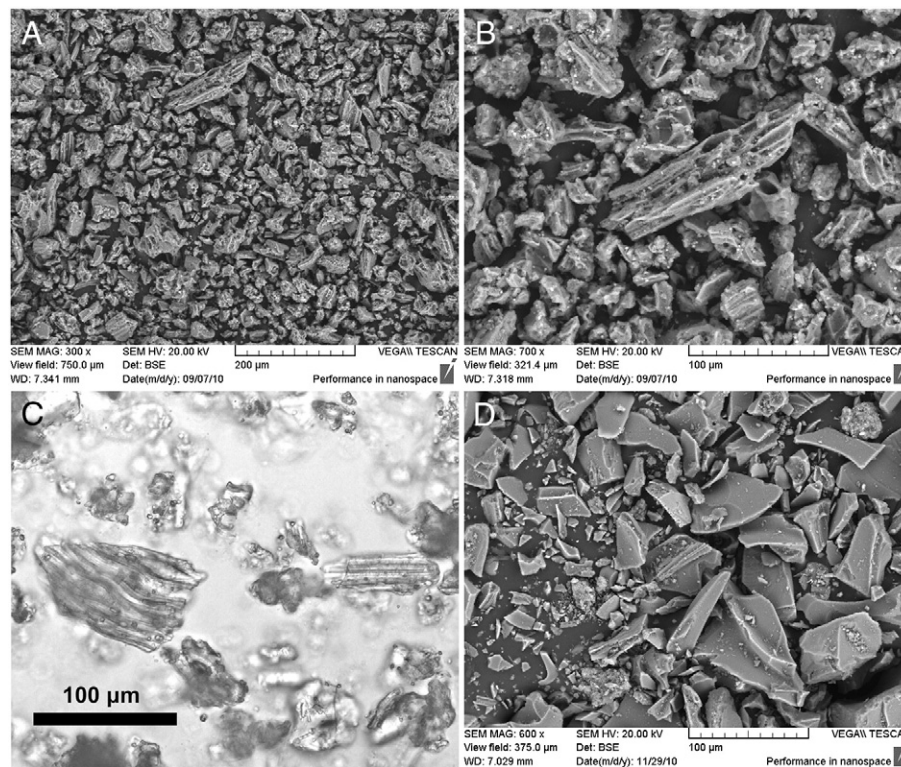
**Figure 3.** A. SEM image of Clay Hall tephra, general view; B SEM image of Clay Hall tephra, detail showing elongate shard and tubular vesicles; C. Light microscope image of Clay Hall tephra; D. SEM image of Younger Toba Tephra.

on the Wentworth scale) and the average of the longest dimension is ~170 μm (~2.5 phi). The rapid sedimentation (fully clear liquid in 5 cm depth within ~3 min) indicates the low content of clay-sized particles, which, along with the negative allophane test result (below), confirms the general impression from the SEM images of a clean, non-weathered tephra.

*Geochemistry of the tephra*

The XRD diffractogram indicated X-ray amorphous material (typical of shards) and quartz (from the minor fluvial silt component).

Because the early reports suggested that the ash was decomposed to varying degrees, we tested for the presence of allophane, following Laverty (1982), with a negative result.

Major species and trace elemental composition from EPMA on eleven shards along with ICPMS on the ~20–63 μm fraction are shown in Table 1 (individual results for EPMA are available in Table S1). The ~75% $SiO_2$ composition indicates that the tephra is rhyolitic in composition (73.0–80.8% $SiO_2$: Ku et al., 2009a). The ICPMS results on the bulk sample are generally consistent with the microprobe results on shards alone, except for the obvious shift in Al values. The pure shards are 13.0% $Al_2O_3$ (typical of pure rhyolite), the filtered bulk samples are 10.4%, and one unfiltered bulk sample (done to test the effect of filtering) only 7.8%. Table 1 also includes analyses done on white silt collected as a sample of weathered clay with a large component of fluvial silt. It is clear that, although well-washed and filtered, the Clay Hall bulk tephra material does have a small component of fluvial silt.

**Table 1**
Geochemistry of the Mulu tephra, columns A and B: EPMA (electron probe micro-analysis) results and ICPMS (inductively-coupled plasma mass spectrometry) results (wt.% average normalized to 100% and 1σ error), and Sr isotopic ratio (average and 1σ error). Column C shows ICPMS on ash with high silt fraction.

| Species | A | | B | | C |
|---|---|---|---|---|---|
| | EPMA on Clay Hall shards (wt.%, 1σ error, n = 11) | | ICPMS on Clay Hall tephra (wt.%, 1σ error, n = 4) | | ICPMS on white silt (wt.%) |
| | Normalized | Measured | Normalized | Measured | Measured |
| $SiO_2$ | 78.7 ± 0.8 | 74.4 ± 1.9 | 79.93 ± 0.22 | 75.64 ± 0.09 | 87.7 |
| $Al_2O_3$ | 13.8 ± 0.7 | 13.0 ± 0.6 | 11.05 ± 0.49 | 10.40 ± 0.41 | 4.11 |
| FeO | 1.1 ± 0.1 | 1.0 ± 0.1 | 1.74 ± 0.06 | 1.64 ± 0.05 | 4.28 |
| $Fe_2O_3$ | 1.22 ± 0.1 | 1.2 ± 0.1 | 1.94 ± 0.07 | 1.83 ± 0.06 | |
| MnO | 0.08 ± 0.04 | 0.07 ± 0.03 | 0.027 ± 0.004 | 0.025 ± 0.003 | 0.003 |
| MgO | 0.25 ± 0.03 | 0.23 ± 0.03 | 0.21 ± 0.01 | 0.20 ± 0.01 | 0.03 |
| CaO | 1.84 ± 0.14 | 1.74 ± 0.13 | 1.31 ± 0.06 | 1.23 ± 0.05 | 0.09 |
| $Na_2O$ | 1.38 ± 0.57 | 1.31 ± 0.55 | 1.99 ± 0.12 | 1.87 ± 0.10 | 0.02 |
| $K_2O$ | 2.61 ± 0.38 | 2.47 ± 0.35 | 2.49 ± 0.46 | 2.35 ± 0.45 | 0.11 |
| $TiO_2$ | 0.16 ± 0.02 | 0.15 ± 0.02 | 0.24 ± 0.01 | 0.22 ± 0.01 | |
| $Na_2O + K_2O$ | 3.99 ± 0.79 | 3.77 ± 0.65 | 4.48 ± 0.35 | 4.22 ± 0.35 | |
| $Na_2O/K_2O$ | 0.52 ± 0.22 | 0.53 ± 0.23 | 0.83 ± 0.20 | 0.83 ± 0.20 | |
| $^{87}Sr/^{86}Sr$ | | 0.70426 ± 0.00001 | | | |

Trace elemental composition is increasingly being used as a descriptor of tephra (e.g., Westgate et al., 1998; Pearce et al., 2007; Óladóttir et al., 2011). Trace elemental data are given in the Table S2.

For several tephras the $^{87}Sr/^{86}Sr$ ratio is diagnostic (Song et al., 2000). Toba is known to have distinctively high ratios (Whitford, 1975). The values for the Toba Tephras given by Chesner (1998) range from 0.71333 to 0.71521. Five analyses on our Mulu tephra gave a $^{87}Sr/^{86}Sr$ ratio of $0.70426 \pm 0.00002$ ($2\sigma$), clearly not corresponding to the YTT.

### Age of the tephra

Successful direct dating of the YTT by the $^{40}Ar/^{39}Ar$ technique is reported by Chesner et al. (1991). However, in the case of the Mulu tephra, Ar–Ar dating is precluded by the low K content and paucity of sanidine as well as possible non-tephra contaminants. Hence, we focused on U–Th dating of associated calcite deposits (Table 2).

Only a few calcite deposits were found above the tephra and none below it. The one most likely to give the best minimum age on the tephra was the calcite raft deposit (which forms in thin layers on top of a still water body) in Clay Hall some 30 cm above top of the tephra (Fig. 2D). However, the whole deposit is only a couple of millimeters in thickness and made up of easily disaggregated tiny crystals. Individual clean crystals were picked out for dating, but non-repeatable results in three repeat dating attempts, and, in one case, isotopic ratios that plot outside of the dating envelope, confirm that this material was open system and leached. Leaching causes loss of U and usually preferential loss of $^{234}U$. Thus, the sample that yields the youngest age represents the closest to unleached material. The true age has to be $\leq 156 \pm 2$ ka. The sample that plots outside the dating envelope, and is of lower U concentration, was clearly substantially leached. However, the closeness of the other two raft dates suggests that these two samples were not extremely leached and the true date is probably not far off 156 ka.

The other dated sample from Clay Hall, the 2-cm-thick flowstone of non-vuggy, compact laminated calcite some 10 cm above the raft calcite, and separated from it by bat guano, gave a good basal date of $81 \pm 1$ ka.

The only other suitable speleothem material found in the caves we studied was a 1-cm-thick layer of flowstone on top of the gravels overlying the white layer in Lagang Cave (Fig. 2A). A sample taken from the middle (avoiding the more detrital edges) gave a good date of $125 \pm 4$ ka. In order to use this date as a minimum on the age of the tephra it was important to establish that this white layer is truly the same material as the Clay Hall tephra. Because the non-Clay Hall samples are contaminated with fluvial material to varying degrees, bulk chemical analyses and Sr isotopic analyses cannot be used for comparison. Instead, SEM was used to pick out the shards and EDS on these was compared with EDS on the shards from Clay Hall. Results show that shards from the Lagang material have identical major elemental composition to the Clay Hall shards (significant at $p$ of $1 \times 10^{-7}$). (While not directly relevant to this report, the

same is true of shards picked out from Fruit Bat Cave, the most southerly of the caves we studied, giving us confidence that it is the same tephra deposit in all the caves of the region that we have studied).

In summary, the U–Th dates require that the ash predates the $125 \pm 4$ ka speleothem and probably predates the $156 \pm 2$ ka speleothem. The technique of fine-tuning tephra age by reference to spikes of sulphate in ice cores, differentiated by isotopic composition, is not applicable here because of absence of high resolution data on pre-MIS 5e ice cores. EPICA Dome C core EDC3 does cover the right time range, but Antarctica has many local volcanoes that swamp the sulphate signal (e.g., Narcisi et al., 2005: of the 13 tephra layers found, all were identified as local and South Atlantic sources).

## Discussion

The initial hypothesis, that the Mulu ash is Younger Toba Tephra, was rejected, based on Sr isotopic ratios, U–Th dates, and dissimilar shard morphology. The age and source of the eruption remained in question.

### Estimating size of eruption, distance from source, and likely source regions

Almost all the publications modelling tephra transport use thickness of deposit as an indication of distance from source (e.g., Pyle, 1989; Bonadonna and Houghton, 2005). In the absence of these data, we must rely on maximum grain size and comparison with empirical data from the literature.

The distance travelled from the origin to the point of deposition is a function of shard size, as well as column height and wind speed. Without knowing wind speed and eruption column height it is impossible to calculate distance travelled with any degree of certainty, but some limits can be estimated. Fisher (1964) plotted empirical data on median diameter of clasts against distance travelled for four moderately sized eruptions with VEIs (volcanic explosivity index: Newhall and Self, 1982) close to 4. Comparing the mean length of the ten biggest shards from the Mulu tephra (~170 μm) on Fisher's maximum clast curve would indicate a transport distance of only ~500 km. We know that no large volcanic centres exist within 500 km of Mulu, so we conclude that the eruption must have had a larger VEI than 4.

To better estimate distance from source we compared grain size of Mulu tephra against Younger Toba Tephra. The Younger Toba eruption, with VEI of 8, is the largest known eruption in the Quaternary (Chesner et al., 1991); hence, it is unlikely that the Mulu ash eruption was as large as this. Using the YTT sample from India (3000 km from source), shard sizes were measured in the same way from SEM images, yielding a mean Feret diameter of the ten largest shards of ~73 μm (~3.8 phi), and mean length of the ten largest shards of ~190 μm (~2.4 phi). Our shard particles are a little smaller than those of Toba. We can, thus, conjecture that the Mulu ash travelled

**Table 2**
U–Th isotopic data and ages.

| Sample | Age ± 2σ (ka) | [a]Age ± 2σ (adjusted) | U conc μg/g | $^{230}Th/^{234}U$ | $^{234}U/^{238}U$ | $^{230}Th/^{232}Th$ | $^{234}U/^{238}U$ initial |
|---|---|---|---|---|---|---|---|
| Lagang Overhang Calcite | 130 ± 1 | 125 ± 4 | 0.42 | 0.652 ± 0.002 | 0.729 ± 0.002 | 14 | 0.608 ± 0.001 |
| Clay Hall flowstone | 82.0 ± 0.5 | 81 ± 1 | 1.41 | 0.476 ± 0.001 | 0.524 ± 0.001 | 38 | 0.400 ± 0.001 |
| Clay Hall raft 2 | 156 ± 2 | 156 ± 2 | 1.22 | 0.644 ± 0.001 | 0.559 ± 0.001 | 1283 | 0.314 ± 0.001 |
| Clay Hall raft 1 | 163 ± 3 | 163 ± 3 | 1.19 | 0.655 ± 0.003 | 0.564 ± 0.002 | 1306 | 0.308 ± 0.001 |
| Clay Hall raft 3 | Not dateable | | 1.00 | 0.731 ± 0.002 | 0.556 ± 0.001 | 1065 | |

| | $^{238}U$ conc μg/g | $^{232}Th$ conc μg/g | $^{230}Th/^{234}U$ | $^{230}Th/^{238}U$ | $^{234}U/^{238}U$ | $^{230}Th/^{232}Th$ | $^{238}Th/^{232}Th$ |
|---|---|---|---|---|---|---|---|
| Clay Hall tephra | 2.6 | 6.6 | 0.894 ± 0.056 | 0.821 ± 0.052 | 0.919 ± 0.002 | 0.983 ± 0.065 | 1.197 ± 0.102 |

[a] All ages were adjusted for detrital contamination using the typical silicate activity ratio $^{230}Th/^{232}Th$ of $0.83 \pm 0.42$, derived from $^{232}Th/^{238}U$ activity ratio of $1.21 \pm 0.6$, $^{230}Th/^{238}U$ activity ratio of $1.0 \pm 0.1$, and $^{234}U/^{238}U$ activity ratio of $1.0 \pm 0.1$ (following Cruz et al., 2005).

less than 3000 km, but probably over 1000 km (since the nearest possible source, Colo, in Sulawesi, is ~900 km distant).
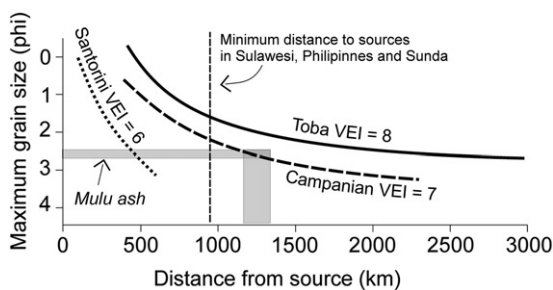
Carey and Sigurdsson (2000) use data from Fisher and Schmincke (1984) to plot variation in maximum grain size versus distance from source for marine tephra fall layers. Figure 4 shows maximum grain size versus distance for Toba (VEI of 8: Rampino and Ambrose, 2000), Campanian (VEI of 7: Pappalardo et al., 2008) and Santorini (VEI of 6: Newhall and Self, 1982). The Mulu ash is shown as the grey line at 2.5 phi and the vertical line shows the closest possible source at 900 km. The intersection of the Mulu ash with the curve for VEI of 6 is the wrong side of the distance cut-off line, so Mulu cannot have had a VEI as low as 6, and it was not as large as Toba. Thus, the Mulu ash most likely corresponds to a VEI of ~7: on this figure it impinges the Campanian curve at a distance of ~1200–1300 km.

Centering this range on Mulu, Figure 1A shows both the narrow grey circle enclosing 1200–1300 km and the wider pale grey circle enclosing 1000–1500 km. This encompasses the major volcanic centres of the Sunda arc (Sumatra and Java), Sulawesi, and the Philippines. Consideration of the prevailing wind systems suggests that the Philippines is the most likely source in winter and Sulawesi in summer (see also similar conclusion for tephras in the Celebes Sea basin in Pubellier et al., 1991). The evidence that, of the two biggest Quaternary eruptions in the region (both on Sunda; Tambora, with VEI of 7, is the next biggest after Toba), neither placed tephra on Mulu suggests that Sunda as a source for the Mulu tephra is highly unlikely. Knowing that the ash cloud from the 1991 Pinatubo eruption missed Mulu by ~600 km (Wiesner et al., 1995), we can assume that the northernmost island of the Philippines, Luzon, although well-known as a center for Quaternary volcanism (Ku et al., 2009b) is probably too far to the north. Further refinement of the most likely source requires comparison of geochemistry of known tephras of the Philippines and Sulawesi.

*Tephrochronology*

Tephrochronology – the matching of unknown tephras to known ones, based on their physical and chemical properties – can be an invaluable tool, but, as Pearce et al. (2007) observe, a single tephra deposit may have geographic variations in the proportions of shards, phenocrysts, lithic clasts, and detrital material. Pearce et al. (2007) overcome such problems by focusing on shards alone, although it may be difficult to separate out the shards in a bulk sample. In the case of the Mulu tephra we have both bulk sample analyses and individual shard analyses.

Tephra matching clearly depends on the availability of similar data on other tephras. Most of the tephrochronological studies published are based on major species (e.g., Liang et al., 2001; Pattan et al., 2010). A limitation is that different eruptions sometimes can have similar major species compositions (Pearce et al., 2007), a problem addressed in some cases by trace elemental comparisons (e.g.,

Bichler et al., 2004). We collected trace elemental data as an additional test but, as yet, we have no trace elemental data on tephras of the region for the right time frame against which to compare the Mulu tephra, so the discussion below is on major species alone.

One of the best repositories of tephras is ocean cores, partly because the tephras are less likely to be altered and mixed with other material than on land, and partly because the foraminifera in the ocean core provide a means of assessing age. Lee et al. (1999) report one tephra from a core to the north of Borneo in the South China Sea, but it proved to be Toba, and the core only goes back to ~150 ka. Pubellier et al. (1991) report tephras from Celebes and Sulu Sea basins, including some likely layers (e.g., up to 95% glass of rhyolitic composition produced by large-magnitude plinian to ultraplinian eruptions); two of these tephras (Pouclet et al., 1991)
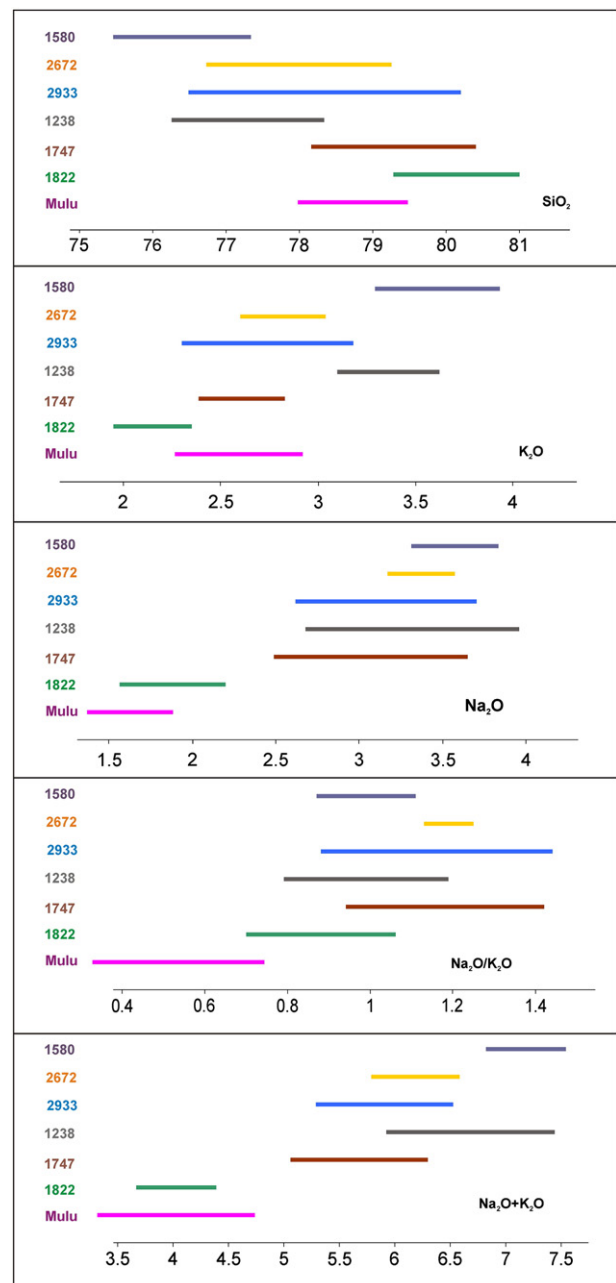


**Figure 5.** The 2 sigma ranges for the six most likely pale-glass tephras (numbered according to depth in cm in core MD01-2387) from Ku et al. (2009a) plotted with the Mulu data.



**Figure 4.** Maximum grain size against distance from source for three large well-studied eruptions, Toba, Campanian, and Santorini (modified from Carey and Sigurdsson, 2000). On this figure the Mulu tephra indicates a most likely distance from source of ~1200–1300 km.

have the right physical characteristics to match the Mulu tephra, but the chemistry does not match.

The best comparison tephras proved to be those documented by Ku et al. (2009a) in ocean core MD01-2387 Celebes Sea basin (marked in Fig. 1A), which is geographically very close to Ocean Drilling Program (ODP) core 767. These authors classify the volcanic provinces into two major groups: Group I includes the Mindanao Island and Philippines and the eastern-most tip of Sulawesi; Group II are further south and include most of Sulawesi. Ku et al. (2009a) were able to estimate ages of the tephra layers quite precisely, based on foraminifera in their core matched against the well-dated ODP core 767.

Separating material into classes according to $^{87}Sr/^{86}Sr$ isotopic ratio and $SiO_2$ (wt.%) immediately eliminates the much more basic materials of Groups I and II of Ku et al. (2009a) and allows us to narrow the most likely source to Mindanao and the other Philippine islands. The group into which our ash falls is Pale-glass-particles, Type A. Of the 15 possible Type A tephras from Ku et al. (2009a), we can eliminate 7 based on age. Our TIMS dates constrain the possible date of the ash to definitely older than $124.5 \pm 3.6$ ka and probably older than $156 \pm 2$ ka. Thus our ash can be tested against any of the pale-glass particles of Type A at depths at least greater than 1200 cm (125 ka) of Ku et al.'s core MD01-2387 and probably greater than 1450 cm.

We can also eliminate the very thin tephra layers (the majority are less than 4 cm thick) on the assumption that the eruption that left thick ash in Mulu was probably substantial, and a VEI of 7 would probably leave a thick layer in the Celebes Sea core (closer to the source than Mulu). Of the nine Type A tephras older than 125 ka, one is of dark glass and thus not relevant (2346 cm). The other eight are potential matches but two are only 4 cm thick and thus less likely to be relevant (1902 and 2913). The remaining six, more likely, layers are: 1238 (5 cm thick, ~130 ka, probably too young); 1580 (5 cm thick, ~165 ka); 1747 (16 cm thick, ~181 ka); 1822 (6 cm thick, ~189 ka); 2672 (11 cm thick, ~274 ka); and 2933 (5.5 cm thick, ~302 ka). These six are plotted in Figures 5–7.

When comparing the geochemical properties several species are of little value because they either show a great range, or both ours and all of the Ku et al. layers plot in the same range. This is true for $Al_2O_3$, MnO, MgO, CaO, $TiO_2$, and FeO. The most useful species include $K_2O$, $Na_2O$, and $SiO_2$. Figure 5 shows the 2-sigma ranges of the useful chemical species for the six Ku et al. layers. The Mulu ash can be seen to match most closely to layer 1822 for $SiO_2$, $Na_2O$, $Na_2O/K_2O$, and $Na_2O + K_2O$. For $K_2O$, it overlaps with layers 1822, 1747, 2933, and 2672. Figure 6 plots $Na_2O + K_2O$ against $SiO_2$ (wt.%): Mulu ash is
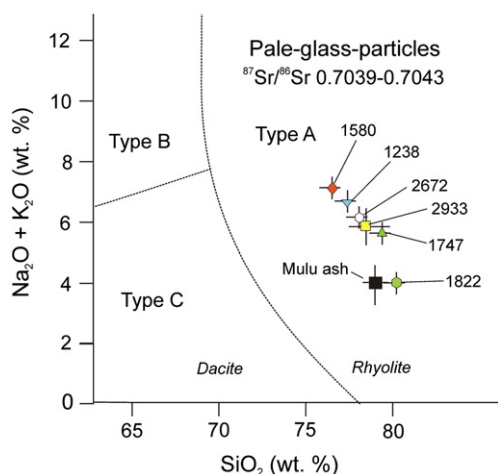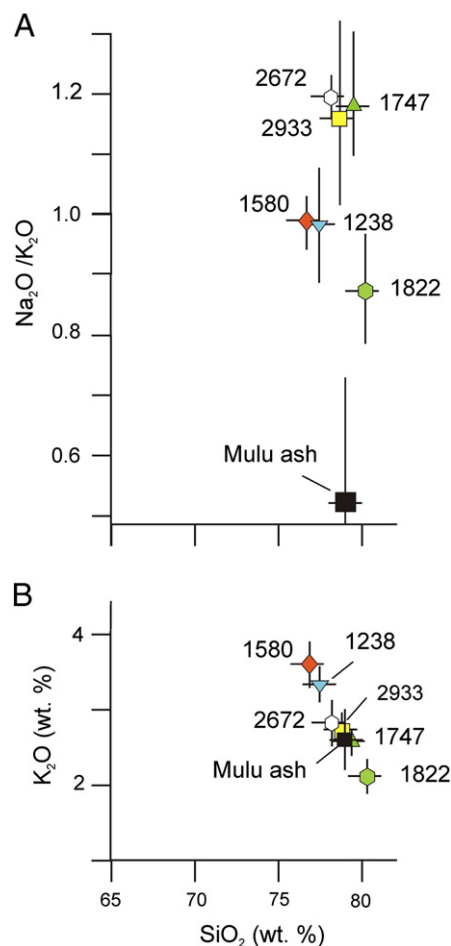


Figure 7. Plots of $Na_2O/K_2O$ and $K_2O$ (wt.%) against $SiO_2$ (wt.%) comparing the Ku et al. (2009a) layers with the Mulu ash.

closest to layer 1822. Figure 7A shows $Na_2O/K_2O$ against $SiO_2$ (wt.%). Here the matches are not so close because the $Na_2O/K_2O$ ratio for Mulu is a larger range and lower than any of the other layers. However, the closest layer is still 1822. For the plot of $K_2O$ (wt.%) against $SiO_2$ (wt.%) (Fig. 7B) all the layers are relatively close and Mulu overlaps with layers 1747, 2933, and 2672.

These comparisons suggest that the Mulu ash is closest geochemically to 1822 (6-cm-thick layer, dated at 189 ka). Layers 1747 (16 cm thick, 181.5 ka), layer 2933 (5.5-cm-thick, 302 ka), and 2672 (11 cm thick, 274 ka) are equally, and much more, distant. Clast and vesicle morphology of Mulu ash matches well with 1822, 2933 and 2672 (rod-like clasts with elongate vesicles) but less well with 1747 (irregular clasts with spherical vesicles). Thus we conclude that, within the limitations of these comparisons, the 189-ka tephra, although not the thickest, is the most likely match.

It may be impossible to further pinpoint the source. According to Ku et al. (2009a) the source is most likely Mindanao Island, but the K–Ar dates of the volcanics on Mindanao reported in Sajona et al. (1994) are not precise enough to allow us to choose a single most likely event. The closest is PH 92–28 at $0.21 \pm 0.10$ Ma (marked on Fig. 1A). However, comparing our $^{87}Sr/^{86}Sr$ data ($0.70426 \pm 0.00001$) to those reported in DuFrane et al. (2006) suggests that the source may be further north on Luzon Island ($^{87}Sr/^{86}Sr$ ratios from the more northerly Bataan arc are 0.7042–0.7046, while those of the Bicol arc, closer to Mindanao, are only 0.7037–0.7039). Countering this argument is the absence of matching between the Mulu tephra and the only rhyolite in their samples, from Pinatubo, northern Luzon Island, and the closer matching of the Mulu Th–U isotopic



Figure 6. Plot of $Na_2O + K_2O$ (wt.%) against $SiO_2$ (wt.%) (EPMA values) showing position of the Mulu tephra in relation to the six most likely pale-glass tephras from Ku et al. (2009a). Mulu ash is shown as the black-filled square.

ratios (Table 2) with the Bicol arc volcanoes than with the Bataan arc ones.

## Conclusion

A distinctive white layer within the sedimentary sequence of many of the caves of Mulu has been shown to be a well-preserved tephra. This represents a surface air-fall deposit of tephra that was fluvially transported and re-deposited inside the caves, where it was sealed by rapid further sedimentary deposition, protecting it from significant weathering. Most of the layers of tephra have been admixed with fluvial material except for Clay Hall, Cave of the Winds, where almost 100% tephra was found. This study has shown that:

- the tephra shards are generally rod-shaped with elongate tubular vesicles. The material is fine-grained, the largest grains being ~170 μm in length;
- geochemically the tephra is pale glass of rhyolitic composition with $^{87}Sr/^{86}Sr$ ratio of $0.70426 \pm 0.00001$;
- U–Th dating of associated calcites suggest that the date of deposition of the tephra is some time before $125 \pm 4$ ka, and probably before $156 \pm 2$ ka;
- grain size and distance from the closest potential source suggests that this eruption probably had a VEI of 7;
- considerations of prevailing winds, grain size, thickness of deposit, location of potential sources, and Sr isotopic ratio limited the potential sources to the Philippines;
- comparisons with the published literature give the best match geochemically with layer 1822 from Ku et al. (2009a), which is dated by stratigraphy in the ocean core to 189 ka;
- this tephra is definitely not the ~74 ka Younger Toba Tephra, formerly considered to be the most likely.

This tephra represents a rare terrestrial repository indicating a very substantial Plinian/Ultra-Plinian eruption that covered the Mulu region of Borneo with ash, a region that rarely receives tephra from even the largest known eruptions in the vicinity. It is unlikely that this tephra layer would be preserved in the weathering conditions external to caves. However, it ought to be present in caves across southeast Asia (an area rich in karst) and would thus provide a definite marker for evaluating denudation and sedimentation rates in Borneo and elsewhere. A concerted effort to locate these ash deposits would provide a much more detailed understanding of the pattern of ash fall throughout the region, and would provide an invaluable chrono-marker horizon for the rich archaeological and paleontological records in these caves.

In light of the probable large-scale ecological impacts of such a substantial eruption (Rampino and Self, 1992; Oppenheimer, 2003; Petraglia et al., 2007), further research is now focused on assessing the isotopic record in speleothems covering this time period.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.yqres.2012.01.007.

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