Supplementary Materials

Methods

To quantify the potential temporal clustering of large volcanic eruptions (>400 km^3 bulk volume) we used the Large Magnitude Explosive Volcanic Eruptions database (LaMEVE)(Crosweller et al., 2012). The LaMEVE database provides the best global compilation of aerial volcanic eruption ages and magnitudes during the Quaternary (Brown et al., 2014; Crosweller et al., 2012). We choose a large volume range cutoff (typically corresponding to magnitude (M) >7 eruptions or Volcanic Explosivity Index (VEI) 7–8 eruptions) since the largest eruptions are most likely to be recorded in the geologic record. This conclusion is further supported by the observation that the cumulative number of eruptions through time (Fig. 4, 28 eruptions total) has an approximately linear relationship in our dataset. Assuming the eruption rate is effectively time-invariant, strong decreases in the eruption recording probability back in time would show up as a convex non-linearity in this plot (Guttorp and Thompson, 1991; Rougier et al., 2018) and this is observed for less well preserved lower volume eruptions (Papale, 2018; Rougier et al., 2018). Nevertheless, there are some gaps in the LaMEVE eruption record (e.g., between 100–275 kyr, 1275–1500 kyr, Fig. 4) which may either be indicative of unrecorded eruptions (more likely though there is no clear relationship with glacial-interglacial periods) or some episodic tectonic process. An analysis of the database biases is beyond the scope of our analysis and we refer the reader to the original LaMEVE papers (Brown et al., 2014; Crosweller et al., 2012), 2014) and Deligne et al. (2017) for a detailed discussion. We choose a lower volume threshold of 400 km^3 to ensure that we have enough eruptions in the dataset to allow robust statistical analysis. Additionally, this threshold ensures that our dataset is very similar to the VEI-8 category dataset in Papale (2018) analysis of recurrence interval for large eruption dataset. We find that the main results of our analysis are not sensitive to the specific volume threshold and are valid as long as we are only considering large (typically a few hundred km^3 eruptions).

We assess any temporal eruption clustering using the coefficient of variation (CV): the ratio of the standard deviation and the mean time interval between two successive volcanic eruptions. The CV (also called: relative standard deviation) is a commonly used statistical measure for analyzing the clustering of discrete events in time (e.g., earthquakes) (Hooker et al., 2018). Typically, CV values are close to 1 for randomly distributed data, >1 for clustered eruptions, and <1 for eruptions with a constant inter-eruption recurrence time (Hooker et al., 2018). Since some volcanic eruptions in the LaMEVE have a significant age uncertainty, we use a Monte-Carlo method to generate 50,000 different possible eruption histories by sampling from the reported eruption ages and their 1σ uncertainties. Using these eruption histories, we calculated the CV value as well as the mean and median recurrence time between large eruptions (inset Fig. 4 and Fig. S2). The median value of the CV distribution is ~1.035, indicating an approximately random distribution. Similarly, the median value of the mean time between eruptions is 76.28 kyr which is close to the value expected for a random distribution (28 eruptions in 2.054 Ma) as well as the results from (Papale, 2018) for VEI 8 eruptions (*ca*. 78 kyr). Finally, although the median value of time between LaMEVE eruptions has a large spread between different possible eruptive histories (Fig. S3), the peak of the probability distribution is *ca*. 35 kyr.

Since our eruption catalog only has a small number of data points (N_{erupt} = 28 eruptions) which can bias statistical interpretations, we generate synthetic random eruption sequences with the same number of eruptions and total sequence duration as our catalog. Using the CV values from these synthetic sequences, we find the LaMEVE distribution lies within the 5–95th percentile values for the random distribution (inset Fig.4; Fig. S2). Thus, on the scale of the whole dataset, the LaMEVE >400 km^3 eruptions do not have any significant non-randomness at the 95%

confidence limit. This conclusion is further supported by the clear overlap between the mean time between eruptions for the synthetic random sequences and the LaMEVE data. However, there is a difference between probability density functions of the median time between eruptions and the synthetic histories. As discussed in more detail later, we interpret this observation to suggest that the LaMEVE dataset likely has a few eruption groups. Since our LaMEVE dataset includes the potential YTT-LCY and Huckleberry Ridge Tuff (HRT)-Cerro Galán Ignimbrite (CGI) pairs, this conclusion is not unexpected.

As a test to illustrate that our statistical analysis is robust and compare CV for clustered and periodic eruption scenarios, we generate 50,000 synthetic eruptive histories with either 2/3/4 clustered eruptions or with periodic eruptions. For the clustered eruption cases, we chose the maximum spacing between individual eruption clusters to be 5% (as well as 40% for the 2-cluster case, this is close to a random case) of the mean time between eruption clusters. Individual eruptive histories are generated by first sampling a random eruptive history with $N_{erupt}/2$; $N_{erupt}/3$; or $N_{erupt}/4$ eruptions and then adding the clustered eruption pairs with random spacing between 1 yr and the maximum spacing (e.g., 5% of the spacing between eruption clusters). For the periodic eruption histories, we set N_{erupt} eruption ages equally spaced over the LaMEVE dataset duration (~2.054 Ma) and assign a 1σ age uncertainty equal to 5% or 30% of the eruption spacing. Then, we generate synthetic histories with the same number of eruptions (N_{erup} = 28) as the LaMEVE dataset. As shown in Supplementary Fig. 2, the CV values for these eruptive histories are distinctive from the LaMEVE dataset with CV >1 for clustered eruptions and CV <1 for periodic eruptions as expected. Additionally, a random eruptive history with ~1000 eruptions has a CV ~1 as theoretically expected (Hooker et al., 2018). Among non-random histories, the closest match with the observed CV values is the 2-cluster case with maximum spacing between individual eruption clusters equal to 40% of the mean inter-cluster temporal spacing. We find the same qualitative result when comparing the median time between eruptions (Fig. S4) where the random and 2-cluster (with 40% variation) is the closest match to the observations. Since the presence of very closely spaced eruption clusters decreases the median time between eruptions (see Fig. S4), we posit that the most parsimonious explanation for the LaMEVE dataset is that it represents a combination of mostly randomly distributed eruptions along with a few closely spaced pairs (e.g., YTT-LCY, HRT-CGI). Given the significant uncertainties in eruption ages for many eruptions in the LaMEVE catalog as well as open questions regarding catalog completeness, it is challenging to presently make any stronger conclusions regarding eruption clustering of large volcanic eruptions.

Finally, we estimate how closely spaced two eruptions can be in a N_{erupt} (=28) random eruptive history. We also assign a bulk eruption volume to every volcanic eruption in each random eruptive history by randomly shuffling the volumes of the eruptions in our LaMEVE dataset. Thus, by construction, the probability density of eruption volumes in each synthetic history is the same as the observed dataset. We would note that herein we have assumed that there is no correlation between eruption volumes and when they erupt. Although this may not be exactly true in practice, this assumption provides a clear statistical end-member to compare against the observations. We use a similar methodology to assign a spatial location for each synthetic eruption by randomly shuffling the locations of our LaMEVE eruptions. Among the 50,000 synthetic histories, we find 2%, 10.15%, 16.392%, and 20% histories with a minimum time between two eruptions being < 80 years, 80–400, 400–1000, and 1000–2000 years, respectively (Fig. S5). However, if we also consider the volume of these eruption pairs, the joint probability of two eruptions spaced between 80 to 400 years and having ≥1000 km^3 volumes are much lower (Fig. 4). Finally, we can consider a constraint that a close-in-time (80-400 yr) supereruption pair must have a small distance (<3000 km) between the antipodal location of the first eruption in the pair and the location of the second eruption. This is motivated by the small similar distance between YTT-LCY (~2200 km). With this additional constraint, the total probability is even lower (Fig. S7).

In conclusion, for a randomly distributed eruption sequence, it is unlikely to observe close eruption pairs like Toba and LCY. As a final note, we acknowledge that our statistical results are weakly dependent on the choice of the underlying statistical model for eruption spacing (Papale, 2018; Rougier et al., 2018; Wang et al., 2020). For instance, a common model for eruption return times is the homogeneous Poisson process with exponential distribution of return times naturally leading to some long-time gaps (Papale, 2018). With this model, they find a very similar result for the mean recurrence time between eruptions (*ca*. 78 kyr) as our results as well as the probability of having a YTT-LCY eruption pair. It is noteworthy that Rougier et al. (2018), also using the LaMEVE database, but only for the last 100 kyr eruptions, find a much shorter (*ca*. 17 ka) recurrence time between M8 eruptions with their statistical model compared to other analysis. They argue for systematic biases in the volume estimates of very large explosive eruptions due to spatially widely distributed deposits for older eruptions. This illustrates that ultimately the accuracy of our conclusions is dependent on the veracity of the geologic constraints for large eruptions especially the accuracy of volume values reported in the LaMEVE database and the confidence in database completeness. To assess how a higher eruption recurrence rate (as argued by Rougier et al. (2018)) may affect our results, we repeated the statistical analysis with only eruptions in the last 100 kyr ($n = 6$ eruptions). Given the smaller number of eruptions, the statistical results for CV are less clear with a larger possible range from synthetic eruption histories. Nevertheless, CV from the 100 kyr LaMEVE dataset is consistent with random eruption distribution as a whole. Additionally, the likelihood of having two eruptions between 80 to 400 years and each having greater than 1000 km^3 volume is still less than 5% (Fig. S6).

Overall, our results are consistent with previous work in showing that presently, there is no strong evidence for eruption clustering for > 400 km^3 Bulk Volume eruptions in the LaMeVE database as a whole. Instead, the dataset is consistent with randomly distributed eruptions within only a few double eruption couplets. This result further highlights that the YTT-LCY eruption doublet is a unique circumstance.

Finally, we plotted published glass shards major and trace-element data for YTT and LCY (Cisneros de León et al., 2021; Pearce et al., 2020) in order to test whether both supereruptions can be easily discriminated if tephra was to be found in ice-core records (Fig. S8).

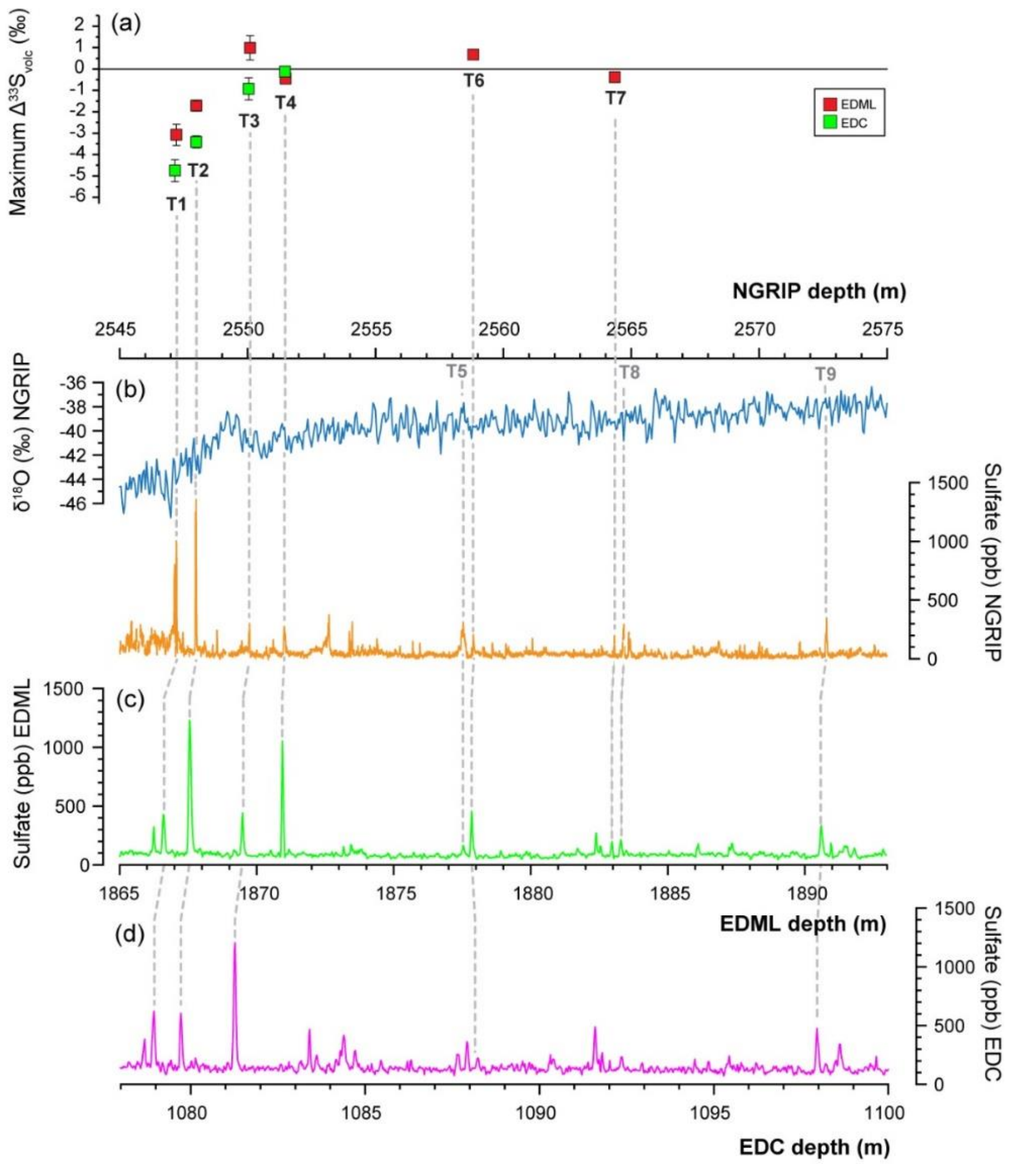

Fig. S1. Correlation of sulfur isotope compositions ($\Delta^{33}S$) from ice core layers containing the sulfate anomalies that are potentially associated with YTT and LCY with records of oxygen (NGRIP) and sulfate from the NGRIP, EPICA Dronning Maud Land (EDML, Antarctica), and

EPICA dome C (EDC, Antarctica) bipolar ice core records. Modified from (Crick et al., 2021) and (Svensson et al., 2013).

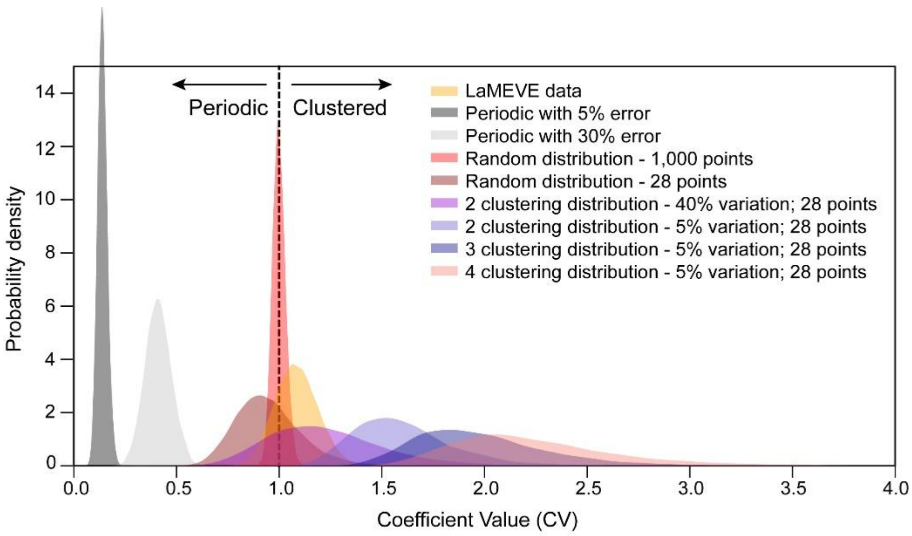

Fig. S2. Coefficient of variation (CV) Monte Carlo synthetic results. Analysis of the Coefficient of Variation for 50,000 synthetic eruptive histories with different statistical models - random, clustered, and periodic. We also plot the results from the LaMEVE dataset for comparison.

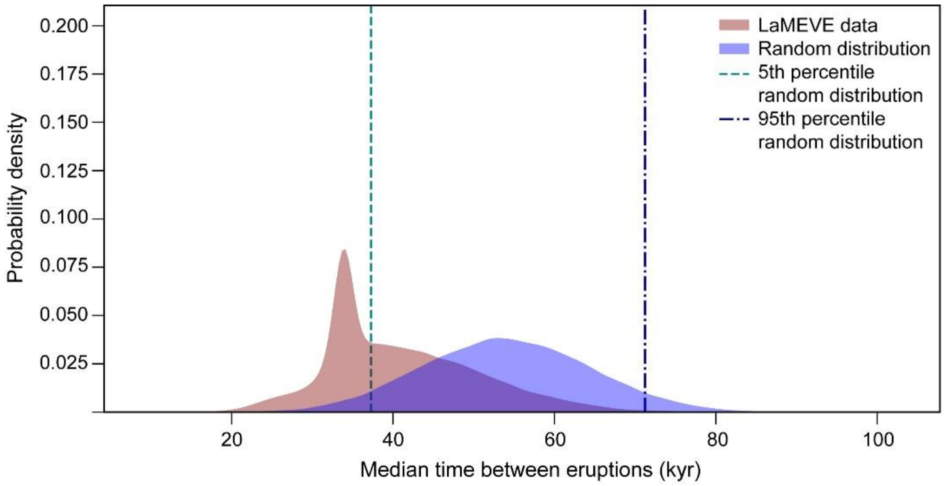

Fig. S3. Median time between eruptions (Monte Carlo results). Analysis of the Median value of the time between individual eruptions for the LaMEVE Dataset and 50,000 synthetic eruptive histories wherein the eruptions (28 eruptions, same as LaMEVE dataset) are randomly distributed in time.

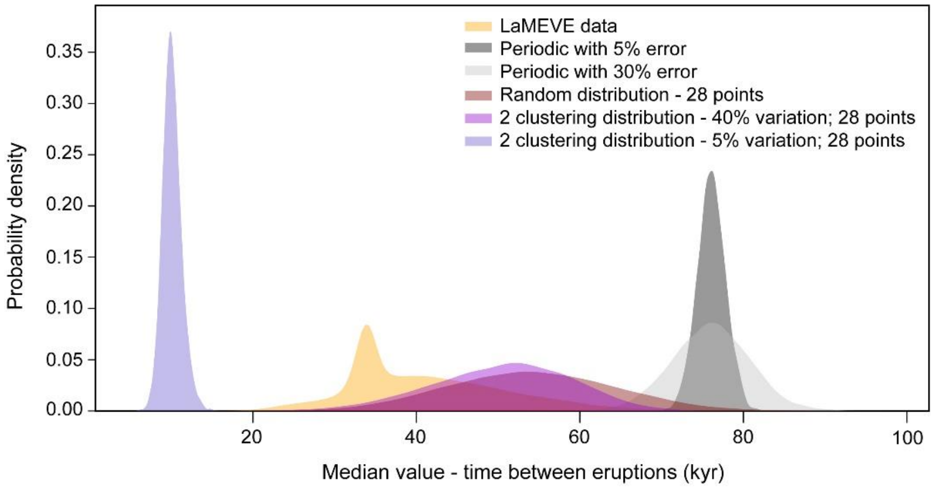

Fig. S4. Median value for time between eruptions. Analysis of the median value of the time between individual eruptions for 50,000 synthetic eruptive histories with different statistical models - random, clustered, and periodic. We also plot the results from the LaMEVE dataset for comparison.

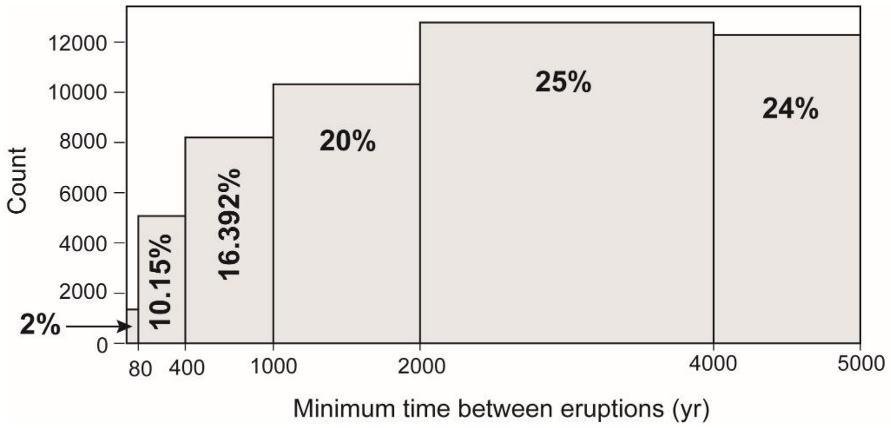

Fig. S5. Histogram of minimum time between subsequent eruptions for 50,000 synthetic eruptions histories assuming that eruptions are randomly distributed. The numbers on each histogram show the percentage probability of being in that bin based on the synthetic eruptive histories. We would note that here we are only considering the time between eruptions and not the volumes of each eruption which provides additional constraints on the likelihood of a large volume YTT-LCY pair.

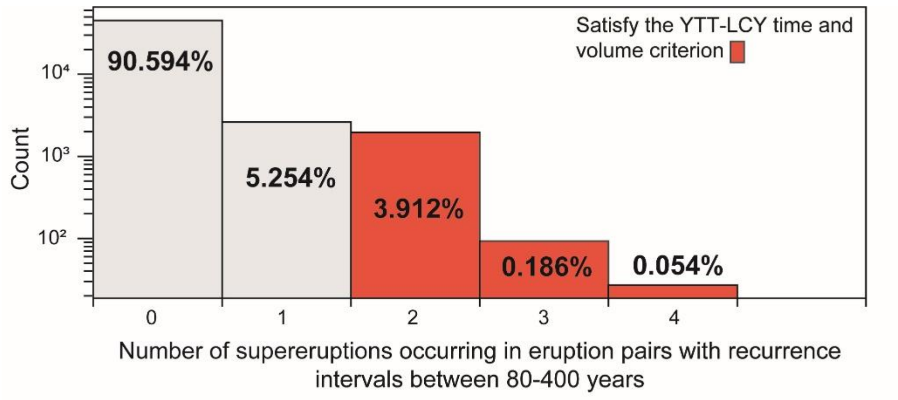

Fig. S6. Histogram showing how many eruption histories (among 50,000 synthetic eruptions histories assuming that eruptions are randomly distributed) have two eruptions within 80-400 years and volumes ≥1000 km^3 (supereruption). In contrast to Fig. 4, we only use the eruption frequency estimates from eruptions over the past 100 kyr. A supereruption pair like YTT-LCY would be represented by the '2' bin. On the other hand, if only one of the two closely spaced eruptions is a supereruption, it would be represented by the '1' bin. The numbers on each histogram show the percentage probability of being in that bin based on the synthetic eruptive histories.

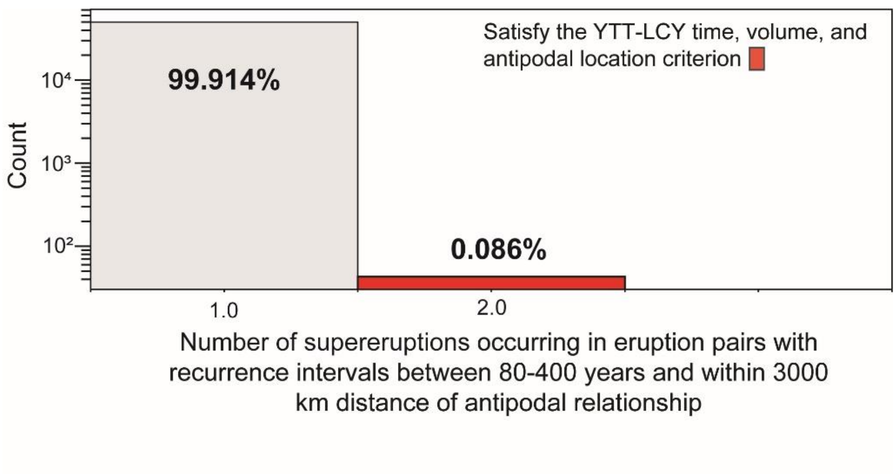

Fig. S7. Histogram showing number of eruption histories (among 50,000 synthetic eruptions histories assuming that eruptions are randomly distributed) that have two supereruptions within 80-400 years and a spatial relationship <3000 km distance between the antipodal location of the first eruption in the eruption pair and the second eruption's location. The numbers on each histogram show the percentage probability of being in that bin based on the synthetic eruptive histories.

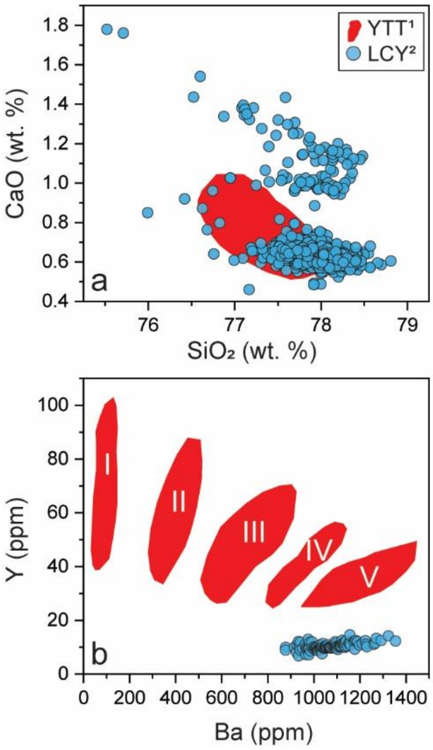

Fig. S8. Major and trace elements compositions for YTT and LCY glass shards. a) Major element compositions and b) Trace element compositions for YTT and LCY glass shards. YTT data from (Pearce et al., 2020) and LCY data from (Cisneros de León et al., 2021).