SUPPLEMENTAL TEXT 1: LUMINESCENCE DATING METHODS

Luminescence dating is based on the principle that charged particles or ions become trapped in defects in crystalline minerals, such as quartz or feldspar, and that these particles accumulate over time (Aitken 1985). These ions are produced by the absorption of environmental radiation, including cosmic radiation, and through radioactive decay of naturally occurring radioisotopes and the release of alpha, beta, and gamma particles. The abundance of the charged particles within a sample is known as the luminescence signal and is, therefore, a function of time. When sufficient energy is applied to a sample, the trapped ions that comprise the luminescence signal are released in the form of light (Feathers 2003:1493). The amount of light released is a function of the time over which the luminescence signal is accumulated; the greater the amount of time, the greater the amount of light that is released. If, in addition to this information, one can measure the amount of radiation in the environment, as well as the propensity for a material to accumulate energy from radiation, one can calculate a date for a sample. This date reflects the amount of time passed since the sample was initially free of charged particles up until it was re-stimulated and the stored energy was released. In the case of ceramics, the starting point of the particle accumulation corresponds to the moment the vessel was fired. While luminescence can accumulate in a variety of semi-conductive materials found in ceramics, quartz and feldspar are common minerals that have properties that result in stable and well-known accumulation of luminescence over time.

For this study, we used optically stimulated luminescence (OSL) dating to estimate the ages of surface collected sherds. OSL is distinct from thermoluminescence (TL) only in that the source used to stimulate samples in order to release accumulated energy is light rather than heat (Aitken 1998). While OSL is generally associated with the dating of sediments, it has an advantage over TL due to the benefits of the single aliquot regenerative (SAR) technique (Murray and Wintle 2000). This allows all necessary measurements to be made on a single aliquot, greatly reducing sources for systematic error inherent in multiple aliquot approaches. Multiple aliquots are used to produce means and standard deviations for the ages.

All of our OSL analyses were completed in the Luminescence Lab at the Institute for Integrated Research in Materials, Environment, and Society (IIRMES) located at California State University, Long Beach. We prepared samples using standard procedures modified from Aitken (1985) and adopted from the University of Washington Luminescence Dating Laboratory (Feathers, personal communication). We processed our ceramic samples and generated dates using aliquots of coarse-grained quartz in the 90–125 μm size range. For comparison, we also generated dates from fine-grained mixed minerals that are in a 1–8 μm size range. The coarse-grained quartz aliquots have the advantage that the single mineral emits reliable and measurable luminescence emissions between 410 and 430 nm (Bøetter-Jensen et al. 2003). Fine-grained mixed mineral samples have advantages in terms of the steps required for processing and the fact that the feldspars that are common in these aliquots tend to produce stronger luminescence signals. Nevertheless, measurements from mixed minerals can also include signals from other less understood minerals. Feldspar grains often experience fading, where the signal is systematically lost over time. The use of both sample types gave us the ability to compare our results and to integrate both data sets to produce a final age estimate.

We made all luminescence measurements using an automated Risø TL/OS 12B/C reader that incorporates calibrated beta (^{90}Sr) and alpha (^{241}Am) radioactive sources. The calibrated beta source exposes a sample to known amounts of radiation. This facilitates calculation of the sample response as well as the equivalent dose—the amount of radiation required for the sample to have the measured amount of luminescence. When combined with measures of the amount of radioactivity present in the environment, the equivalent dose enables one to calculate the age of a sample. Knowing this response rate is central for evaluating the rate of luminescence signal accumulation over time.

We stimulated the samples for luminescence using blue-light OSL (BOSL) stimulation within a range of 400–550 nm (centered at 470 ± 30 nm). This range is well suited to samples with the single aliquot regenerative dose (SAR) protocol outlined by Murray and Wintle (2000). For the fine-grained mixed mineral samples, in order to help eliminate luminescence contribution due to minerals other than quartz and to isolate the quartz emissions, we used an SAR method with an IR-wash that involved exposing the sample to infrared light before making measurements with the blue-light source (Banerjee et al. 2001). For the double-IR wash step, the samples were stimulated using infrared diodes in the 800–900 nm transmission range. A U-340 filter was used to eliminate spillover from stimulation blue light and to restrict the measurement to light being emitted from the sample.

Using the facilities at IIRMES, we measured elemental abundances to estimate the amount of radioactivity that is present in the sample and the surrounding environment. For this analysis, we utilized our GBC OptiMass 8000 ICP Time of Flight (TOF) Mass Spectrometer attached to a New Wave Research UP-213 Laser Ablation system (LA-TOF-ICP-MS). In order to compensate for internal heterogeneity of radioactivity within sherds, we ball-milled a small portion of each sherd until the material consisted of particles of approximately 5 μm in size and was thoroughly mixed. For an internal standard, we dosed each sample with a 40 ppm indium standard. We mixed the final sample with a briquetting additive and pressed the powder into a pellet with a 15-ton geological sample press. Using laser ablation to sample areas across the pellet, we then measured U, Th, and K concentrations in our TOF ICP-MS. In these analyses, we conducted repeated five-second data acquisitions that we then averaged and calculated a standard error. All intensity counts were normalized using an internal standard that we placed in each pellet. We also calibrated the abundance of each element using external calibration standards. In addition to samples of material from ceramic sherds, we also estimated radiation from the surrounding sediment for each location with pellets of sediment made in the same fashion as the sherds. We incorporated all of the data from the luminescence measurements, characterizations of the rate of luminescence signal generation, and the abundance of environmental radiation into a set of calculations that provided us with the number of years since the event of firing when the luminescence signal was assumed to have been reset.

Although we started with coarse-grained quartz for our study, we focused our analyses on fine-grained polymineral samples ranging from 1–8 μm once we learned that we could obtain equivalent results with both techniques. The analysis of fine-grained mixed mineral samples allowed us to avoid a step involving the etching of coarse grains using HF, an unpleasant chemical in laboratory use. Nevertheless, given the small size of the fine-grained samples we had to add a step to estimate the efficiency of alpha radiation. Alpha radiation generally contributes to the luminescence signal by accumulating in traps in a mineral grain for a depth of only the first few microns from the surface. In the case of coarse-grained samples, the contribution due to alpha radiation is eliminated through the etching of grains with HF acid. Fine-grained minerals, on the other hand, are too small to etch since the process would dissolve the grains entirely. Consequently, one must measure alpha efficiency, the measure of luminescence induction during exposure to alpha radiation, a value that is generally lower than exposure to beta and gamma rays (Liritzis et al. 2013:21). The contribution due to alpha radiation is measured using a calibration procedure. In this calibration procedure, two aliquots for each sample for which the equivalent dose is already known are chosen for the alpha correction. These aliquots are given a known amount of alpha radiation and the equivalent dose is then re-measured using the SAR sequence and beta irradiation. Different doses for alpha are applied to each of the aliquots, depending on the original equivalent dose of the aliquot. The equivalent dose is then compared with the given alpha in order to determine the alpha efficiency, a value known as the b-value. The b-value is then used to correct the originally measured paleodose. For pure quartz, b-values range from .4 to .6. This value may vary if the sample contains minerals other than quartz. In a case in which there are feldspars, the b-value may be larger than 0.6. In our samples, we found only a few samples with large b-values. We used the averaged b-values for the two aliquots to correct the remaining aliquots for each sample. For three of the samples (LB0745, 754, 755) we were unable to make alpha measurements due to insufficient materials. Although the dates are reported below, we excluded these samples from the final analysis.

For each ceramic sample, we used the SAR technique to generate a set of equivalent dose measurements, one for each aliquot. Determining the central tendency of this distribution of values, we considered the dispersion of the equivalent dose. Based on their dispersion, we used one of two statistical models to derive an estimate of age. When the distribution of equivalent dose values were the same or very similar for all aliquots with no over-dispersion, we used a "common age" model that averaged the equivalent dose and error. When the equivalent doses were dispersed, as indicated by the high values for the rate of overdispersion, we used the "central age model" (Galbraith and Roberts 2012). The overdispersion rate consists of the ratio of the aliquots that exceed two standard deviations based on the averaged value. In the case of these samples, we found the overdispersion rates to be less than 15 percent, indicating a small amount of dispersion for the equivalent doses. Thus we used the “common age” model for calculating the mean age values for the sets of aliquots.

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