Luminescence Analysis of Ceramics from the Dongshanzui Site

James K. Feathers
Luminescence Dating Laboratory
University of Washington

This report presents the results of luminescence analysis on eight ceramic specimens from the Hongshan period ceremonial center of Dongshanzui in Liaoning Province, northeastern China. All specimens were collected from the surface within a 50 m radius of previous excavations. Laboratory procedures are given in the appendix.

Dose Rate

The dose rate was measured on each ceramic and on associated sediments which were collected within 30 cm of the sherds. The sediments are homogeneous across the site and currently in cultivation. Dose rates were mainly determined using alpha counting and flame photometry. The beta dose rate calculated from these measurements on the ceramics was compared with the beta dose rate measured directly by beta counting. These were in statistical agreement for all samples. Moisture content was estimated as 50±20% of saturated value for the ceramic sherds, reflecting the surface provenience, and 10±5% for the sediments. Table 1 gives the radioactivity data and comparison of the beta dose rate calculated in the two ways mentioned earlier. Table 2 gives all dose rates for each sample. The samples had similar radioactivity, dose rates of around 4 Gy/ka. The lower dose rates of some of the samples is due to low b-value. The radioactivity of the sediments was not too different from that of the samples, suggesting most of the ceramics were made locally.

Table 1. Radionuclide concentrations

Sample	^{238}*U* (*ppm*)	^{233}*Th* (*ppm*)	*K* (*%*)	*Beta dose rate (Gy/ka)*	
				ß-counting	α-counting/flame photometry
UW2617	2.06±0.18	10.01±1.31	2.48±0.12	2.62±0.29	2.56±0.10
Sediment	1.93±0.21	14.79±1.50	2.31±0.11		
UW2618	3.48±0.23	8.05±1.20	2.32±0.11	2.61±0.24	2.59±0.10
Sediment	2.17±0.20	12.24±1.36	2.22±0.19		
UW2619	2.65±0.23	12.95±1.53	1.89±0.09	2.34±0.23	2.26±0.09
Sediment	1.01±0.23	23.14±2.11	2.36±0.08		
UW2620	1.53±0.21	16.84±1.75	2.12±0.08	2.29±0.20	2.38±0.09
Sediment	0.65±0.22	22.93±2.09	1.87±0.06		
UW2621	2.81±0.21	9.12±1.27	2.31±0.08	2.50±0.21	2.51±0.08
sediment	1.54±0.23	19.56±1.92	2.38±0.08		
UW2622	3.29±0.24	10.39±1.37	1.84±0.13	2.32±0.20	2.24±0.11
Sediment	0.45±0.23	24.89±2.19	2.33±0.06		
UW2623	2.07±0.19	10.17±1.32	1.94±0.10	2.17±0.20	2.14±0.09
Sediment	1.56±0.19	14.92±1.49	2.33±0.07		
UW2624	1.87±0.24	19.49±1.92	2.29±0.09	2.40±0.24	2.64±0.10
Sediment	1.67±0.19	13.26±1.50	2.34±0.08		

Table 2. Dose rates (Gy/ka)*

Sample	*alpha*	*beta*	*gamma*	*cosmic*	*total*
UW2617	0.85±0.06	2.34±0.12	0.75±0.05	0.27±0.06	4.20±0.16
UW2618	0.76±0.05	2.32±0.13	0.69±0.05	0.27±0.06	4.04±0.16
UW2619	0.71±0.05	2.08±0.11	0.88±0.07	0.27±0.06	3.94±0.15
UW2620	1.34±0.08	2.19±0.11	0.83±0.06	0.27±0.06	4.62±0.16
UW2621	0.89±0.10	2.23±0.12	0.83±0.06	0.27±0.06	4.22±0.18
UW2622	1.15±0.12	1.99±0.13	0.89±0.07	0.27±0.06	4.30±0.20
UW2623	0.43±0.03	1.99±0.10	0.73±0.05	0.27±0.06	3.41±0.13
UW2624	0.40±0.04	2.40±0.13	0.76±0.05	0.27±0.06	3.82±0.15

*Dose rates for ceramics are calculated for OSL. They will be higher for TL due to higher b-values. Also the beta dose rate is lower than that given in Table 2 due to moisture correction.

Equivalent Dose

Equivalent dose was measured for TL, OSL and IRSL as described in the appendix. TL plateaus (Table 3), were broad except for two samples, UW2621 and UW2622. All the others had plateaus of 100°C or more. Scatter in the growth curves was low in all samples except UW2624. Five of the eight samples showed a sensitivity change with heating. TL anomalous fading was evident in all sherds. Anomalous fading rates, or g-values, ranged up to 16% per decade, and 3 had values higher than 8%. Such values are untypically high, but on only one sample was the g-value so high that a fading correction could not be made. A value higher than about 14% cannot be accurate because if it was the sample would have no signal. Fading rates must therefore have changed through time, which also might be the case for three samples with high g-values, reducing the TL reliability. The TL age was corrected for fading following Huntley and Lamothe (2001).

Table 3. TL parameters

Sample	*Plateau (°C)*	*1st/2nd ratio**	*fit*	*Fading g-value***
UW2617	250-400	1.0	linear	15.9±5.6
UW2618	250-350	1.0	linear	5.2±2.0
UW2619	250-430	1.62±0.22	linear	12.8±4.8
UW2620	250-390	1.53±0.12	linear	7.9±2.7
UW2621	250-270	1.23±0.18	linear	11.0±1.9
UW2622	290-320	1.38±0.04	linear	4.5±1.1
UW2623	260-430	2.28±0.14	linear	4.5±2.7
UW2624	250-400	1.0	linear	6.7±3.7

*Refers to slope ratio between the first and second glow growth curves. A glow refers to luminescence as a function of temperature; a second glow comes after heating to 450°C.

**A g-value is a rate of anomalous fading, measured as percent of signal loss per decade, where a decade is a power of 10.

OSL/IRSL was measured on 4-10 aliquots per sample (Table 4). Scatter was high (more than 10% over-dispersion) on three samples. The IRSL signal on all samples was weaker than the OSL signal, by 2 to 25 times, but it was not negligible. IRSL stems from feldspars, which are prone to anomalous fading. A relatively large IRSL signal may suggest the OSL signal partly stems from feldspars and therefore may fade. The OSL b-value, which is a measure of alpha luminescence efficiency, was higher than is typical of quartz, which usually has a value less than 0.7, for several samples. The IRSL signal and the OSL b–value were large enough on these samples that fading of the OSL signal (which was not measured due to lengthy machine time) cannot be ruled out. As a test of the SAR procedures, a dose recovery test was performed. The recovered dose was nearly the same as the given dose for all samples. Equivalent dose and b-values are given in Table 5.

Table 4. OSL/IRSL data

Sample	# aliquots*		OSL Over-dispersion (%)	Dose Recovery (OSL)	
	OSL	IRSL		Given Dose (sß)	Recovered Dose (sß)
UW2617	6	5	10.2±4.5	200	190±24
UW2618	6	6	11.0±4.0	40	44±2.3
UW2619	7	7	0.7±5.4	150	155±6
UW2620	6	6	0	150	157±6
UW2621	6	6	0	100	108±7
UW2622	9	10	18.8±5.0	200	190±13
UW2623	6	6	0	150	144±5
UW2624	4	4	0	100	94±3.1

*Denotes number of aliquots with measurable signals.
**Over-dispersion after one outlier removed.

Table 5. Equivalent dose and b-value – fine grains

Sample	*Equivalent Dose (Gy)*			*b-value (Gy μm^2)*		
	TL	*IRSL*	*OSL*	*TL*	*IRSL*	*OSL*
UW2617	21.6±1.31	15.5±1.09	20.3±1.09	0.95±0.15	1.09±0.04	1.13±0.04
UW2618	22.2±1.72	16.1±0.69	21.8±1.06	1.11±0.39	1.00±0.03	0.87±0.03
UW2619	26.7±3.59	17.9±0.36	22.0±0.32	3.25±0.41	1.31±0.04	0.72±0.04
UW2620	16.9±1.32	15.6±0.26	21.2±0.32	1.27±0.10	0.99±0.02	1.38±0.04
UW2621	17.5±0.79	13.4±0.61	17.4±0.46	1.42±0.05	1.46±0.11	1.10±0.10
UW2622	20.0±0.57	22.1±0.64	26.0±1.73	1.03±0.04	1.18±0.06	1.29±0.10
UW2623	21.5±1.38	16.5±0.52	20.6±0.28	1.77±0.12	0.97±0.03	0.56±0.01
UW2624	31.8±4.55	10.8±0.26	14.6±0.24	1.28±0.16	0.64±0.03	0.35±0.02

Ages

On four samples the age derived from OSL agreed within 1-sigma of the age derived from TL after correction for anomalous fading: UW2618, UW2620, UW2622 and UW2623. These should be the most reliable dates, but some caveats are in order. First, three of them had relatively high OSL b-values, suggesting some of the OSL signal derived from fading-prone feldspars. Agreement with the fading-corrected TL suggests that OSL fading is minimal, but at least on two of them, UW2618 and UW2620, the agreement was only because the corrected TL had high error terms. It is therefore possible the ages on these two could be slightly underestimated. It should be noted that the IRSL ages on all samples were lower than TL or OSL, which is easily explained by fading. The IRSL results therefore played no role in age determination.

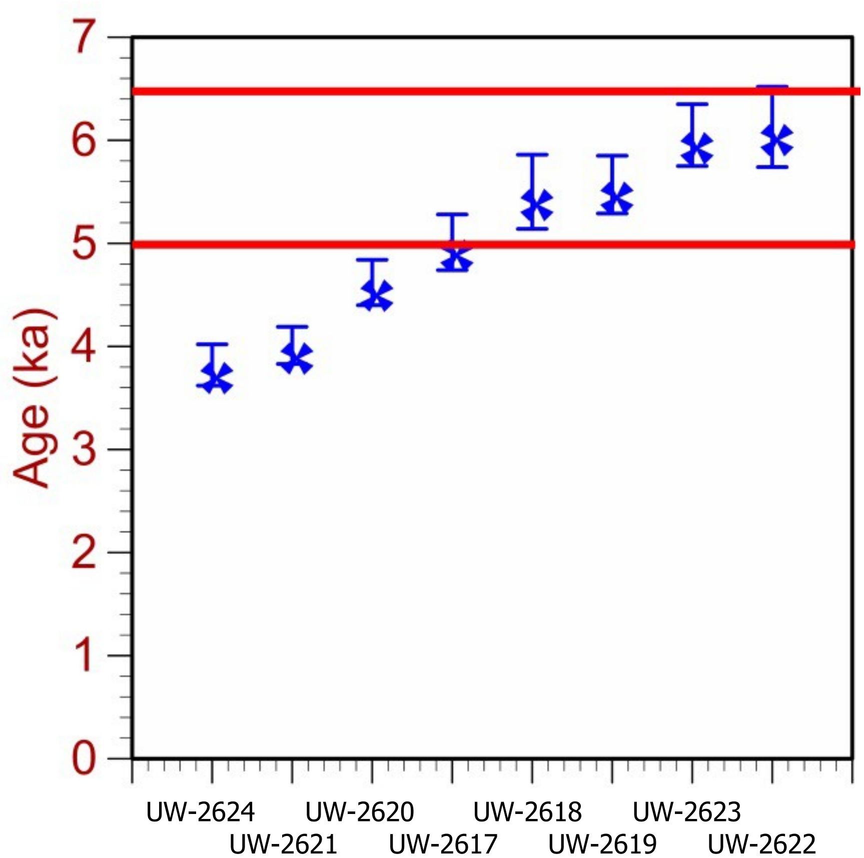
On two samples, UW2617 and UW2621, the age derived from OSL agreed with the uncorrected TL age. On both, the fading rate, g-value, was high, 11 to 16%. In the case of UW2617, no correction was possible. The fading rate must have changed through time for this sample and likely for the other one as well. The OSL b-value for both was relatively high as well, so feldspar contribution to the OSL signal is likely. These two ages are probably underestimated to some degree, but because the TL and OSL ages agreed, despite likely different fading rates, the under-estimation should not be much.

On UW2619, the TL age is younger than the OSL age. The g-value was high, and fading correction on the TL signal produced a Pleistocene age but of such low precision, because the fading data were scattered, that it actually did not differ significantly from the uncorrected age. A better measure might have produced a fading correction that brought the TL age into agreement with OSL. The OSL b-value is also relatively low, suggesting that mainly quartz is contributing to the OSL signal. The OSL age is therefore the best estimate. ON UW2624, the TL age was fully unreliable. The data were scattered, the slide method could not be used because of a high negative intercept, and the fading correction produced a Pleistocene age. OSL provides the best age, and it likely does not fade because the OSL b-value is quite low.

Table 6. Ages

Sample	Age (ka)	% error	Basis for age	Calendar date (years BC)
UW2617	5.01±0.27	5.5	OSL/uncorrected TL	3000±270
UW2618	5.50±0.36	6.6	OSL/corrected TL	3490±364
UW2619	5.57±0.28	5.0	OSL	3560±280
UW2620	4.62±0.22	4.8	OSL/corrected TL	2610±220
UW2621	4.01±0.18	4.4	OSL/uncorrected TL	2000±180
UW2622	6.13±0.39	6.4	OSL/corrected TL	4120±390
UW2623	6.05±0.30	4.9	OSL/corrected TL	4040±300
UW2624	3.82±0.20	5.3	OSL	1800±200

Figure 1 plots the eight samples against time. The red bars indicate the timing of the Hongshan period. The ceramic dates cover the full range of Hongshan and extend to younger ages. Of the two youngest sherds, one might be an underestimate but the other one is not likely underestimated.



Appendix

Procedures for Thermoluminescence Analysis of Pottery

Sample Preparation—Fine Grain

The sherd is broken to expose a fresh profile. Material is drilled from the center of the cross-section, more than 2 mm from either surface, using a tungsten carbide drill tip. The material retrieved is ground gently by an agate mortar and pestle, treated with HCl, and then settled in acetone for 2 and 20 minutes to separate the 1-8 µm fraction. This is settled onto a maximum of 72 stainless steel discs.

Glow-outs

Thermoluminescence is measured by a Daybreak reader using a 9635Q photomultiplier with a Corning 7-59 blue filter, in N_2 atmosphere at 1°C/s to 450°C. A preheat of 240°C with no hold time precedes each measurement. Artificial irradiation is given with a ^{241}Am alpha source and a ^{90}Sr beta source, the latter calibrated against a ^{137}Cs gamma source. Discs are stored at room temperature for at least one week after irradiation before glow out. Data are processed by Daybreak TLApplic software.

Fading test

Several discs are used to test for anomalous fading. The natural luminescence is first measured by heating to 450°C. The discs are then given an equal alpha irradiation and stored at room temperature for varied times: 10 min, 2 hours, 1 day, 1 week and 8 weeks. The irradiations are staggered in time so that all of the second glows are performed on the same day. The second glows are normalized by the natural signal and then compared to determine any loss of signal with time (on a log scale). If the sample shows fading and the signal versus time values can be reasonably fit to a logarithmic function, an attempt is made to correct the age following procedures recommended by Huntley and Lamothe (2001). The fading rate is calculated as the g-value, which is given in percent per decade, where decade represents a power of 10.

Equivalent dose

The equivalent dose is determined by a combination additive dose and regeneration (Aitken 1985). Additive dose involves administering incremental doses to natural material. A growth curve plotting dose against luminescence can be extrapolated to the dose axis to estimate an equivalent dose, but for pottery this estimate is usually inaccurate because of errors in extrapolation due to nonlinearity. Regeneration involves zeroing natural material by heating to 450°C and then rebuilding a growth curve with incremental doses. The problem here is sensitivity change caused by the heating. By constructing both curves, the regeneration curve can be used to define the extrapolated area and can be corrected for sensitivity change by comparing it with the additive dose curve. This works where the shapes of the curves differ only in scale (i.e., the sensitivity change is independent of dose). The curves are combined using the "Australian slide" method in a program developed by David Huntley of Simon Fraser University (Prescott et al. 1993). The equivalent dose is taken as the horizontal distance between the two curves after a scale adjustment for sensitivity change. Where the growth curves are not linear, they are fit to quadratic functions. Dose increments (usually five) are determined so that the maximum additive dose results in a signal about three times that of the natural and the maximum regeneration dose about five times the natural. If the regeneration curve has a significant negative intercept, which is not expected given current understanding, the additive dose intercept is taken as the best, if not fully reliable approximation.

A plateau region is determined by calculating the equivalent dose at temperature increments between 240° and 450°C and determining over which temperature range the values do not differ significantly. This plateau region is compared with a similar one constructed for the b-value (alpha efficiency), and the overlap defines the integrated range for final analysis.

Alpha effectiveness

Alpha efficiency is determined by comparing additive dose curves using alpha and beta irradiations. The slide program is also used in this regard, taking the scale factor (which is the ratio of the two slopes) as the b-value (Aitken 1985).

Radioactivity

Radioactivity is measured by alpha counting in conjunction with atomic emission for ^{40}K. Samples for alpha counting are crushed in a mill to flour consistency, packed into plexiglass containers with ZnS:Ag screens, and sealed for one month before counting. The pairs technique is used to separate the U and Th decay series. For atomic emission measurements, samples are dissolved in HF and other acids and analyzed by a Jenway flame photometer. K concentrations for each sample are determined by bracketing between standards of known concentration. Conversion to ^{40}K is by natural atomic abundance. Radioactivity is also measured, as a check, by beta counting, using a Risø low level beta GM multicounter system. About 0.5 g of crushed sample is placed on each of four plastic sample holders. All are counted for 24 hours. The average is converted to dose rate following Bøtter-Jensen and Mejdahl (1988) and compared with the beta dose rate calculated from the alpha counting and flame photometer results.

Both the sherd and an associated soil sample are measured for radioactivity. Additional soil samples are analyzed where the environment is complex, and gamma contributions determined by gradients (after Aitken 1985: appendix H). Cosmic radiation is determined after Prescott and Hutton (1988). Radioactivity concentrations are translated into dose rates following Adamiec and Aitken (1998).

Moisture Contents

Water absorption values for the sherds are determined by comparing the saturated and dried weights. For temperate climates, moisture in the pottery is taken to be 80±20% of total absorption, unless otherwise indicated by the archaeologist. Again for temperate climates, soil moisture contents are taken from typical moisture retention quantities for different textured soils (Brady 1974:196), unless otherwise measured. For drier climates, moisture values are determined in consultation with the archaeologist.

Procedures for Optically Stimulated or Infrared Stimulated Luminescence of Fine-grained Pottery

Optically stimulated luminescence (OSL) and infrared stimulated luminescence (IRSL) on fine-grain (1-8μm) pottery samples are carried out on single aliquots following procedures adapted from Banerjee et al. (2001) and Roberts and Wintle (2001. Equivalent dose is determined by the single-aliquot regenerative dose (SAR) method (Murray and Wintle 2000).

The SAR method measures the natural signal and the signal from a series of regeneration doses on a single aliquot. The method uses a small test dose to monitor and correct for sensitivity changes brought about by preheating, irradiation or light stimulation. SAR consists of the following steps: 1) preheat, 2) measurement of natural signal (OSL or IRSL), L(1), 3) test dose, 4) cut heat, 5) measurement of test dose signal, T(1), 6) regeneration dose, 7) preheat, 8) measurement of signal from regeneration, L(2), 9) test dose, 10) cut heat, 11) measurement of test dose signal, T(2), 12) repeat of steps 6 through 11 for various regeneration doses. A growth curve is constructed from the L(i)/T(i) ratios and the equivalent dose is found by interpolation of L(1)/T(1). Usually a zero regeneration dose and a repeated regeneration dose are employed to insure the procedure is working properly. For fine-grained ceramics, a preheat of 240°C for 10s, a test dose of 3.1 Gy, and a cut heat of 200°C are currently being used, although these parameters may be modified from sample to sample.

The luminescence, L(i) and T(i), is measured on a Risø TL-DA-15 automated reader by a succession of two stimulations: first 100 s at 60°C of IRSL (880nm diodes), and then 100s at 125°C of OSL (470nm diodes). Detection is through 7.5mm of Hoya U340 (ultra-violet) filters. The two stimulations are used to construct IRSL and OSL growth curves, so that two estimations of equivalent dose are available. Anomalous fading usually involves feldspars and only feldspars are sensitive to IRSL stimulation. The rationale for the IRSL stimulation is to remove most of the feldspar signal, so that the subsequent OSL (post IR blue) signal is free from anomalous fading. However, feldspar is also sensitive to blue light (470nm), and it is possible that IRSL does not remove all the feldspar signal. Some preliminary tests in our laboratory have suggested that the OSL signal does not suffer from fading, but this may be sample specific. The procedure is still undergoing study.

A dose recovery test is performed by first zeroing the sample by exposure to light and then administering a known dose. The SAR protocol is then applied to see if the known dose can be obtained.

Alpha efficiency will surely differ among IRSL, OSL and TL on fine-grained materials. It does differ between coarse-grained feldspar and quartz (Aitken 1985). Research is currently underway in the laboratory to determine how much b-value varies according to stimulation method. Results from several samples from different geographic locations show that OSL b-value is less variable and centers around 0.5. IRSL b-value is more variable and is higher than that for OSL. TL b-value tends to fall between the OSL and IRSL values. We currently are measuring the b-value for IRSL and OSL by giving an alpha dose to aliquots whose luminescence have been drained by exposure to light. An equivalent dose is determined by SAR using beta irradiation, and the beta/alpha equivalent dose ratio is taken as the b-value. A high OSL b-value is indicative that feldspars might be contributing to the signal and thus subject to anomalous fading.

Age and Error Terms

The age and error for both OSL and TL are calculated by a laboratory constructed spreadsheet, based on Aitken (1985). All error terms are reported at 1-sigma. The reference for ka (thousand years before present) is 2010.

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