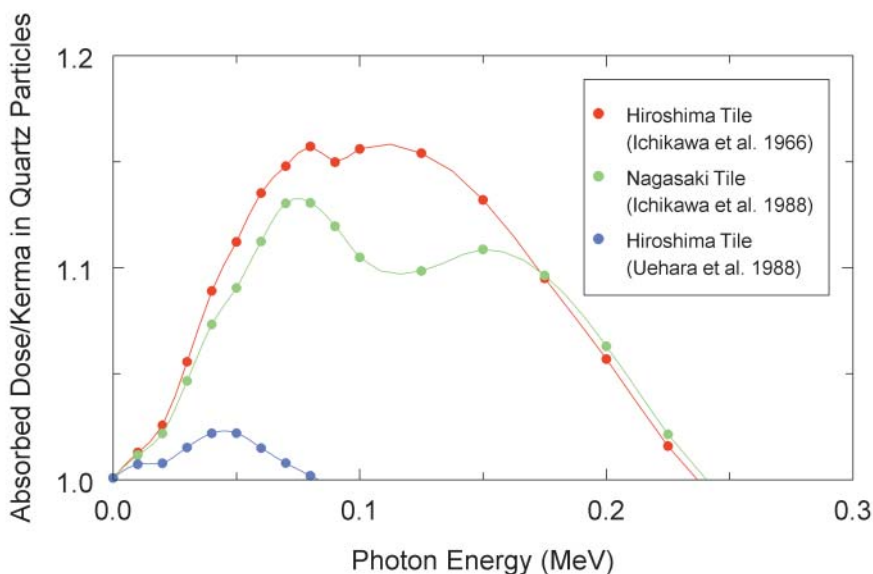


**Figure 6.** Ratios of the mass energy-absorption coefficient for photons in TLD sample matrices of several compositions (Ichikawa et al. 1966; Uehara et al. 1988) to the mass energy-absorption coefficient for photons in quartz ( $\text{SiO}_2$ ). The mass energy-absorption coefficients ( $\mu_{\text{en}}/\rho$ ) used in these calculations were taken from a report by Hubbell and Seltzer (2001).

**Table 8. Dimensions of quartz particles used in TLD measurements**

Investigators	Particle sizes ( $\mu\text{m}$ )	DS86 Final Report reference
Maruyama, Kumamoto, and Noda	105-210	Vol. 2, Appendix 1 to Chapter 4, pp. 113-124.
Ichikawa, Nagatomo, Hoshi, and Kondo	50-74 74-150	Vol.2, Appendix 2 to Chapter 4, pp. 125-136
Ichikawa, Nagatomo, and Hoshi	74-149	Vol. 2, Appendix 3 to Chapter 4, pp. 137-144.
Hoshi, Ichikawa, and Nagatomo	74-149	Vol. 2, Appendix 5 to Chapter 4, pp. 149-152
Haskell, Kaipa, and Wrenn	150-250	Vol. 2, Appendix 6 to Chapter 4, pp.153-171
Baliff	90-150	Vol. 2, Appendix 7 to Chapter 4, pp. 172-183.
Huxtable	90-150	Vol. 2, Appendix 8 to Chapter 4, pp. 184-189.
Stoneham	Not specified	Vol. 2, Appendix 9 to Chapter 4, pp. 190-197.

crystal by electrons that were produced in both the quartz crystal and ceramic tile shell about the quartz crystal was determined using the F6 and F8 cell heating and energy distribution tallies, respectively. These two tallies gave absorbed doses that differed by less than a few percent and the average of the two tallies was used as the absorbed dose in a 100-micron quartz crystal for monoenergetic photons of a given energy. The F4 tally was used to obtain a track length estimate of the photon flux in the spherical quartz crystal, which was convoluted with the fluence-to-kerma conversion factors for quartz from Kaul et al. (1987) to obtain an estimate of kerma in a 100-micron quartz crystal for monoenergetic photons of the same specific energy. The ratios of absorbed dose to kerma in 100-micron quartz crystals that were obtained from these MCNP calculations are plotted in Figure 7 as functions of both ceramic tile composition and photon energy. There is a small enhancement in the absorbed dose to quartz per unit kerma to quartz in the photoelectric region below a couple of hundred keV as shown in Figure 7, but it is not significant when averaged over the entire spectrum of gamma rays at either Hiroshima or Nagasaki. The enhancement is, of course, very dependent upon the composition of the ceramic tiles from which the quartz particles were extracted for the TLD measurements, particularly the percent by weight of elements in the TLD sample with an atomic number greater than that of silicon ( $Z > 15$ ). However, the calculations performed for this work do not suggest any significant error in the calculated dose to quartz of Appendix 11 to Chapter 4 of the Final DS86 Report (Kaul et al. 1987).



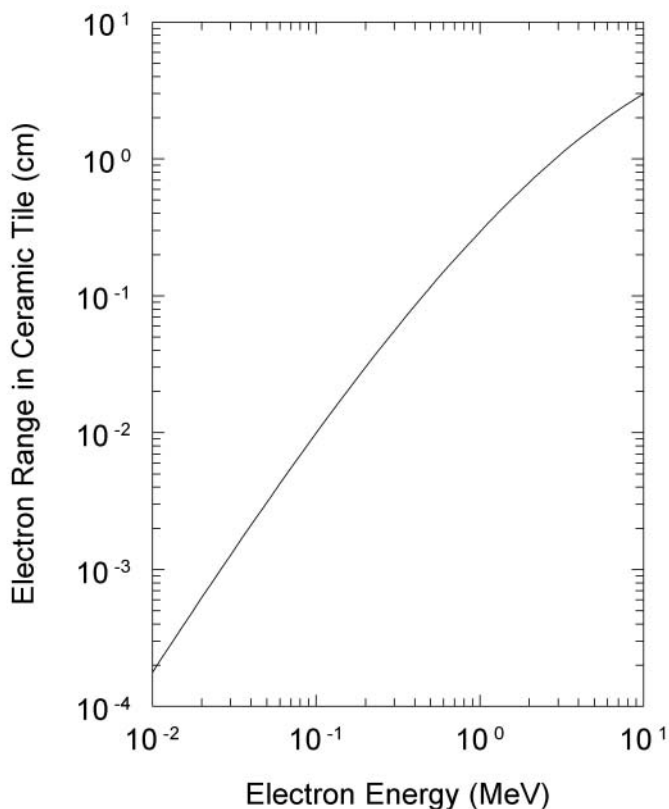
**Figure 7.** Energy response of absorbed dose per unit kerma in 100-micron quartz particles for sample matrices of several compositions (Ichikawa et al. 1966; Uehara et al. 1988). These results were obtained from coupled photon-electron transport calculations with the MCNP computer code (Briesmeister 2000).

Maruyama et al. (1987) state: “All preparation techniques (*for the TLD measurements*) begin by removing several millimeters of material from the outer surface of the potsherd, brick, tile, etc. The purpose of the removal is to improve the uniformity of dose, since, with the limited range of beta particles within a sample, a pronounced drop-off in the (*background*) dose often occurs at the edge of the ceramic. Removing a portion of sample roughly equivalent to the range of particles produced (*by naturally occurring radionuclides*) in the sample (2 to 3 mm) insures that the portion to be used for analysis has been exposed to a beta particle field unaffected by edge effects. Removing several millimeters of outer surface also, in the case of impinging gamma radiation, removes the electron buildup material that would have experienced lower levels of radiation (*dose*) than the remainder of the sample. The removal of this portion is usually accomplished with a water cooled lapidary saw, but surface grinding is common for nonflat and textured surfaces.” The exact amount of surface material removed in the various TLD studies is summarized in Table 9 and varies from as little as 1 mm to as much as 5 mm. Our calculations indicate that there is very little difference in the electron ranges for the various ceramic materials in Table 7, and the removal of as little as 1 mm (0.1 cm) of surface material from the edge of a TLD sample will remove the electron buildup region near the edge of the sample. Figure 8 shows the results of our electron range calculations for the tile composition from the report by Uehara et al. (1988).

Issues pertaining to the free-in-air calculated values associated with measured in situ dose to quartz are treated further in Part B of this chapter, on comparison of calculated and measured values.

**Table 9. Material thickness removed from TLD samples to eliminate contamination and surface layer where electronic equilibrium may not have existed in quartz crystals**

Investigators	Removed material thickness (mm)	DS86 Final Report reference
Maruyama, Kumamoto, and Noda	5	Vol. 2, Appendix 1 to Chapter 4, pp. 113-124.
Ichikawa, Nagatomo, Hoshi, and Kondo	2	Vol. 2, Appendix 2 to Chapter 4, pp. 125-136
Ichikawa, Nagatomo, and Hoshi	2	Vol. 2, Appendix 3 to Chapter 4, pp. 137-144.
Hoshi, Ichikawa, and Nagatomo	1	Vol. 2, Appendix 5 to Chapter 4, pp. 149-152
Haskell, Kaipa, and Wrenn	3-4	Vol. 2, Appendix 6 to Chapter 4, pp. 153-171
Baliff	2-4	Vol. 2, Appendix 7 to Chapter 4, pp. 172-183.
Huxtable	2	Vol. 2, Appendix 8 to Chapter 4, pp. 184-189.
Stoneham	2	Vol. 2, Appendix 9 to Chapter 4, pp. 190-197.



**Figure 8.** Electron range in a ceramic material with a composition and density taken from a report by Uehara *et al.* (1988). The electron ranges were calculated using the ESTAR computer code (Berger *et al.* 2000).

### **Saturation**

The possibility of saturation of the TL response in some samples has been raised, particularly in regard to samples near the hypocenter in Nagasaki, where free-in-air gamma kerma was on the order of 300 Gy. The DS86 report includes a conceptual discussion of saturation in Chapter 4, in Figure 10 and the accompanying text (Maruyama *et al.* 1987). The experience of measurers in Japan, at the Nara University of Education and the National Institute of Radiological Sciences, has been that saturation occurs at levels above 300 Gy for most quartz grains of the coarse size used in the measurements reported here and in earlier studies considered in this work, i.e., grains of diameter at least 74 to 100 microns. This is in contrast to saturation levels sometimes reported for archaeological dating done with finer grains. It is the opinion of the measurers that the methods used were sufficient to avoid any significant inaccuracy due to saturation, even in the early studies of the 1960s.

### ***Annealing by Bomb Thermal Effects***

The possibility of partial annealing of samples due to heat from the bomb or from subsequent fires has been suggested as a theoretical concern since the earliest work (see, for example, Ichikawa et al. 1966) and continues to be a question in some publications to the present day (National Research Council 2001). The work done in the 1960s made allowances for this; for example, roof tiles were taken from the second layer, if those on the top layer showed blistering due to heat (Ichikawa et al. 1966). The DS86 Final Report (Maruyama et al. 1987) states that samples used in that study were tested for partial annealing by the plateau test (high-temperature/quartz inclusion method) or ultraviolet reversal of sensitivity (pre-dose method). Haskell has stated (National Research Council 2001) that the pre-dose method is not affected by the type of annealing that would be associated with bomb thermal effects; furthermore, all of the measurements using the pre-dose technique in the present data are at distances >1,200-m ground distance, beyond the distances where direct bomb thermal effects would be of concern. As documented in the DS86 Final Report, the NRC report, and elsewhere, there appears to be no *a priori* reason to believe that partial annealing is a problem in the present data, although it cannot absolutely be ruled out for the early measurements performed in the 1960s on samples at short distances (Ichikawa et al. 1966; Hashizume et al. 1967a,b).

### ***Calibration Errors***

Various errors of calibration are included in the TL measurement process. They are typically not large errors, but by their multiplicative nature they define a minimum error for any measurement, regardless how strong the signal may be and how small the measurement's relative random error may be as defined by the standard deviation of a set of replicate measurements. A good working definition of "calibration error" as the term is used here is that it is the error between the mean of a set of replicated measurements and the true value of the quantity being measured. One of the immediate implications of this definition is that calibration error so defined includes any errors that are *not* reflected in the natural random variation among replicate measurements on identical samples. That is, for a set of measured values  $x_i$ , the error  $\varepsilon$  among replicate measurements is usually characterized by using the standard deviation of the  $x_i$  (sample standard deviation  $s$ ) as an estimator

$$\hat{\varepsilon} = s = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n-1}} \quad (2)$$

and the calibration error corresponds to  $\bar{x} - \mu$  where  $\bar{x}$  is the mean of the measurements  $x_i$  and  $\mu$  is the true mean of the quantity being measured.

Calibration errors are often thought of as *systematic errors*, because the same calibration value is applied to various measurements. However, an error that is "systematic" at one level of an experiment may be considered as having a random component at some higher level, as discussed further at the end of this section.

For several reasons, measurers tend to focus on the error associated with replicate measurements and not on calibration error.

1. Calibration error is often assumed to be negligible because very accurate standards are available for calibration purposes in many situations; hence the calibrations are assumed to have similar accuracy.
2. Most of the *calibrations* in the methods used here are performed with large enough quantities of the variables involved to have small relative error (e.g., a coefficient of variation  $\sigma/\mu$  of a few percent or less).
3. Natural variation among *replicate measurements*, in contrast, tends to have increasing relative size at lower levels of the measured variable and becomes the error quantity of primary concern as measurements are made at lower and lower levels.

A careful examination of the calibration process reveals that calibration errors of at least a few percent are to be expected in even the most careful experiments. These errors are typically not seen at all in replicate measurements, because the experimental design is such that they are the same for every replicate. That is, for example, the same calibration value relating luminescence to dose might be applied to each of the replicate measurements, and the uncertainty in that calibration value might either not be estimated or its estimate might not be included in calculating the uncertainty of the mean of the replicates.

For example, consider the main measured quantity of interest, the measured gross gamma dose of the aliquots from a given bulk sample being measured for bomb gamma rays, which is denoted “TL” in the treatment beginning on p. 149 of DS86 Final Report, Vol. 1 (Maruyama et al. 1987) and whose standard error is denoted  $\sigma_{TL}$ . For each replicate measurement,  $TL_i$  might be calculated as  $L_iC$ , where  $L$  is an amount of luminescence in arbitrary luminescence units supplied by the TLD reader and  $C$  is a calibration factor in units of Gy per luminescence unit, which is derived from a test irradiation and is the same for all replicates  $L_i$ .  $C$  has some error  $\sigma_C$ , which may or may not be estimated by the measurer. Basic propagation of error suggests (Bevington and Robinson 1992) that, *assuming that there is no correlation between C and L* (see further discussion below), the overall error  $\sigma_{\overline{TL}}$  in  $\overline{TL}$  is given by

$$\overline{TL} = C\overline{L} \Rightarrow \hat{\sigma}_{\overline{TL}} = \sqrt{C^2\hat{\sigma}_L^2 + \overline{L}\hat{\sigma}_C^2}, \text{ where } \hat{\sigma}_L = \frac{\hat{\sigma}_L}{\sqrt{n}} \quad (3)$$

and  $\sigma_L$  is the sample standard deviation of the  $L_i$ , i.e., the same formula shown in equation (2) above with  $x$  replaced by  $L$ . [This product formulation based on first-order partial derivatives is the same one used in equation (4) on p. 149 of the DS86 Final Report, Vol. 1 (Maruyama et al. 1987)]. The term  $\overline{L}\hat{\sigma}_C^2$  is often ignored on the assumption that it is not large enough to be of concern.

The same basic formulation given in equation (3) is considered applicable to all of the measured gross TL doses considered in this work, although those values are arrived at by different methods and calculations whose implications need to be considered, including the pre-dose method as opposed to the high-temperature/quartz inclusion method, and corrections for supralinearity. These and some other physically obvious components of calibration error are discussed in more detail in the following sub-sections. Then an effort is made to characterize the overall size of likely calibration errors in the measurements of interest to the present work, the extent to which those calibration errors may have been included in error estimates provided by measurers, and the overall errors of the measurements.

### ***Test Irradiation Source Output***

The calibration of radiation sources used for test irradiations was identified as a substantial concern in DS86, and was the principal reason for the use of adjustment factors derived from the interlaboratory comparison. However, those factors were within 2% of each other for the two laboratories that continued to make measurements after DS86. There is at present no information to indicate a problem in this area for newer measurements.

### ***Positioning and Geometry of Test Irradiation***

To be consistent with the theory of radiological physics as it applies to dose calibration, the TL material in the test irradiations should be surrounded by a “buildup” thickness of material, of a thickness just sufficient to produce electron equilibrium, but not thick enough to produce any additional attenuation of the incident photon fluence in the irradiation. The thickness, density, and effective atomic number of the buildup layer are thus a factor in correct calibration.

If there is any medium present in the test irradiation setup in locations not between the source and the irradiated material, which would intercept the irradiator beam and produce appreciable dose in the sample by scattering photons from the primary beam, this increment of dose must be considered in the calibration. Such concerns usually arise in regard to “backscatter” from backing materials that are part of the sample assembly, or photons scattered in backward directions from the beam stop or other object that intercepts the beam on the side of the sample assembly that is opposite the source. However, depending on the beam geometry, this consideration applies equally to any source of scattered photons from materials outside the sample and its surrounding buildup layer. (That is, wider beams are more prone to situations in which scatter may arise from “off-axis” directions to the side of the sample assembly.) Any dose due to such scattered photons should be included in the estimated dose delivered by the test irradiation, so that the resulting calibration factor will be an accurate estimate of luminescence per unit deposited dose.

Most sources are typically operated at source-receptor distances on the order of a meter or two in experiments of this type, a distance at which the beam divergence is nontrivial. For a case of a very small source at a source-to-sample distance of 1 m, a 1 cm error in the placement of the source results in approximately a 2% error in the dose, based on inverse-square dose vs. distance due to beam divergence. The effect is slightly less for a source of practical size, although not very much, and the proportional size of the effect is smaller for longer source-to-sample distances. Another related concern regarding exactness of positioning is that the nature of beam divergence means that the dose averaged over the volume of a finite-sized detector or sample is not quite the same as the dose at the center of its volume.

### ***Pre-Dose vs. High Temperature Method***

Various observers have raised concerns that the pre-dose method might have given different values than the high-temperature method. A relatively small (14%) possible discrepancy was suggested by a very small intercomparison study that is documented in the DS86 Final Report, Vol. 2, p. 148 (Nagatomo et al. 1987), but this study was far too small to be reliable, being based on only four pairs of measurements.

The full dataset available for this work includes 83 TLD measurements in Hiroshima at distances greater than 1,200 m ground distance, of which half (42) were made by the pre-dose

method; there are no pre-dose measurements at shorter distances. In order to compare these data, a model (exponential in slant distance) was fitted with a separate intercept and slope for pre-dose measurements, parameterized as an increment in relaxation length times a binary dummy variable and an increment in the fitted dose at 1,400 m slant distance times a binary dummy variable, respectively. In this model, neither the difference in slope nor intercept was close to statistical significance. When a model with a common slope and different intercepts was fitted, the difference in intercepts was not close to statistical significance. This seems to be a reasonable basis for asserting that in the presently available data there is no indication of a difference between the two methods; in fact they are quite similar.

### ***Total Measurement Uncertainty***

Based on the preceding analysis, there appears at present to be no reason to believe that calibration errors or other uncertainties related to experimental method are responsible for unaccounted errors in excess of a few percent. The errors reported by measurers are not less than about 6% and are typically at least 10% (these may be calculated easily from the data in Table 7 of Part B of this chapter). The errors already reported would therefore dominate errors of a few percent in a root-sum-of-squares formulation, e.g., as in equation (2) above. Therefore it does not appear that a detailed quantification of calibration errors or other uncertainties related to experimental method would result in a substantial increase over the errors already reported.

### ***Distance Uncertainty***

The samples used for the analyses in Part B of this chapter were subjected to a detailed map evaluation. Most of the DS86 and newer measurements can be well evaluated with the existing map techniques using the geographical information system, along with aerial photographs, and the errors are reasonably small, i.e., estimated error standard deviations on the order of  $\pm 10$  to 20 m at most in the x and y coordinate of each sample. Measurements made in the 1960s, however, are another matter. Many of these measurements were made at private residences or shrines and temples that can no longer be identified on existing maps; in fact, many such samples were known only to come from a certain “cho,” which is an administrative subdivision of a city that may be several hundreds of meters or more in extent. Several measurements on samples that had clearly conflicting map information have been excluded from the analysis in Part B of this chapter on comparison of measured and calculated values, and the remainder of the measurements in the associated tables of Part B of this chapter that are identified only by “cho” rather than a particular named building should be regarded with caution. Additional discussion of distance uncertainty is given in general terms in Chapter 5 of this report and in particular in Chapter 4.

## **Conclusions**

### ***New Measurements***

New measurements made after 1988 by the JNIRS group and not previously published are reported here and are included in the detailed tables and analyses of this Part (both background

and net measured, bomb-related values). The same measurements are analyzed in Part B of this chapter.

New measurements made after 1987 by the Nara University/Hiroshima University group have been published elsewhere. They are included in the detailed tables and analyses of this section (background only) and are analyzed in Part B of this chapter.

### *Uncertainty Analysis*

**Background.** For most of the ceramic samples that were measured, a period of several decades or more had elapsed between the firing of the pottery and the time of measurement. Various measurers have estimated the total cumulative doses received by samples, from natural background beta rays, terrestrial gamma rays, and cosmic rays, to be in a range between about 100 and 400 mGy. They may fluctuate considerably within this range depending on the environmental conditions around the samples and characteristics of the samples themselves.

There are some indications that newer or more distant measurements might have lower reported background values overall than older or more proximal measurements. This possible trend should be considered in comparisons of measured to calculated values at longer distances and could benefit from further investigation.

The atomic bomb gamma-ray doses at a ground distance more than about 1.5 km from the hypocenters in both cities were approximately on the same order as background, and therefore the measured net doses are strongly affected by the error in estimated background doses.

Therefore, it is impossible to determine precisely the bomb gamma-ray dose with the present TL measurements at these longer distances.

**Energy Response and Calibration Errors.** Energy response may be a problem, if measured values derived from a test irradiation calibrated in terms of dose to tissue or air are used directly as reported estimates of free-in-air dose to tissue or air. Measured values should be converted to dose-to-quartz using appropriate physical constants, and the corresponding free-in-air dose to tissue should be determined by the methods described in this report.

The system used for DS86 and extended for use in this report makes appropriate corrections for energy response-related issues by detailed calculation of dose to quartz, as described in this section and in Part B of this chapter.

Energy response of TLD materials is seen by the analysis above to be only an issue with respect to the gamma-ray attenuation properties of the quartz crystals and the matrix surrounding them in intact samples and the electron disequilibrium at the interface between the two, to the extent that these aspects might not have been adequately treated by the homogeneous sample model used for Monte Carlo calculations of dose to quartz in the samples modeled in DS86. This does not appear to be a significant contributor to random or systematic errors in transmission factors, based on special Monte Carlo calculations performed for this work, as discussed above.

Other calibration errors do not appear to be of sufficient magnitude that they would significantly increase the error estimates already reported by measurers, if they were analyzed in detail and incorporated into those error estimates.

The doses measured by the pre-dose method were compared to those measured by the high-temperature/quartz inclusion method for all reported measurements to date, and no statistically significant difference that would suggest a difference in calibration was found.

**Possible Annealing due to Bomb Thermal Effects.** Annealing of sample materials, whether by direct thermal irradiance or by fires, does not appear to be a problem in the present data, although it cannot be conclusively ruled out for some of the more proximal measurements performed in the 1960s.

**Saturation.** Although the free-in-air kerma near the hypocenter in Nagasaki was on the order of 300 Gy, and some references suggest that saturation occurs at comparable dose levels in some fine-grain quartz used in archaeological dating work, measurers believe that saturation is not a problem in this work, because of the coarse grains and the calibration procedures that were used.

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