

Report of Test

for

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Strontium-90 Activity Calibrations of Brachytherapy Sources

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I. Submitted sources

At the request of Novoste Corporation (Norcross, GA), three batches of sealed sources of ^{90}Sr - ^{90}Y were submitted by AEA Technology QSA GmbH (Braunschweig, Germany) for activity calibrations. The sources were designated, in each case, as "Product code: SICB8184" and "Drawing: VZ-2660" and were further identified as given in Table 1.

Table 1. Submitted sources.

Batch identification by AEA Technology	Number of sources in batch	Reported activity in each source	Reported reference date	NIST assigned identity labels
CO 42795-C2-2A3	8	129.5 MBq	1 August 2001	Y1 through Y8
CO 42795-C2-2A9	8	129.5 MBq	23 August 2001	X1 through X8
CO-44401	3	129.5 MBq	17 October 2001	Z1 through Z3

The first two batches were received on 31 October 2001 and the third batch was received on 30 November 2001. The NIST calibration measurements were initiated on 16 November 2001 and were completed on 4 February 2002. The sixteen sources from the first two batches were returned to AEA Technology on 5 February 2002.

II. Overview of calibration measurements

Calorimetric measurements were performed to obtain the total power from four groupings of the sources; viz., (i) eight sources of Y1 through Y8; (ii) eight sources of X1 through X8; (iii) sixteen sources of Y1 through Y8 and X1 through X8; and (iv) two sources of Z1 and Z2.

These power measurements were converted to total ^{90}Sr activities through the use of a calculated average energy per decay whose value was based on nuclear decay data.

Relative ionization current responses on all 19 seeds, as obtained from two different instruments in standard source geometries, were then used to assign an individual activity to each source from the relative proportion of the total calorimetrically-determined activity.

For confirmatory purposes, source Z3 was radiochemically digested and assayed by liquid scintillation (LS) spectrometry.

III. Assumed nuclear decay data

Relevant nuclear data, as obtained from ENSDF [1], which was used to convert calorimetric power into activity and as used for the LS efficiency-tracing calculations are provided in Tables 2 and 3 below.

Table 2. Nuclear data for the average energy per decay and the power (in units of μW) per unit activity (in units of GBq).

nuclide	average energy per decay (keV)	power per unit activity ($\mu\text{W} \cdot \text{GBq}^{-1}$)	half-life
^{90}Sr	195.80 ± 0.80	31.37 ± 0.13	$10516 \pm 21 \text{ d}$
^{90}Y	933.82 ± 1.22	149.61 ± 0.20	$64.00 \pm 0.21 \text{ h}$
$^{90}\text{Sr}-^{90}\text{Y}$	564.86 ± 0.73	181.02 ± 0.24	--

The activity ratio of ^{90}Y to ^{90}Sr is 1.000254 ± 0.000001 at radioactive equilibrium.

Table 3. Nuclear data used for the LS efficiency-tracing calculations.

nuclide	half-life	maximum β energy (keV)	branching ratio	β transition
^{90}Sr	$10516 \pm 21 \text{ d}$	546.0 ± 1.6	1.0000	1 st forbidden unique $J^\pi = 0^+ \rightarrow 2^-$
^{90}Y	$64.00 \pm 0.21 \text{ h}$	2281.5 ± 2.5	0.999885 ± 0.000014	1 st forbidden unique $J^\pi = 2^- \rightarrow 0^+$
		521.3	0.000115 ± 0.000014	1 st forbidden unique $J^\pi = 2^- \rightarrow 0^+$
^3H	$4500 \pm 8 \text{ d}$	18.619 ± 0.011	1	allowed

IV. Microcalorimetry on batches of the sources

Calorimetric power measurements were made with a commercial dual-cell, near-isothermal (heat flow) calorimeter that was obtained from Calorimetry Sciences Corporation (CSC) of Spanish Fork, Utah and which was denoted as "CSC 4400 Isothermal Microcalorimeter (IMC)." The calorimeter was suitably adapted to utilize specially-designed and -fabricated source-holder cells that are used to maximize the energy absorption of the ionizing radiation and incorporates resistance heaters within these measurement cells to obtain very-accurately-determined independent power calibrations. Refer to Collé and Zimmerman [2] for details on the calorimeter, its operation, and performance evaluations.

Measurements were performed with both thick-walled brass and aluminum source-holder energy absorbers within Hastelloy-C cells. Monte Carlo calculations for the energy deposition in the measurement cells were used to assess and correct for the power loss due to escaping ionizing radiation. The calculated fractions of the total energy absorbed within the cells were 0.99625 for the brass absorber and 0.99680 for the aluminum absorber.

Each calorimetric determination was based on the measurement of the difference between the power obtained with a batch of sources in a sample cell S (as measured against comparable blank materials in a matched reference cell R) and an average baseline power (before and after insertion of the sources and blanks into the cells). Both power measurements used to obtain this power difference were determined at near-isothermal conditions at steady-state heat flow.

The power differences were obtained using an average "canonical" power calibration factor that was determined throughout the course of this work. Twenty-two independent heat-flow power calibrations at applied powers ranging from $60 \mu\text{W}$ to $250 \mu\text{W}$ were performed by applying known power levels to both the S and R cells under both baseline and inserted source conditions.

The results of ten determinations of the calorimetric power and the equivalent ^{90}Sr activity for the various batches of sources are given in Table 4. Uncertainty assessments for both the calorimetric power and equivalent ^{90}Sr activity for each batch are given in Table 5. **Certified values are in bold font.**

Unless specifically noted otherwise, all uncertainties and uncertainty intervals cited within this Report correspond to derived "standard uncertainties" or propagated "combined standard uncertainties." Certified values are reported in terms of "expanded uncertainties" with "coverage factors" of $k = 2$ (see references [3] and [4]).

Table 4. Calorimetric power and equivalent ^{90}Sr activity for the various batches of sources at a Reference Time of 1200 EST 15 December 2001. **Certified values are in bold font.**

source batch	Number of sources in batch	power (μW)	equivalent activity (MBq)	mean power (μW) \dagger	mean activity (MBq) \dagger
all (Xs + Ys)	16	342.70 342.30 344.41	1893.2 1891.0 1902.6	343.14 ± 1.13	1895.6 ± 6.3
all Xs	8	172.45 172.76	952.65 954.37	172.61 ± 0.23	1953.5 ± 1.2
all Ys	8	169.84 170.75	938.25 943.29	170.30 ± 0.65	940.8 ± 3.6
(Z1 + Z2)	2	57.81 58.27 58.09	319.37 321.89 320.93	58.06 ± 0.33	320.7 ± 1.8

\dagger The cited uncertainties correspond to standard deviations of the mean and were derived exclusively from the propagated statistical uncertainty in obtaining the average difference in the two steady-state power levels.

Table 5. Assessment of the uncertainty in the calorimetric power and equivalent ^{90}Sr activity for the various batches of sources as given in Table 4. **Certified values are in bold font.**

uncertainty component	relative standard uncertainty (in %)			
	all (Xs + Ys)	all Xs	all Ys	(Z1 + Z2)
measurement precision in replicate determinations of power differences; propagated standard deviation of the mean (Table 4)	0.33	0.13	0.38	0.56
calorimetric baseline stability; reproducibility on re-insertion of sources	0.2	0.4	0.4	1.2
radiative energy loss from Monte Carlo calculations	0.2	0.2	0.2	0.2
calorimeter calibration factor ($n = 22$ determinations)	0.55	0.55	0.55	0.55
^{90}Sr decay over measurement intervals	0.003	0.003	0.003	0.003
calorimetric power combined standard uncertainty	0.70	0.72	0.81	1.5
calorimetric power expanded uncertainty ($k = 2$)	1.4	1.5	1.6	2.9
average energy per decay	0.14	0.14	0.14	0.14
^{90}Sr activity combined standard uncertainty	0.72	0.76	0.82	1.5
^{90}Sr activity expanded uncertainty ($k = 2$)	1.4	1.5	1.6	2.9

V. Ionization current measurements on the individual sources

Ionization current measurements on all 19 submitted sources were performed with two different instruments: (1) NIST chamber "A", a unique, argon-filled, pressurized (2 MPa) "4 $\pi\gamma$ " re-entrant ionization chamber (hereafter designated as PIC "A"); and (2) a Vinten Instruments Ltd. (Surrey, UK) model 671, nitrogen-filled, pressurized (1 MPa) re-entrant ionization chamber (designated as PIC "V").

Current measurements with PIC "A" were obtained with respect to a NIST, internal-standard, sealed ^{226}Ra reference source, RRS20 and are reported in terms of background- and decay-corrected current ratios $I = I_i / I_R$ at a common reference time of 1200 EST 15 December 2001 (where I_i is the corrected current for source i and where I_R is that for RRS20).

Current measurements with PIC "V" are reported directly in terms of negative-potential currents (in pA) that were corrected for background and radioactive decay to the 1200 EST 15 December 2001 reference time. A check source of ^{226}Ra (nominal 9.3 pA) was used to assure instrument consistency between measurement occasions.

For the ionization current measurements with both instruments, the individual sources were contained within a specially-fabricated, source-holder insert that just fitted within both instruments' standard-ampoule source holder. The insert

consisted of a "Lucite" (methyl methacrylate) right-circular cylinder rod with a 1.5-mm inner diameter and a 7.5-mm wall and whose external dimensions (76-mm length, 16.5-mm outer diameter, and 65-mm well depth) -- and hence source positioning -- matched that for the standard-ampoule source holders.

Average ionization current ratios (for PIC "A") and ionization currents (for PIC "V") were derived from numerous multiple measurements of ionization currents, including evaluations of source positioning variability, on either two or three separate measurement occasions over the course of 20 days. The averaged results for both instruments are given in Table 6, along with estimates of the replicate measurement precision on each.

Table 6. Results of the ionization current measurements on the individual sources at the reference time of 1200 EST 15 December 2001. The results are given in terms of the mean ionization current ratio \bar{I} for PIC "A" and the mean ionization current I (in pA) for PIC "V". The measurement precision on each mean is given in terms of the propagated relative standard deviation of the mean.

source identity	mean \bar{I}	s_m / \bar{I} (in %)	mean I (in pA)	s_m / I (in %)
X1	1.0646	0.11	9.819	0.92
X2	1.0555	0.093	9.718	0.76
X3	1.0007	0.098	9.273	0.78
X4	1.0409	0.072	9.592	0.68
X5	1.0446	0.097	9.631	0.94
X6	1.0472	0.11	9.617	0.82
X7	1.0332	0.11	9.486	0.54
X8	1.0519	0.086	9.671	0.67
Y1	1.0389	0.073	9.523	0.46
Y2	1.0244	0.072	9.381	0.51
Y3	0.9964	0.12	9.144	0.50
Y4	1.0361	0.075	9.530	0.44
Y5	1.0491	0.056	9.640	0.41
Y6	1.0178	0.12	9.325	0.47
Y7	1.0338	0.11	9.477	0.52
Y8	1.0448	0.071	9.606	0.57
Z1	1.4194	0.080	12.94	0.48
Z2	1.4924	0.075	13.57	0.42
Z3	1.4254	0.080	12.99	0.60

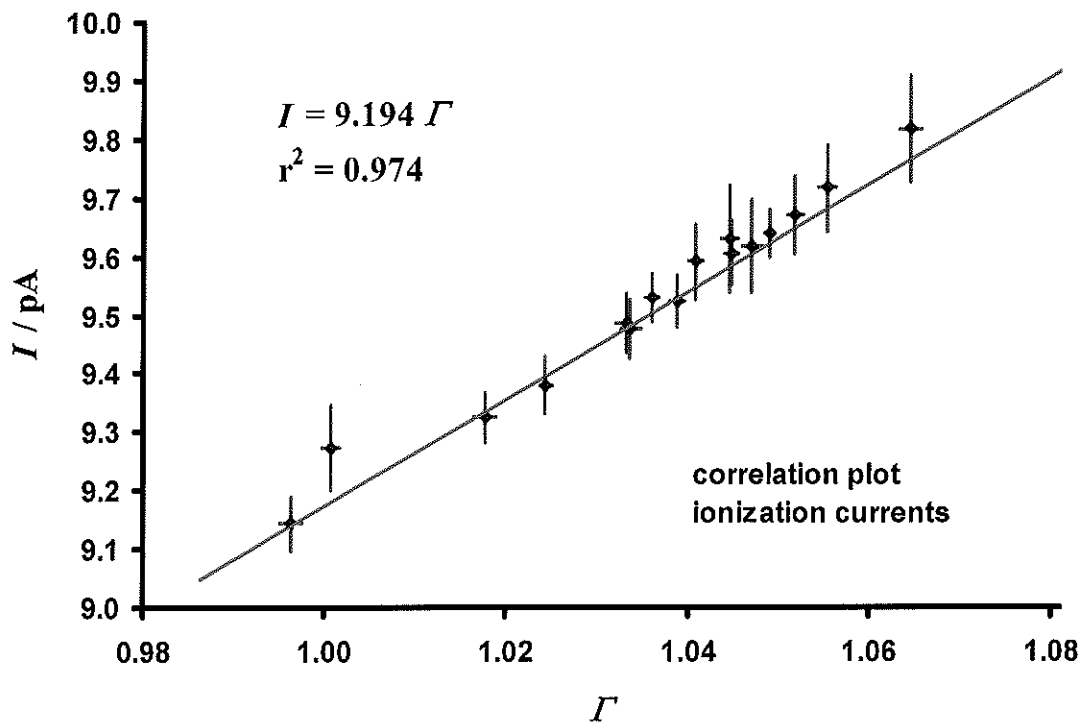
The consistency in the two sets of ionization current measurements amongst the 16 sources in the X and Y source batches is shown in the correlation plot of Figure 1.

VI. Derived calibration factors for the ionization chambers

Calibration factors for both ionization chambers were derived by summing the ionization currents for all of the sources within a batch and combining this result with the calorimetrically-measured equivalent ^{90}Sr activity (Table 4) for that batch. The results, given as $C_V = \sum_i I_i / A$ for PIC "V" and $C_A = \sum_i \bar{I}_i / A$ for PIC "A", are given in Table 7. Table 8 contains the uncertainty assesment for the derived calibration factors.

Table 7. Derived calibration factors for the PIC "V" and PIC "A" ionization chambers at a Reference Time of 1200 EST 15 December 2001.

source batch	$\sum_i I_i$ (pA)	$\sum_i \bar{I}_i$	^{90}Sr activity A (MBq)	$C_V = \sum_i I_i / A$ (pA · GBq ⁻¹)	$C_A = \sum_i \bar{I}_i / A$ (GBq ⁻¹)
all (Xs + Ys)	152.43	16.580	1895.6	80.41	8.747
all Xs	76.805	8.3386	1953.5	80.55	8.745
all Ys	75.624	8.2413	940.8	80.39	8.760
(Z1 + Z2)	26.513	2.9118	320.7	82.69	9.081



page 4 of 8 **Figure 1.**

Correlation of PIC "V" ionization current I with the PIC "A" ionization current ratio Γ for the 16 sources identified as X1 through X8 and Y1 through Y8.

Table 8. Uncertainty assessment for the derived calibration factors given in Table 7.

source batch	relative standard uncertainties (in %)				
	$\Sigma_i I_i$	$\Sigma_i \Gamma_i$	^{90}Sr activity A	$C_V = \Sigma_i I_i / A$	$C_A = \Sigma_i \Gamma_i / A$
all (Xs + Ys)	0.034	0.27	0.72	0.72	0.77
all Xs	0.031	0.17	0.76	0.76	0.78
all Ys	0.023	0.16	0.82	0.82	0.84
(Z1 + Z2)	0.055	0.32	1.5	1.5	1.5

The apparent, nearly 4 % inconsistency between the calibration factors determined with the X and Y sources compared to that for Z1 and Z2 is inexplicable and not understood.

The mean value for C_A obtained from the three determinations with the various X and Y source batches is $8.751 \text{ GBq}^{-1} \pm 0.8 \%$.

VII. Assigned activity values on the individual sources

Table 9 contains the assigned ^{90}Sr activities on the individual sources as determined from the individual mean ionization current ratios Γ (Table 6) from the PIC "A" chamber and its calorimetrically-determined C_A calibration factors (Table 7). The activity values for sources X1 through X8 and Y1 through Y8 were assigned using the mean value of $C_A = 8.751 \text{ GBq}^{-1} \pm 0.8 \%$; whereas that for Z1, Z2 and Z3 was obtained with $C_A = 9.081 \text{ GBq}^{-1} \pm 1.5 \%$.

The relative expanded uncertainties for a coverage factor of $k=2$ on the individual activity values, as given in Table 9, were propagated from the standard uncertainties on the calorimetrically-determined PIC "A" calibration factors (Table 9) and on the decay- and background-corrected PIC "A" relative ionization current measurements. The uncertainty includes

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contributions due to within- and between-measurement-occasion precision, source-positioning variability, long-term temporal variability in the RRS20 relative ionization current measurements, charge collection effects, livetime determinations, background subtractions, and decay corrections.

The certified activity values assume that there are no radionuclidic impurities in the sources that could have significantly contributed to either the calorimetric power measurements or to the ionization currents.

Table 9. Assigned values of the ^{90}Sr activity for the individual sources at a Reference Time of 1200 EST 15 December 2001.
Certified values are in bold font.

source identity	^{90}Sr activity (MBq)	relative expanded ($k = 2$) uncertainty (in %)
X1	121.7	1.6
X2	120.6	
X3	114.4	
X4	119.0	
X5	119.4	
X6	119.7	
X7	118.1	
X8	120.2	
Y1	118.7	
Y2	117.1	
Y3	113.9	
Y4	118.4	
Y5	119.9	
Y6	116.3	
Y7	118.1	
Y8	119.4	
Z1	156.3	3.1
Z2	164.3	
Z3	157.0	

There are no apparent statistically-significant differences in the mean values for the ^{90}Sr activity of the X1 through X8 and Y1 through Y8 batches of sources. The activity of the 16 sources in the two batches can be assumed to be normally distributed (Figure 2) such that they can be considered to have been sampled from a distribution having a mean of about 118 MBq and with a standard deviation of about 2 MBq.

VIII. Confirmatory results from the destructive LS-based assay of source Z3

Source Z3 was wholly dissolved in a small volume of a strong acid solution (nominally 88 % concentrated HCl and 12 % concentrated HNO_3) that was subsequently quantitatively transferred and diluted (serially) to form a master solution having a massic activity of about $6 \text{ kBq} \cdot \text{g}^{-1}$ in approximately $1 \text{ mol} \cdot \text{L}^{-1}$ HCl with carrier ion concentrations of about $50 \mu\text{g} \text{ Sr}^{+2}$ per gram of solution and about $50 \mu\text{g} \text{ Y}^{+3}$ per gram of solution. The total residual activity in the in the source-dissolution vessel and on all solution-transfer tools, as determined by direct LS measurements, was found to be less than 0.01 % of the activity in source Z3. The kind of quantitative procedures and techniques that were employed for this destructive assay have been described in a generalized way previously (see Collé [6] and references therein).

The master solution was assayed for ^{90}Sr - ^{90}Y (in radioactive equilibrium) by an efficiency tracing $4\pi\beta$ LS spectrometric method that is somewhat routine for this laboratory. It is based on well-established procedures for the assay of gravimetrically-determined aliquants of solutions containing β -emitting nuclides (see Collé and Zimmerman [7] and references therein). The assays were performed with two series of LS cocktail samples that were prepared with two different commercially-available scintillation fluids. Each series of the prepared cocktails was measured in all three of the Group's LS spectrometers. The ^{90}Sr and ^{90}Y detection efficiencies were calculated using the CIENIST2000A Code

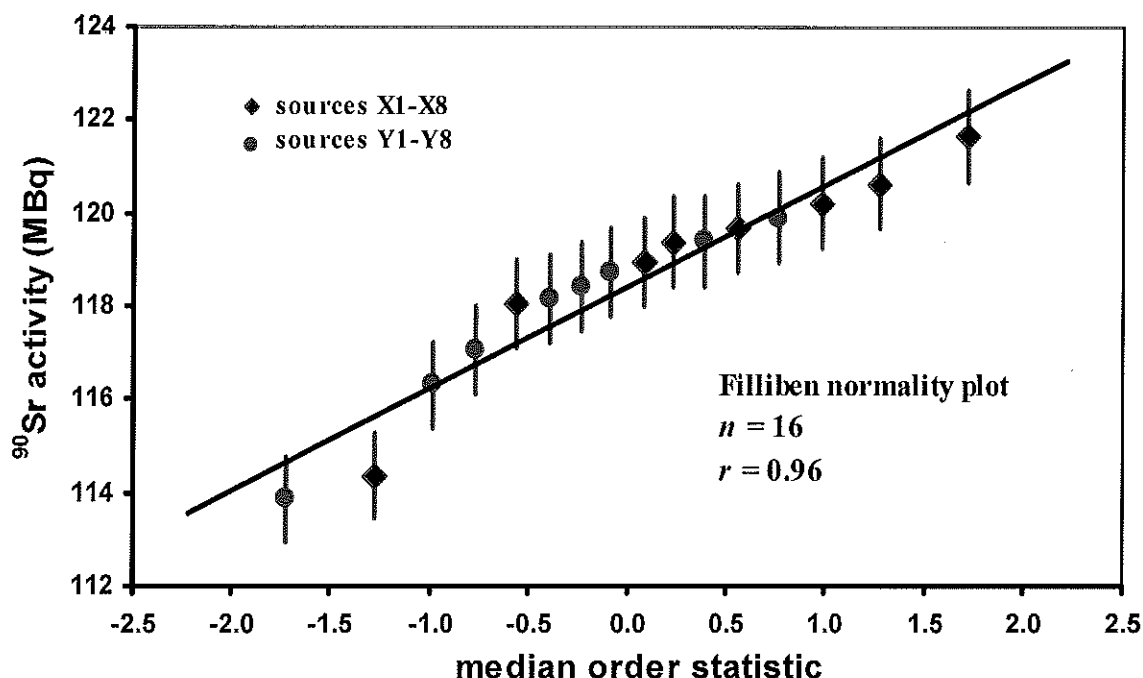


Figure 2.

Normal probability plot for the ^{90}Sr activity in the 16 sources identified as X1 through X8 and Y1 through Y8. The abscissa is the order statistic medians from a normal $N(0,1)$ distribution as given by Filliben [5]. The test statistic r is the normal probability plot correlation coefficient. Based on the percent points p of r for a sample size of $n = 16$, the observed r lies between the $p = 25\%$ and $p = 50\%$ points of the null distribution, and is well above a $p = 5\%$ critical value (where p may be interpreted to be the probability that the values are not non-normally distributed).

(reference [8]) with ^3H as the efficiency monitor. Confirmatory calculations were performed with the EFFY4 code [9]. The calculated efficiency results from the two codes were in agreement to better than 0.05 %. Table 3 summarized the relevant nuclear data that was used as input for the detection efficiency calculations. The ^3H -standard cocktails used for the efficiency tracing were prepared with aliquants of carefully-prepared gravimetric dilutions of a NIST ^3H (tritiated water) standard (reference [10]). Efficiency variations for the tracing were achieved by using varying additions of a 10 % solution of CH_3NO_2 in ethanol as an imposed quenching agent. The ^3H cocktails were in all cases composition matched to the ^{90}Sr - ^{90}Y cocktails by the addition of proportionate amounts of the blank carrier solution that was used to prepare the master solution.

The ^{90}Sr activity in source Z3, as obtained from the LS-based destructive assay, was 155.2 MBq with an relative expanded ($k=2$) uncertainty of $\pm 0.4\%$. The result was found to be invariant of the measurement occasion, of the spectrometer used for the measurement, and of the cocktail series. The uncertainty includes components from the dissolution and gravimetric dillution scheme, gravimetric determinations of LS-sample aliquants, LS measurement precision (for both within- and between-measurement-occasion variability), spectrometer livetime determinations, nuclear decay data and detection efficiency calculations, ^3H standard activity, background subtraction, and decay corrections.

This assay result differs from the calorimetrically-determined value (Table 9) by about -1.1% and is well within the latter's $\pm 3.1\%$ uncertainty for a $k = 2$ interval. This finding confirms the efficacy of the calorimetry to within its measurement uncertainty for this case. For completeness, we note that the calorimetric result cited here was based on use of the $C_A = 9.081 \text{ GBq}^{-1} \pm 1.5\%$ calibration factor (as obtained from Z1 and Z2 measurements). If one instead invokes the average $C_A = 8.751 \text{ GBq}^{-1} \pm 0.8\%$ calibration factor (as obtained from the X and Y source measurements), then the LS assay result will differ by - 5 %. We believe that use of the latter calibration factor is valid for the X and Y sources, but is not valid for the Z sources.

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