Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

Disclaimer:

Gamma-Pulse-Height Evaluation of a USA Savannah River Site Burial Ground Special Configuration Waste Item

R. A. Dewberry, R. A. Sigg, and S. R. Salaymeh
773-41 A
Savannah River National Laboratory
Aiken, SC 29808
USA

Keywords: ISOTOPIC, Microshield, γ-PHA assay, solid waste stream, TRU waste.

Abstract

The Savannah River Site Burial Ground had a container labeled as Box 33 for which they had no reliable solid waste stream designation. The container consisted of an outer box of dimensions 48"x46"x66" and an inner box that contained high density and high radiation dose material. From the outer box Radiation Control measured an extremity dose rate of 22mrem/hr. With the lid removed from the outer box, the maximum dose rate measured from the inner box was 100mrem/hr extremity and 80mrem/hr whole body. From the outer box the material was sufficiently high in density that the Solid Waste Management operators were unable to obtain a Co-60 radiograph of the contents. Solid Waste Management requested that the Analytical Development Section of Savannah River National Laboratory perform a γ-ray assay of the item to evaluate the radioactive content and possibly to designate a solid waste stream. This paper contains the results of three models used to analyze the measured γ-ray data acquired in an unusual configuration.

Introduction

Solid waste is managed at the US Department of Energy Savannah River Site (SRS) to assure that external exposure to waste results in an effective dose equivalent that does not exceed 25 mrem/year to any member of the public.[1] To accomplish compliance with the Department of Energy (DOE) Order of reference 1, solid waste at SRS is designated in one of several categories. These designations are Green-is-Clean (GIC), Low Level Waste (LLW), Transuranic Waste (TRU), Hazardous Waste, and Mixed Waste. The first three of these have waste acceptance criteria that are defined by the level and type of radioactivity. Each of these three excludes various types of wastes that would cause the waste to be designated as hazardous or mixed. Having a transuranic nuclide content greater than 100 nCi/g or a measurable content of lead or RCRA metals would cause the solid waste to be listed in one of the last three categories.

To ensure that low level solid waste does not exceed the environmental release requirements, LLW must also meet the waste acceptance criteria for the SRS E-area vaults. Each segment of solid waste is evaluated for the content of 24 long-lived radioactive species designated as performance assessment radioisotopes (PAR). Since most of the 24 species can not be easily measured, each waste generating facility at SRS was required to designate a solid waste stream that specifies the makeup of each of the 24 PAR's. Each radioisotope must be directly measurable or calculated from an easily measured species or dose rate. It is generally true that all solid waste is characterized by either a total dose measurement that is then distributed to the PAR's using the designated waste stream or by a measure of easily observed Cs-137 to which all other PAR's are normalized. Such normalization is very important to designate solid waste as LLW or to classify it as TRU.

In mid-2005 the SRS Burial Ground was processing a container labeled as *Box 33* for which no reliable solid waste stream designation was available. The facility suspected the high density waste was recovered residue from development tests of the In-Tank-Precipitation (ITP) facility. This facility was originally planned as the processing plant for liquid high level radioactive fission product waste that contained only trace TRU quantities.[2] Selective in-tank precipitation of radioactive Cs-137 and Tc-99 would yield a supernatant liquid with reduced activity suitable for processing as low-level liquid waste while producing a highly-radioactive sediment with nearly exclusively Cs-137 content. Confirming the genesis of this solid waste would allow determination of the remaining 24 PAR's by calculation from the assayed Cs-137 content.

The container labeled *Box 33* consisted of an outer box of dimensions 48"x46"x66" and an inner box that contained high density and high radiation dose material. From the outer box, Radiation Control (RCO) measured an extremity dose rate of 22mrem/hr. With the lid removed from the outer box, the maximum dose rate measured from the inner box was 100mrem/hr extremity and 80mrem/hr whole body. From the outer box the material plus container was sufficiently high in density that the Solid Waste Management (SWM) operators were unable to obtain a Co-60 radiograph of the contents. SWM requested that the Analytical Development Section (ADS) of Savannah River National Laboratory (SRNL) perform a γ-ray assay of the item to evaluate the radioactive content and to confirm the designation of solid waste stream. To confirm the waste stream required a measure of Cs-137 content with an estimation of limit of content for Co-60, Eu-152, and the TRU species Pu-238,239 and Np-237/Pa-233.

Experimental

Field γ-ray acquisitions were obtained using the ADS down-looking Ortec HPGe detector coupled to a Canberra Inspector multichannel analyzer and Genie2000 acquisition system using a laptop computer. The down-looking detector was designed for ADS to perform culvert γ-ray monitoring in 1998[(3)] and had been stored in various ADS laboratories since then. A photograph of the detector and frame is shown in Figure 1.

Figure 1. Photograph of the down-looking detector.

The detector was cooled with liquid nitrogen and tested in the SRNL nuclear nondestructive assay facility for use in this project. The system was energy and efficiency calibrated using a point source of mixed

radioactivity by placing the detector and frame on blocks with the mixed source on the floor. The mixed source contains Cs-137 and eight other radioactive species with well-known decay attributes that provide γ-rays with energy ranging from 60 keV up to 1836 keV.[4] A best agreement to the efficiency calibration curve is fit with the polynomial of equation (1) and is shown in Figure 2. The point source absolute efficiency for the 662-keV peak from Cs-137 decay was measured to be 1.75×10^{-4} at twelve inches.

$$Ln[Eff(E)] = -8.705 + 0.4315x + 0.1683x^2 + 0.3003x^3 - 0.3060x^4, \quad (1)$$

where γ-ray energy E is in units of keV, and x equals the unusual form of ln[706.8/E].

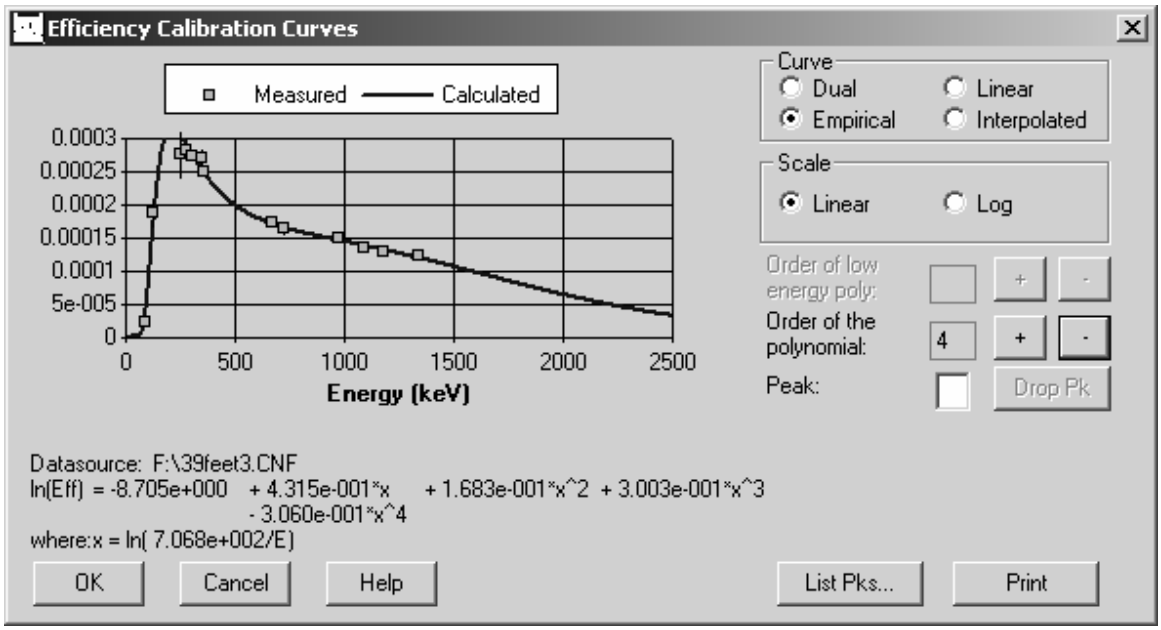

Figure 2. Graphical Representation of the Down-Looking HpGe Efficiency Calibration Curve.

After conferences with the appropriate Solid Waste Management Facility (SWMF) operations personnel to identify and mitigate hazardous job performance obstacles, the SWMF management and operators performed an Automated Hazard Analysis and developed a pre-job briefing packet for γ-ray acquisitions and subsequent gamma-radiography transmission measurements. The down-looking detector was transported from SRNL to E-Area using the Site Rigging group. E-Area Rigging became responsible for transport and lifting of the detector as well as transport and handling of the *Box 33* waste item.

Photographs of the acquisitions and preparations for the acquisitions are shown in Figures 3 and 4. Figure 3 shows the *Box 33* item with the detector and frame sitting beside it. The crane is also shown in Figures 3 and 4 as it is being used to remove the lid from *Box 33* and to lift the detector. Figure 4 shows the detector being lifted above Box 33 for subsequent γ-ray acquisitions. Survey acquisitions were obtained at multiple detector heights in order to obtain an estimate of the detector dead-time. Because of the very high activity coming from the top of the box with the lid removed, it was necessary to raise the detector to a height of 33 feet above the box to obtain an acceptable dead-time of 15%.

Figure 3. A photo of the field acquisitions showing the cover being lifted from the outer container.

Figure 4. A photo of the field acquisitions showing the box, detector, and crane.

The data acquired are summarized in Table 1. We list acquisition name, height above the sample, and area in the 661.6-keV peak from Cs-137 decay. Figure 5 shows a typical spectrum. In all ten spectra Cs-137 was the only γ-ray peak observed. A single background spectrum was obtained that is not shown in this report. Compared to the activity observed in the item, the background was so low that we disregarded it as insignificant when performing the calculations described below.

Table 1. List of Gamma-Ray Spectra Acquired for Box 33.

Acquisition Name	Distance (ft)	Count Time (sec)	661.6 keV Area	σ	Cs-137 (cps)
15Feet1	15	60	92689	380	1544.8±6.3
33Feet1	33	60	25037	170	417.3±2.8
33Feet2	33	60	25371	167	422.8±2.8
33Feet3	33	60	25347	170	422.4±2.8
33Feet4	33	60	25213	167	420.2±2.8
39Feet1	39	60	17926	142	298.8±2.4
39Feet2	39	60	17938	142	299.0±2.4
39Feet3	39	60	18087	143	301.4±2.4
39Feet4	39	60	17937	144	299.0±2.4
12Feet1	12	64.57	42507	272	658.3±4.2

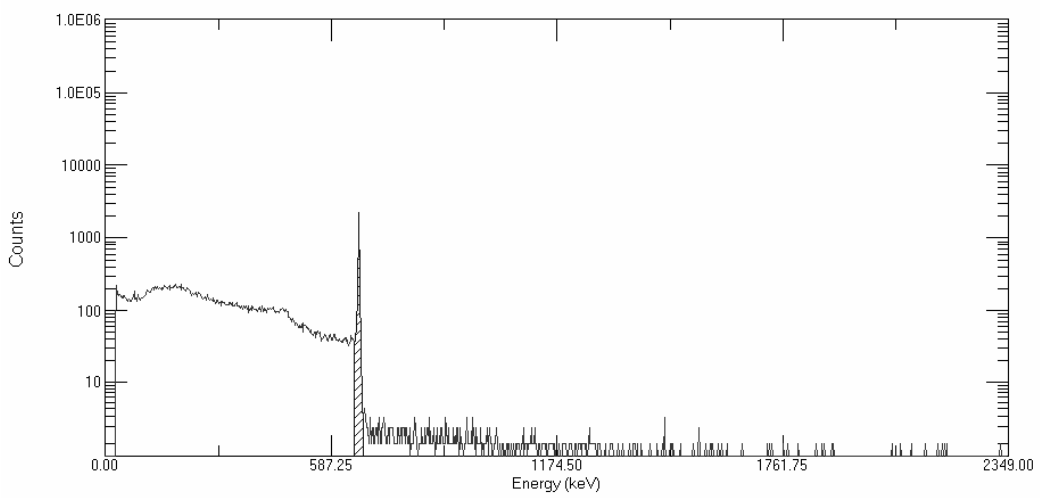

Figure 5. Spectrum *39feet3*.

Results

Because of the very large acquisition distances, each spectrum is approximately a point source acquisition configuration. The eight spectra obtained from 33 feet and from 39 feet were evaluated as both a simple point source and as an extended point source using the Ortec computer code ISOTOPIC.[5] We used the point kernel photon transport code MicroShield[(6)] to obtain a good estimate of the sample self-absorption of the 662-keV photon from Cs-137 decay. All of the calculations are described in this section.

The point source content of Cs-137 in each acquisition is obtained from the measured detection rate of the 662-keV γ-ray using equation (2).

$$Activity\ {}^{137}Cs = K_p (d)^2 (cps)(Cf_T), \quad (2)$$

where K_p is the point source calibration factor for the 25% HPGe detector, d is the source to detector distance in units of inches, cps is the measured detection rate of the 662-keV photopeak, and Cf_T is the transmission correction factor for the item observed. For the down-looking HPGe detector the point source efficiency for Cs-137 is 1.75×10^{-4}, which then yields a point source calibration factor of 0.00126 μCi-sec/in^2.

$$K_p = \mu Ci/\{(37000)(eff)(branch)(d^2)\} = 1/(37000)(1.75 \times 10^{-4})(0.851)(12)^2 = 0.00126\ \mu Ci\text{-}sec/in^2. \quad (3)$$

The transmission correction factor was determined using the computer codes ISOTOPIC and Microshield.[5,6] The Microshield code uses operator input of sample dimensions, content, density, and chemical makeup of the absorbing matrix to determine γ-ray fluence at a selected energy onto a selected point (i.e. the detector). By comparing the fluence of a given photon calculated with the item full of air with the calculated fluence with the sample full of matrix, the operator is able to gain a direct measure of the photon absorption characteristics of the actual sample. Using this technique we were able to calculate a transmission correction factor for the 662-keV γ-ray that we applied to all eight spectra that we analyzed. For all of the calculations we obtained a transmission correction factor of Cf_T(Microshield) = 15. For acquisition *33feet3* and using eq. (2) this yields a content of 1.25Ci Cs-137.

Note this point source calculation underestimates the Cs-137 content because the 66-inch item is not a point source. The code ISOTOPIC is able to convert the point source calibration to an extended point source item. This conversion effectively inserts a correction C_w into equation (1) to account for the object's shape. The extended point source correction is both sample shape and distance dependent and is described further in references 7 – 9.

The code ISOTOPIC also uses operator input of sample dimensions, content, density, and chemical makeup of the absorbing matrix to determine γ-ray transmission through the sample and matrix and to perform a geometry correction to transform the point source calibration factor to an extended source. For the 33-foot acquisitions the correction C_w is 1.166, and for the 39-foot acquisitions the correction is 1.145. The results of all of the calculations are listed in Table 2.

Applying the geometry correction to (2) yields

$$Activity\ ^{137}Cs = K_p (d)^2 (cps)(Cf_T)(C_w). \quad (4)$$

The results of (4) are listed in column 4 (Microshield) of Table 2.

The ISOTOPIC code calculation uses equation (5).

$$\text{Activity } ^{137}\text{Cs} = (\text{cps})(Cf_T)(\text{geometry})/[\text{efficiency}(662, 12")][\text{branch}], \quad (5)$$

where Cf_T comes from the ISOTOPIC calculation of sample and matrix absorption. The geometry factor is a product of the C_w factor above and of the acquisition distance relative to 12 inches. Recall in the discussion above, the down-looking detector was efficiency calibrated in the point source acquisition configuration at a distance of 12 inches. Therefore the geometry factor determined by the code ISOTOPIC is intended to correct for both acquisition distance and shape.

For acquisition 33feet3 ISOTOPIC determines an activity of

$$\text{Activity } ^{137}\text{Cs} = (422.8)(17.42)(1270.19)/[0.000175][0.8521] = 6.272 \times 10^{10}\ \text{dps}. \quad (5)$$

Converting to Ci, we obtain the values listed in the last column of Table 2. All are in good agreement with the Microshield calculations. The uncertainties in both results columns of Table 2 are dominated by the uncertainty in the transmission factor calculated by the Microshield and Isotopic programs. The transmission-corrected results were reported in reference 10.

Table 2. Measured Cs-137 Content calculated by MicroShield, ISOTOPIC, and by GADRAS for the eight far field acquisitions of Table 1.

Acquisition	Area (cps)	sigma (cps)	Cs-137 (Ci) MicroShield	Cs-137 (Ci) Isotopic	Cs-137 (Ci) GADRAS
33Feet1	417.3	2.8	1.44±0.64	1.71±0.85	1.22±0.54
33Feet2	422.8	2.8	1.46±0.65	1.73±0.87	1.21±0.60
33Feet3	422.4	2.8	1.46±0.65	1.73±0.87	
33Feet4	420.2	2.8	1.45±0.64	1.72±0.86	
39Feet1	298.8	2.4	1.42±0.63	1.65±0.83	1.65±0.83
39Feet2	299.0	2.4	1.42±0.63	1.65±0.83	1.61±0.71
39Feet3	301.4	2.4	1.43±0.64	1.66±0.83	1.69±0.85
39Feet4	299.0	2.4	1.42±0.63	1.65±0.83	

We have also included a determination of Cs-137 content in five of the spectra using the GADRAS fitting program.[11] This software uses previous detector characterization to fit an entire spectrum including the Compton continuum to estimate the contribution from single or multiple sources. With GADRAS we were able to fit the residue of the photopeak plus the Compton to determine contributions from selected species. A screenshot of the fit obtained for spectrum *39feet1* including contributions from Co-60, Eu-152, and Np-237/Pa-233 is shown in Figure 6.

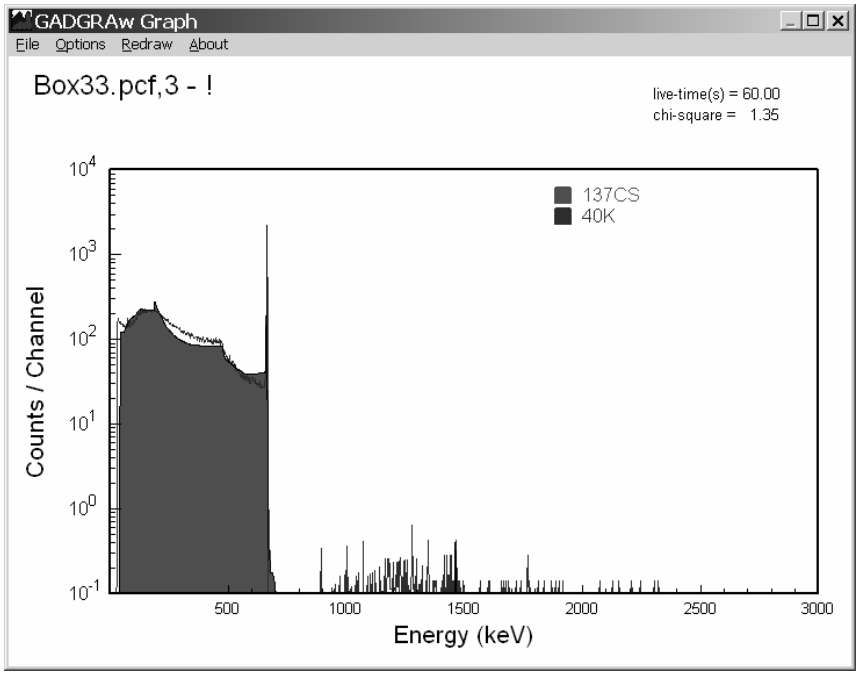

Figure 6. GADRAS Screenshot fit of photopeak and Compton continuum for spectrum 39feet3.

Discussion

Using the technique of equation (5) the limits of content were calculated for each of the species Co-60, Eu-152, Pu-238,239 and Np-237/Pa-233. The results are listed in Table 3. Clearly the calculated limits of content for Pu-238 and Pu-239 are so high that they provide no assistance to characterize the waste as LLW. While the actual limits of detection in the γ-ray spectra are not especially large, the branching ratios of 0.0000101 for the Pu-238 γ-ray and 0.0000151 for the Pu-239 γ-ray are too small. The branching ratios dominate the calculation in (5), and for Pu-238 the transmission correction factor is also very large due to the low energy of the 152-keV transition.

Table 3. Limits of Content for Co-60, Eu-152, Np-237/Pa-233, and Pu-238,239.

	Co-60 (1172 keV γ-ray)	Eu-152 (1408 keV γ-ray)	Np-237/Pa-233 (312-keV γ-ray)	Pu-238 (153-keV γ-ray)	Pu-239 (414-keV γ-ray)
Experimental (ISOTOPIC)	< 0.00019 Ci	< 0.00016 Ci	< 0.011 Ci (< 1.5 g)	< 630 Ci (< 37 g)	< 250 Ci (< 4000g)
Experimental (GADRAS)	< 0.0005 Ci	< 0.005	< 1		
Predicted	< 0.0003 Ci	< 0.0003 Ci	< 0.0000006 Ci	< 0.0006 Ci	< 0.000006 Ci

From measured recovery of selected atomic and radionuclide species in references 12 and 13, we can predict the expected activities shown in the last row of Table 3. Because of the branching ratio problem elaborated above, the predicted and measured limits of content for the TRU species differ widely, however those limits agree fairly well for Co-60 and Eu-152. Another important factor is that the analytical γ-rays for the two non-TRU species are well above the Cs-137 Compton Scatter continuum. The contributions from Co-60, Eu-152 and Np-237 are estimated by GADRAS in the second row of Table 3.

Conclusion

We observed only Cs-137 in all of the acquisitions. The transmission-corrected results were reported in reference 10. The likely identification of this waste is the Cs-loaded ammonium molybdatophosphate (amp-1) cation exchange resin from the former In-Tank-Precipitation project. The yellow color and density values observed agree well with those of the matrix material.

A very positive result is that we observe good agreement between the three modeling techniques ISOTOPIC, Microshield, and GADRAS. The former two modeling techniques use matrix characteristics to determine sample self-absorption and then use the observed photopeak to determine Cs-137 content and uncertainty. The GADRAS technique uses no knowledge at all of the sample characteristics, but rather use detector characterization to model the photopeak plus Compton continuum. Only the source to detector distance is used in the final calculation of content. The three models yield excellent agreement within uncertainty. The uncertainty is driven by the large matrix absorption. We also observe good agreement between limits of detection for the species with γ-ray branches larger than 1%.

References

1. US Department of Energy Order 435.1, (1999).

2. D. V. Osteen, Westinghouse Savannah River Company internal report, "Process Requirements for the In-Tank Precipitation Process", WSRC-IM-91-63, (1992).

3. 4. K. J. Hofstetter, and R. A. Sigg "High Resolution Gamma-Ray Spectrometry of Culverts Containing Transuranic Waste at the Savannah River Site", Proceedings of the 1992 Topical Meeting on Advances in Reactor Physics, 1, (1992), 1-155.

4. R. J. Haslett, Analytics Inc. standard radionuclide source number 65996-147 Certificate of Calibration, April (2003).

5. Ametek Ortec Gamma-ray analysis computer code Isotopic, version 2.0.6, (2004).

6. Grove Engineering, Radiation transport computer code Microshield, 5.03, (1992).

7. P. A. Russo, "Gamma-Ray Measurements of Holdup Plant-wide: Application Guide for Portable, Generalized Approach", LA-14206, Los Alamos National Laboratory, (2006).

8. R. A. Sigg, V. R. Casella, and R. A. Dewberry, "Nondestructive Assay Efficiency Calibration of HPGe Detectors for the Ortec ISOTOPIC Method, WSRC-TR-2005-00431, (2005).

9. R. A. Dewberry, V. R. Casella, R. A. Sigg, S. R. Salaymeh, F. S. Moore, and D. J. Pak "Holdup measurements for three visual examination and TRU remediation glovebox facilities at the Savannah River Site", **Journal of Radioanalytical and Nuclear Chemistry**, Vol. 275, No.3, (2008) 541.

10. R. A. Dewberry, R. A. Sigg, and S. R. Salaymeh, "Evaluation of γ-ray Acquisitions of Box33 Item", SRNL-ADS-2005-00526, August (2005).

11. D. Mitchell, Gamma Detector Response Analysis Software (GADRAS), Gamma Analysis Software, Version 15.1.2, Sandia National Labs, (2004).

12. R. A. Dewberry and C. J. Coleman, "Tests of Cs-137 Removal from DWPF Samples Prior to Analyses", WSRC-MS-94-0590, Presented at the Nuclear Chemistry Symposium at the annual meeting of the American Chemical Society, Wash. DC, August 1994..

13. J. L. Thomas, Westinghouse Savannah River Company internal report, "Tank 48 Radionuclide Characterization", CBU-PIT-2005-0004, (2006).

Acknowledgements

We are especially grateful for the guidance and assistance provided by Kevin Tempel, Chris Bush, and Todd Horton throughout the experimental acquisitions of the Box33 item.