Radioactivity in Trinitite - a review and new measurements

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Abstract

Samples of Trinitite and soil from Trinity site were studied in the radioactivity measurements laboratory at the University of Bremen and at the authors' facilities in Anaheim (JCR) and Edgewater (WMK). Gamma spectroscopy was used to identify and quantify radionuclides in Trinitite and to perform a radiometric characterization of soil at the Trinity site. Additionally, a similar material ("atomsite") formed during a soviet test at the Semipalatinsk nuclear test site was investigated. Fission products (^{137}Cs, ^{155}Eu) together with activation products (^{60}Co, ^{133}Ba, ^{152}Eu, ^{154}Eu, ^{241}Am) and ^{239}Pu were identified. A literature search including some publicly available archive sources was conducted and our data compared to previously published results. Obtained data on Trinitite were also compared to literature data on atomsite formed during atmospheric nuclear tests in Algeria. Variability of radioactivity in Trinitite and relationship of distance from the ground zero and activation were discussed.

Introduction

Trinity

The terms Trinitite or atomsite are used for a fused glass-like material formed during the first nuclear test in the desert in White Sands Missile Range, New Mexico, USA, on July 16, 1945. Its colour is usually greyish-green, the top surface (facing the explosion) is smooth and typically more active than the bottom side, which is rougher and contains sand and small stones from the desert surface. The thickness of the Trinitite layer varies usually between 0.5 and 1 cm and the material contains plenty of air inclusions. Trinitite was cleared from the area by the Atomic Energy Commission in 1952, but visitors of the Trinity site can still find small pieces on the ground. Although it is illegal to remove Trinitite from the site now, specimens can be found for sale on the internet and from mineral dealers. The Ground Zero (GZ) of the Trinity test, which is a National Monument since 1965, lies within the military area and can be visited by the public twice a year, always in April and in October (WSMR, 2010).

The physical properties of the glass formed during the first nuclear explosion are described in LA-1126 report (Staritzky 1950). The glass covered an area of about 610 m diameter with a total estimated mass of $17 \cdot 10^5$ kg. A health physics survey more than 20 years after the Trinity test was made to determine the radiological risk for the public visiting the site (Fey 1967). In this study, monitoring of samples of Trinitite carried away from the site for gamma-exposure rates, measurement of surface exposure rate from individual Trinitite pieces and calculation of deposition of radioactivity in the body from ingested Trinitite were performed. In a later report (Hansen, Rodgers 1985), radiological conditions were evaluated for the Trinity site and the associated fallout zone. Here also qualitative data for activity concentrations of selected radioisotopes in the soil at GZ were given.

As for scientific literature easily accessible to the public, Atkatz & Bragg published a Trinitite NaI gamma spectrum in 1995 and from ^{137}Cs activity they calculated the explosive yield of the device. In a response to the paper, Schlauf et al. (1997) showed advantages of HPGe spectroscopy and identified other gamma emitters in Trinitite. Sixty years after the Trinity test, a comprehensive study of Trinitite radioactivity by means of alpha-, beta- and gamma- spectroscopy was published (Parekh et al., 2006). Among other findings, the authors used ^{152}Eu as a slow neutron flux monitor and conducted isotopic analyses of Pu in Trinitite samples. In a subsequent study (Semkov et al 2006) based on isotopic data, calculations and modeling, parameters characterizing the Trinity test were determined. Additionally, based on non-radioactive Trinitite properties, Hermes and Strickfaden (2005) devised a new theory on its formation during the explosion. The majority of the Trinitite layer was formed not on the ground, but by a rain of molten glass. After falling to the ground, the surface of Trinitite was further heated by the fireball and developed a smooth surface.

Fig. 1. A specimen of Trinitite. Left: top side. Middle: edge. Right: bottom side. Mass 3.170 g.

Semipalatinsk test site

The USSR conducted its first nuclear explosion at the Semipalatinsk test site in Kazakhstan on August 29, 1949. The Soviets coded it RDS-1 (the acronym is not well understood), while the American intelligence designated it Joe-1, after Joseph Stalin. The tested weapon was a plutonium bomb design similar to that in Trinity, detonated on a tower. The yield was estimated to 20 kt TNT.

Although not much information has been published about radioactivity at the Semipalatinsk test site, there is evidence that the first Russian explosion also created pieces of fused rock at GZ (Kruglov 2002, Hodge and Weinberger 2008). They are called Kharitonchiki in honour of one of the leading Russian nuclear weapons scientists, Yuly Khariton, but are more broadly referred to as atomsite. The authors are not aware of any clean-up involving removal of atomsite, as in the case of Trinity site.

Information on activity concentrations of radionuclides in the upper 2-3 cm of soil at the site of the RDS-1 test at the Semipalatinsk test site has been published by Yamamoto et al. (1996). A mainly in-situ gamma spectroscopic study was performed by Shebell & Hutter (1998).

Fig. 2. A specimen of atomsite from Semipalatinsk test site. Left: top side. Middle: edge. Right: bottom side. Mass: 2,545 g.

Algeria

France performed 4 atmospheric nuclear tests in the Algerian part of the Sahara desert between 1960 and 1961 at the Saharan Military Test Centre near Reggane. The first of the tests, conducted on February 13, 1960, was called Gerboise Bleue. The fission device was detonated on a 100 m tower with an estimated test yield of 40-80 kt. In 1999 the IAEA conducted a field expedition with the goal of evaluating residual activity due to the atmospheric nuclear tests (IAEA, 2005, Danesi et al. 2008). The atomsite from Gerboise Bleue test is described as black, vitreous and porous material, typically 100-1000 times more active than unmelted sand.

Material and methods

Trinitite[1] samples analyzed in the Radioactivity Measurements Laboratory, University of Bremen (Landesmessstelle für Radiaktivität, LMS) included 6 individual larger pieces of 1.4-3.8 g each (one of them in Figure 1), 3 plastic cylindrical containers filled with many small pieces (68, 50 and 14 g each) and 2 samples of powdered Trinitite (1.9 and 1.1 g). Additionally, a 103 g bulk sample of soil from the upper 5 cm of soil collected outside the inner fence, in the distance of approx. 100 m from GZ, was measured. The samples were analyzed by low-level low-background gamma

[1] Most of the Trinitite in the present study is from the Derik Bower collection. Based on personal communications with Ralph Pray (April 2006 to September 2009) and Derik Bower (January 2003 to October 2008), it is almost certain this material was collected by Ralph Pray in the summer of 1951. It would have been located on the south road leading to GZ, possibly between 150 and 220 m from GZ.

spectroscopy using a coaxial HPGe detector (Canberra Industries) of 50% rel. efficiency housed in a 10 cm Pb shielding with Cu, Cd and plastic lining, operated under Canberra Genie 2000 software. Efficiency calibration was performed for each individual sample separately based on its individual size, density and geometry relative to the detector using the Monte-Carlo based LabSOCS Genie 2000 calibration tool (Bronson 2003). Since some of the analyzed gamma emitters have complicated decay schemes (^{133}Ba, ^{152}Eu), the probability of detecting two photons emitted by the same decaying nucleus as one (a phenomenon known as cascade summing), is not negligible (up to 25%), therefore a cascade summing correction has been applied.

Spectra of another piece of Trinitite (#26) purchased from United Nuclear and a sample of atomsite from the Semipalatinsk test site (Figure 2) were obtained using the author's (JCR) Ortec LO-AX-51370/20 coaxial HPGe detector shielded by approximately 7cm Pb, lined with Cu and Sn foil within PVC and acrylic layers. Samples collected by the author (WMK) in October 2006 at Trinity site with known locations around GZ were measured using the author's (WMK) 5" Bicron NaI(Tl) well detector housed in 2 cm of Pb shileding with Cu foil grading. For the two later mentioned setups the efficiency calibration as described above was not achievable, therefore the resulting spectra were used for qualitative and comparative purposes only.

Results

Gamma spectroscopy of Trinitite enabled identification of several natural and artificial radionuclides, formed as a result of fission and activation or found as remains of nuclear fuel. A gamma spectrum of one of the samples is shown in Figure 3. Radionuclides are listed in Table 1 together with remarks on their origin. Some of the radionuclides (^{60}Co and ^{155}Eu, which were not reported in previous studies) were initially present in very high concentrations; therefore it is possible to detect them, even after more than 10 half-lives. Table 2 gives the specific activity of individual isotopes in Trinitite compared to literature values (Atkatz & Bragg 1995, Schlauf et al. 1997 and Parekh et al 2006).

Table 1. List of detected artificial gamma emitters artificial in Trinitite and their origin

Isotope	Half-live (yr)	Origin
^{60}Co	5.3	Activation of ^{59}Co – from test tower steel and from soil
^{133}Ba	10.5	Activation of ^{132}Ba. Baratol - Ba $(NO_3)_2$ - was part of explosive lens system of the Gadget
^{137}Cs	30.0	Fission product (beta decay of ^{137}Xe and ^{137}I and also independently)
$^{152, 154}$Eu	13.3 / 8.8	Activation of stable isotopes ^{151,153}Eu in soil by slow neutrons
^{155}Eu	4.8	Fission product
^{239}Pu	24110	Principle isotope of nuclear fuel
^{241}Am	433	Mostly present as daughter product of ^{241}Pu (beta emitter), produced mainly from ^{239}Pu during the explosion via double-neutron capture. Based on ^{241}Am ingrowth activity of ^{241}Pu is possible to determine.

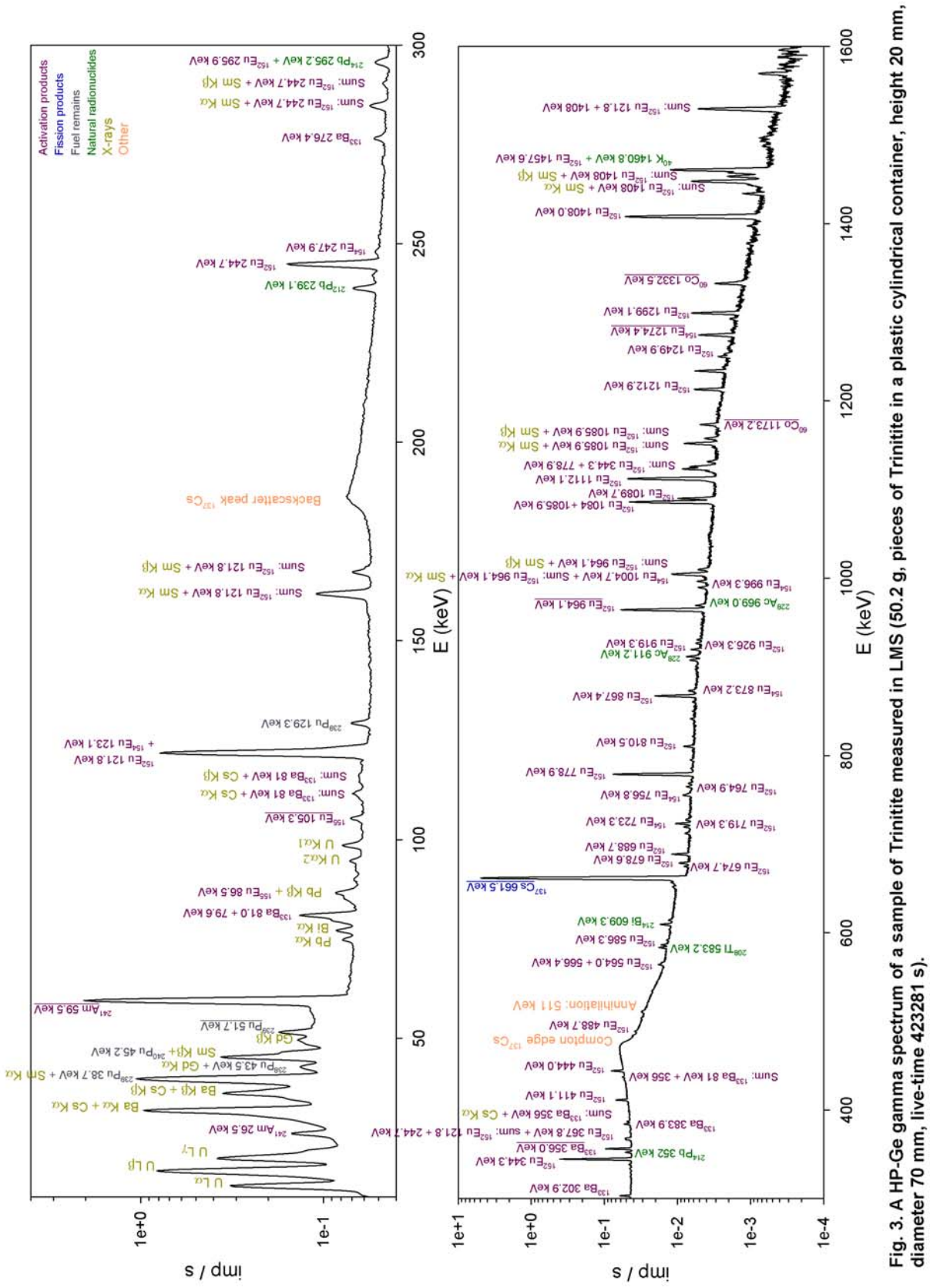

Fig. 3. A HP-Ge gamma spectrum of a sample of Trinitite measured in LMS (50.2 g, pieces of Trinitite in a plastic cylindrical container, height 20 mm, diameter 70 mm, live-time 423281 s).

Table 2. Table of comparison of activity concentrations (Bq/g, uncertainties 1σ) of artificial radionuclides detected in Trinitite and soil from nuclear test sites recalculated to the date of respective nuclear explosions (^{241}Am, which is ingrowing from ^{241}Pu, is reported to the date of analysis - in brackets).

Radio-isotope	LMS, 2010	Atkatz & Bragg, 1995	Schlauf et al., 1997	Parekh et al., 2006	IAEA, 2005	LMS, 2010	Hansen & Rodgers, 1985	Yamamoto et al., 1996
	Median, Min - Max			Min, Max	Min - Max	Min, Max	Min - Max	
	TRINITITE				ATOMSITE ALGERIA	SOIL TRINITY		SOIL SEMIPALATINSK
Bq/g	11 samples of Trinitite (see text)	Small piece	8.3 g piece	3 samples: several small pieces and 2 bigger individual pieces	Black fragments of fused sand	2 samples of soil (see text)	Samples at GZ area	2-3 upper mm of soil
^{60}Co	47.7 ± 4.6 <51.8[1] - (72.5 ± 4.8)	-	44 ± 4	44.4 ± 4.6 62.0 ± 4.9	168.9	2.9 ± 0.8 20.8 ± 1.3	64 - 320	1946 ± 43
^{133}Ba	9.10 ± 0.77 (5.04 ± 0.51) - (17.8 ± 1.3)	-	9.9 ± 0.6	7.55 ± 0.45 9.80 ± 0.26	48.9	0.104 ± 0.009 0.36 ± 0.03	0.73 - 1.73	-
^{137}Cs	48.3 ± 1.4 (16.26 ± 0.88) - (80.9 ± 2.4)	83.2	90 ± 9	27.33 ± 0.08 121.8 ± 0.1	78.1	0.142 ± 0.004 0.879 ± 0.026	0.38 - 1.87	236 ± 2
^{152}Eu	26.0 ± 1.1 (14.2 ± 1.3) - (55.5 ± 1.3)	-	27 ± 1	22.61 ± 0.38 78.89 ± 0.61	54.0	7.92 ± 0.19 33.42 ± 0.79	3.2 - 347	998 ± 10
^{154}Eu	7.08 ± 0.24 <6.03[2] - (12.76 ± 0.36)	-	4.8 ± 0.6	2.45 ± 0.60 16.1 ± 1.3	27.2	1.07 ± 0.08 4.80 ± 0.28	-	100.8 ± 4.5
^{155}Eu	274 ± 25 <361[3] - (461 ± 54)	-	-	-	241.2	<28.4 10.8 ± 2.8	-	-
^{239}Pu	73.6 ± 3.5 (25.7 ± 3.6) - (133 ± 25)	-	-	86.3 ± 2.7	<230	< MDA < MDA	-	-
^{241}Am	1.870 ± 0.080 (0.741 ± 0.048) - (4.47 ± 0.19) (2009)	-	2.9 ± 0.5 (1997)	1.841 ± 0.053 4.137 ± 0.058 (2006)	2.3 (1999)	0.0082 ± 0.0004 0.0369 ± 0.0018 (2009)	-	0.52 ± 0.01 (1994)
^{241}Pu[4]	63.6 ± 2.7 (25.2 ± 1.6) - (152.0 ± 6.5)	-	100 ± 17	62.9 ± 1.8 141.3 ± 2.0	85	1.254 ± 0.061 0.279 ± 0.014	-	18.6 ± 0.4

Several values measured by LMS were below detection limit, mainly due to low sample mass and/or shorter measurement times.
[1] 3 values under MDA, minimal value above MDA is 34.5 ± 5.6. **[2] 3 values under MDA, minimal value above MDA is 2.70 ± 0.07.**
[3] 5 values under MDA, minimal value above MDA is 115 ± 38. **[4] Calculated from ^{241}Am ingrowth**

The spectrum of radionuclides (with exception of ^{239}Pu) found in Trinity soil was similar to that of Trinitite. The absolute activities of ^{152,154}Eu and ^{60}Co, isotopes being formed by activation of elements present in soil itself, were present in the same order of magnitude as in Trinitite. The activity concentrations of ^{137}Cs, ^{133}Ba and ^{241}Am were 1-2 orders of magnitude lower in soil than in Trinitite (Table 2).

Discussion

Correlations

Correlation analysis was performed for the set of 11 Trinitite samples measured in LMS using Pearson test (normality was positively tested by Shapiro-Wilk test). Significant positive correlation was found for ^{152}Eu and ^{154}Eu (r=0.823;P=0.016). That is in agreement with similar origin of both isotopes (activation of stable Eu isotopes from soil). On the other hand, activation product ^{152}Eu and fission product ^{155}Eu are not correlated (r=0,305; P=0,557). ^{239}Pu and ^{241}Am, which is present as a decay product of ^{241}Pu (activation of fuel during the explosion), show strong positive correlation (r=0.967; P=0,000001). Activation product ^{133}Ba is positively correlated with ^{239}Pu (r=0,768; P=0,006) and ^{241}Am (r=0.785; P=0,004). Fission products ^{137}Cs and ^{155}Eu were significantly correlated with ^{239}Pu (r=0,827; P=0,0017 and r=0,836; P=0,038, respectively) and similarly with ^{241}Am. Positive correlation was also found for ^{137}Cs and ^{152}Eu (r=0,703; P=0,0159). ^{60}Co is not correlated to any other radioisotope.

Inhomogeneous distribution of radioactivity in Trinitite

The difference of activity at the top surface and the bottom surface of Trinitite is remarkable, mainly for beta activity. Measurements performed on 6 Trinitite specimens collected by WMK in 2006 using a 2" pancake tube inside a plastic bag to stop alpha radiation showed the top/bottom surface ratios ranging between 2.6 and 22.9 (with exception of one sample of unusual appearance showing ratio <1). Gamma measurements do not show such a pronounced difference due to higher penetrability of gamma radiation. A repeated measurement of one piece of Trinitite on a HPGe detector (LMS) top side up and top side down showed top/bottom ratios significantly increased for ^{137}Cs (1.24±0.05), ^{239}Pu (1.49±0.11) and ^{241}Am (1.25±0.08). Top/bottom ratios for ^{133}Ba and ^{152}Eu,however, were close to 1 (1.0±0.1 and 0.96±0.09, respectively).

Variability of radionuclides in Trinitite

While some variation in radionuclide activity (Table 2) can be attributed to sample size and normal variance, differences due to location are also expected and have been observed. Semkow et al (2006) found a power-law dependence between ^{152}Eu and slant range to the nuclear explosion center. Semkow calculated a least-squares exponent of -6.257 using 1946 data from 300 to 500 m (instruments closer to GZ were destroyed), whereas a spherical spread of neutrons would have resulted in an exponent of -2. Trinitite collected by the author (WMK) in October 2006 from 18 locations between 60 and 260 meters from GZ did, however, exhibit a power-law dependence with an exponent of -2.03 and least-squares correlation of R^2=0.777. The activity in counts-per-hour per gram at 1112 keV was found to be 68350 $s^{-2.03}$, where s is the slant range in meters to the top of the 30 m tower. The same specimens showed no significant

relationship between ^{137}Cs activity (a fission product) and slant range using the 662 keV peak. It is not known how site clean-up activities may have affected the distances measured to GZ but evidence found during the 2006 survey suggests the specimens were probably within a meter of two of their original locations.

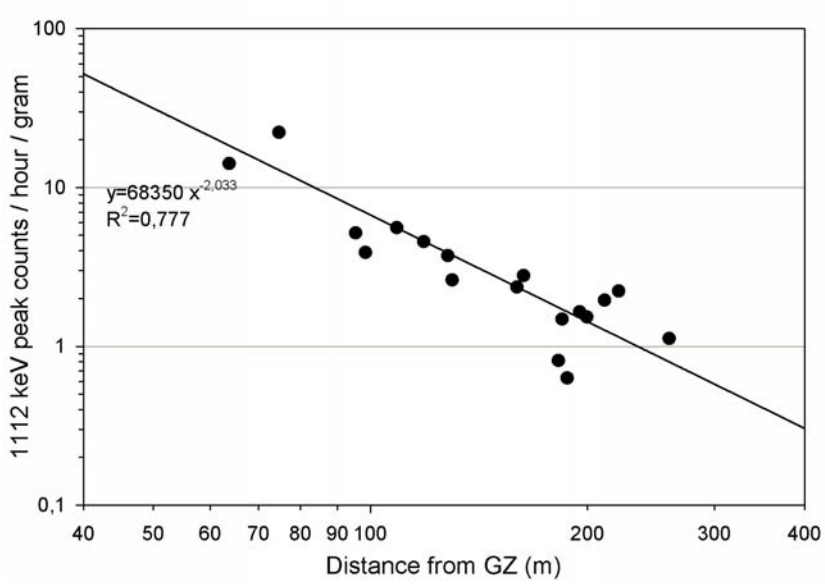

Fig. 4. ^{152}Eu activity vs distance from GZ in Trinitite samples collected in October 2006 by WMK measured with 5'' NaI(Tl) well detector.

Comparison of Trinitite to atomsite from Semipalatinsk

Gamma activity of a 2.545 g specimen of atomsite (Figure 2) from Semipalatinsk was compared to a typical 6.5 g specimen of Trinitite (gamma spectra in Figure 5). The net peak areas of radionuclides measured in both spectra were corrected by respective masses and decay corrected to the date of explosions (1945 and 1949, respectively). Atomsite from Semipalatinsk showed significantly higher activity than Trinitite. The activity ratios of Semipalatinsk/Trinity atomsite were: ^{137}Cs: 16.1±0.1, ^{152}Eu: 23.0±1.5, ^{241}Am (^{241}Pu): 3.4±0.1 and ^{239}Pu: 3.4±0.6. ^{154}Eu and ^{60}Co were under detection limits in Trinitite. ^{133}Ba was not detected in atomsite from Semipalatinsk. This is surprising due to the fact, that Baratol was also used in explosive lenses in the first Soviet nuclear test (Kruglov 2002).

Comparison of Trinitite to atomsite from Algeria

Literature values of a sample from Gerboise Bleue atomsite from Algeria (IAEA 2005 – sample No. ALG 4) described as black fragments of fused sand were compared to values of Trinitite measured by LMS (Table 2). Generally, the Algerian atomsite is more radioactive (activities recalculated to dates of explosions). ^{137}Cs value is about 1.5 times higher than median Trinitite values, ^{152}Eu 2 times higher, ^{154}Eu 4 times higher, ^{60}Co 3.5 times higher, ^{133}Ba 5 times higher. ^{155}Eu and ^{241}Am values are comparable for both Trinitite and atomsite from Algeria. Higher radioactivity in Algerian atomsite is likely due to the higher yield of the tested device.

Comparison of soil from Trinity to soil from Semipalatinsk test site

Soil data collected from the First Experimental site near the hypocenter where the first Soviet nuclear bomb was tested were published by Yamamoto et al (1996). They were compared to samples of soil from Trinity site. All activities were 1 or 2 orders of magnitude higher in soil from Semipalatinsk site than Trinity. The main difference was the absence of ^{133}Ba, as the activation product of stable Ba in baratol in Semipalatinsk site.

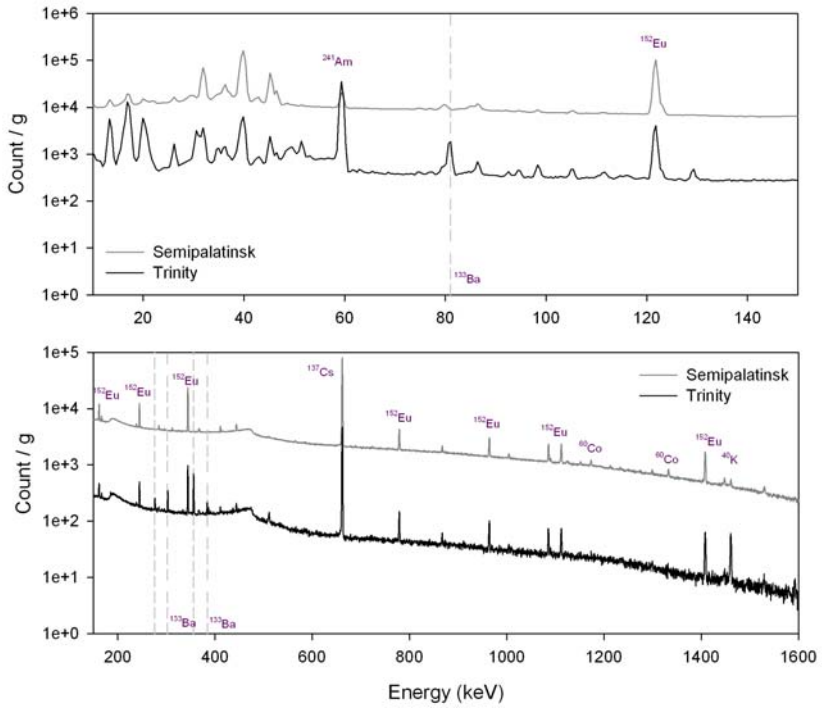

Fig. 5. 24 hour comparison spectra of atomsite from Semipalatinsk and Trinitite (sample #26) measured by Ortec LO-AX HPGe detector. Dashed lines indicate the main ^{133}Ba gamma lines in Trinitite and the lack of ^{133}Ba in the sample from Semipalatinsk.

Conclusions

Gamma spectroscopic studies of Trinitite, atomsite from Semipalatinsk and soil from Trinity was conducted. The experimental data were compared to literature data for Trinitite, atomsite from Algeria and soil from Semipalatinsk. ^{133}Ba (an activation product of Ba in explosive lenses) is present in Trinitite and atomsite from Algeria, but absent in atomsite and soil from Semipalatinsk. ^{155}Eu, a fission product, was found to be detectable in Trinitite after more than 13 half-lives. Correlations between radionuclides in Trinitite and the inhomogeneous distribution of radioactivity in Trinitite were discussed. Power-law dependence with an exponent close to -2 between ^{152}Eu and distance from GZ was found.

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