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RADIONUCLIDES IN RAINWATER AND THEIR IMPACT ON BACKGROUND RADIATION

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BY

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THESIS

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ABSTRACT

Radioactive waste has accrued throughout the continental United States and in the Oceans surrounding the country. Significant quantities of the waste are poorly documented regarding their location and the radioisotopes contained in the waste. Locating the waste is not an easy matter, as its locations are not well documented, and its method of disposal may be covered with plants, soils, and sediments. A common tool used to locate the waste is a gamma spectrometer, which measure the gamma emissions spectrum of radionuclides. The effectiveness of this tool when utilized to locate gamma emitting waste may be reduced due to the presence of various waters. Of the types of waters that may be present, including groundwater, rivers, floodwaters, rainwater, etc., rainwater may present the most significant obstacle. While all waters cause a degree of attenuation, rainwater will also introduce additional radionuclides into the area, increasing the quantity of gamma emissions present, and obscuring the signal that the wastes emit. Rainwater has been found to increase the quantity of gamma emissions by as much as 20% over the emissions present in the absence of rainwater. Depending on the radionuclides present in the waste, this may present a significant obstacle to accurately locating the waste.

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Chapter 1: Introduction

Radioactive waste has accumulated worldwide for the past 70 years and, as documented by nearly every medical organization and the U.S. Environmental Protection Agency (EPA) (EPA 2018), this waste presents a risk to living organisms. The location of much of this waste is well known, with these sites routinely inspected to determine if they are leaking and/or are causing harm to living creatures (IAEA 2007; FUSRAP 2018). However, there are many undocumented or poorly surveyed waste sites globally, with more than 500 sites listed in the Department of Energy's listing of sites "considered" for waste cleanup (WSJ 2014). The risks to the public of these undocumented sites are not fully understood, due to a lack of information concerning waste type stored and precise locations of the waste. There have been, and continue to be, concerted efforts to locate and assess the dangers associated with these potentially dangerous localities (NIOSH 2011).

Characterizing poorly documented or undocumented waste sites has become increasingly important, as a great deal of early radioactive waste was disposed of in metal containers or disposed of without any container at all, including being literally washed down the drain (IAEA 2007). Over time these containers corrode and eventually leak their contents. Since the effect that the contents may have on living organisms is poorly understood until the effects are manifest, it is important to locate the waste prior to the degradation and dispersal into the environment begins.

Attempts to locate the waste are hampered by several factors. These include poor record keeping of waste operations, imprecise location of dump sites, and lack of information on the contents of the waste containers (AEC 1956; IAEA 1999). In addition, waste may be covered by soil/sediment, which both limits the distance energetic particles can travel and impedes detection by remote measurement. The cost to locate waste sites is often high, as finding waste's location may require many man hours and specialized hardware. These factors are well understood, and usually taken into account when attempting to locate radioactive waste. In contrast, a poorly researched and understood factor is the role of water in the soil/sediment matrix. Specifically, the impact of rainfall on the attenuation of gamma rays and the subsequent impact on measurement/detection of these sources is poorly known.

This study will evaluate this last factor, namely determining the impact of water in the matrix, with an emphasis on rainfall, to ascertain the effects on attenuation and detection. Secondarily, since most technicians who run these detection protocols are not trained geologists, the research will help characterize different lithologies and provide guidance for subsequent surveys. The practical end goal of this research will be to provide valuable information to non-geologists that will make it easier to locate waste, make the detection more precise, and do so at lower cost. The scientific goal of this study will be to assess whether rainfall produces a measurable increase to the background radiation emitted in combination with different lithologies present in central and northern New Mexico.

Chapter 2: Scientific Background:

Historical Development & Approaches Used to Study Radionuclides Presence in Waters, and Transport Through Varying Lithologies, Sediment

The problem of radioactive waste arose at the dawn of the nuclear age, and arguably with the Manhattan Project, due to ignorance of the health effects of nuclear waste. Subsequently, facilities across the US began to manufacture radioactive components for the first nuclear weapon. After World War II, research continued with weapons applications, but also encompassed energy generation, medical applications, and food production. Early waste products from these endeavors were not disposed of in a manner that would be considered appropriate by today's standards (AEC 1956). Later, when the danger posed by the waste was better understood, it was still disposed haphazardly with little or no documentation, often due to the cost and effort necessary for proper disposal (AEC, 1956; IAEA 2007). Admittedly, the proper method for radioactive waste disposal is still a debatable topic nearly 70 years after the Manhattan Project.

The Formerly Utilized Sites Remedial Action Program (FUSRAP) identified 517 sites in the US alone that may necessitate cleanup (FUSRAP, 2017). Further investigative work by the Wall Street Journal (WSJ) uncovered ~100 additional sites that likely necessitate remediation (Vine et al., 2014) ***(Figure 2-1)***. Outside the US, the website Intercontinental Cry has identified sites throughout the world that are either radioactive waste sites, nuclear testing sites, or toxic sites (Ryser, 2016) ***(Figure 2-2)***. Other undocumented sites exist and may number in the 1000's. In addition, numerous reports,

most notably by the Atomic Energy Commission (AEC) and the International Atomic Energy Agency (IAEA) have detailed waste disposal at sea, which was apparently managed with even less oversight than waste sites on land (AEC, 1956; IAEA, 1999). However, and while of interest for future detection, the sites at sea fall outside the scope of this research.

Difficulties associated with locating radioactive waste are both manmade and natural. Manmade include improper record keeping with respect to the location and contents of the waste and in some cases, there was outright obfuscation of relevant data. The AEC showed that there was a lack of good record keeping prior to the early 1960's and suggested that significant quantities of radioactive waste were disposed of without mentioning the contents of the waste, method of disposal, nor the location of the waste (Joseph, 1957). In addition, an IAEA report noted that ocean dumping of waste continued through the late 1970s with little oversight in terms of properly documenting where the waste was disposed (IAEA, 1999). Additional work determining waste site locations was performed by FUSRAP, reporters for the WSJ (Vine et al., 2016), and a reporter from Intercontinental Cry (Ryser et al., 2014). These publications either specify the locations of waste sites, as is the case of the article released by FUSRAP (2017) or discuss the locations of possible sites that have not been properly investigated. The number of sites specified by the WSJ (2016) exceeds 600, and the Intercontinental (2014) lists another 40 locations.

The very nature of the soils and sediments that the waste is buried in or covered by are also a significant impediment in locating radioactive waste. Al-Masri et al. (2012) and

Costa et al. (2014) quantified the attenuation of gamma radiation by soils. Work by Toelke (1955), Brannon (1956), Michaelis (1980), Vasiliev (2003), McCay (2014) and a myriad of others have examined the attenuation of gamma rays by various lithologies through their work in and with the oil industry. Since most radioactive waste is likely to be buried or partially obscured by soils and sediments, its gamma radiation emission may be attenuated by the soil itself. This attenuation is caused by several factors, including soil elemental composition, density and moisture content.

Work performed by Reginato et al. (1964) and Abdel-hady (1996) involved assessing the attenuating impacts of the soil's moisture content. They found that it is also a factor in impeding the travel of gamma radiation through soils and sediments. The attenuation is caused primarily by the water filling the pore spaces of the soils and sediments, displacing air, thereby increasing the density of the soil or sediment. This attenuation would have an impact on the gamma emissions of any anthropogenic radioactive waste but would also attenuate the emissions that originate from the soils themselves. These emissions, and their origin, were the focus of work by Abdel-Hady (1996) and Shahbazi-Gahrouei (2013). They found that the soils themselves are also radioactive, and worked to quantify this radioactivity. Subsequent work by Mir at al. (2014), Ademola et al. (2014), and Mohammed et al (2013) sought to do the same. This research provided valuable information that was used to base expectations of gamma emissions and attenuation on.

The work that was performed by these researchers indicate that the waste disposed of in soils, sediments, etc., may be hidden by the soil or sediments through attenuation or

the soils or sediments natural emissions, density, and percentage of moisture in the soil. The degree to which detection was impaired depended on the types of radioisotopes present in the soil and waters, and their concentration. While most of these studies were performed in the laboratory, several field-based studies (Mohammad et al.,2013; Ajayi et al., 2015) utilized similar tools to what I used in the field,

This natural background radiation can be compounded; input from surface water, groundwater, and rain may mask the signal even further. Since most waste sites are in natural environments, there is an expectation that water (meteorologic, groundwater, soil moisture) will be present. The presence of radionuclides in these waters may raise the level of background radiation, making differentiation of waste gamma radiation from natural background radiation more difficult. Studies (Drndarski et al., 1988; Stralberg et al., 2003; Avwiri et al., 2007; Gainon et al., 2007; IAEA, 2016) have also sought to characterize the waters themselves, separate from the soils present. Duenas et al. (2010), Koike et al. (2013), and Ibikunle et al. (2016) detailed the presence of radionuclides in rainwater, which can also raise the level of background radiation.

Al-Masri et al. (2012) focused on soil attenuation of gamma radiation. The sites analyzed included a total of 60 different surface soil and sediment deposits, differentiated into 180 samples. The samples were air-dried and then analyzed with multiple energies of gamma radiation. Al-Masri et al. (2012) showed that attenuation decreased with increasing gamma ray energy ***(Figure 2-3)***. In addition to physical measurements, the authors also utilized a simulation program called X-com that programmatically determines the mass attenuation coefficient of a given sample, provided its elemental

constituents ***(Figure 2-4)***. The authors determined that sediment elemental composition and density have the greatest effect on gamma attenuation ***(Figure 2-6)*** and note that at low gamma energies (< 165 keV), soils with greater quantities of iron and calcium attenuate more than soils with lower concentrations. They did not seek to quantify the effects of soil moisture.

Costa et al. (2013), sampled a Red-yellow Latosol and a Red Nitosoil from Brazil, also utilizing X-Com for analysis but with a slightly different method, placing a source below the sample to measure the extent of attenuation by the intervening sample. Their results reinforced the well-expected observation that attenuation decreases with an increase in gamma radiation energy. They also chemically analyzed the samples ***(Figure 2-7)*** and found that the soils with the greater concentrations of heavy elements had, as expected, a higher level of attenuation ***(Figure 2-8)***. They also found that an increase in the thickness of the sample also caused the signal from a source to attenuate more. Like Al-Masri et al. (2012), they did not seek to quantify the effects of soil moisture.

Toelke (1955) laid the groundwork for utilizing gamma spectroscopy in well logging. The work was performed with core samples retrieved from boreholes. Work by Toelke was continued and built upon by Brannon et al. (1956), who also utilized a gamma spectrometer to examine core samples retrieved from a borehole. Their work was referenced through others, and was instrumental in work performed by Serra (1984), and Michaelis (1980), Vasiliev (2011), McCay (2014), and many others. Serra's article was the culmination of 50 years of work, and presented the fundamentals of well-logging interpretation, as well as acquiring the data to so. While Serra focused on

hydrocarbon exploration, Michaelis focused on utilizing spectrometer data to locate marine metal reserves. Work by Vasiliev (2011) and McCay (2014) examined new and novel ways to use gamma spectroscopy in locating hydrocarbon or geothermal sites. While these articles, and many others written for the hydrocarbon, geothermal, and mineral exploration industry, provide useful data concerning the attenuation of a huge variety of lithologies and natural materials, the research is typically geared towards borehole and well applications, and may not be as useful for this avenue of research, which is focused on surficial lithologies and materials, including waters.

Reginato et al. (1964) wrote the seminal paper on soil moisture and its effect on gamma attenuation. They utilized a Cesium 137 source that placed under nine representative soil samples from around the US. They concluded that determining the water content of a soil by its level of gamma attenuation was not only possible, but in fact they found that the addition of water had a profound effect on gamma attenuation, possibly equal to the effect of the elemental composition of the soil itself.

The assumption that soil moisture has a great impact on attenuation has been consistently reinforced by subsequent work. Abdel-hady et al. (1999) showed that both soil moisture and the bulk density of the soil could be estimated with respect to the degree of gamma attenuation produced by soils. Abdel-hady et al. saturated a soil sample until ponding occurred, and then performing the analysis. After a further 24 hours had passed, the analysis was repeated. These tests provided the values necessary to determine the effect of water in attenuating gamma radiation, which in turn allowed

calculation of the sample's soil moisture content, water content (cm/depth) and the bulk density of the soil samples.

Shahbazi-Gahrouei et al. (2013) provided a review on the natural background radiation that occurs nearly planetwide, and what causes this background. They note that uranium-238, thorium-232, and potassium-40, as well as their daughter products, play a large role in this radiation. Because those elements are nearly ubiquitous worldwide their emissions are expected in every survey taken of the environment, sample, and waters I take. It is due to this ubiquity that they are also expected to be present in rainwater because the dust and particulates in the atmosphere are often composed of erosional materials that have become airborne.

Ajayi et al. (2015) used a sodium iodide (NaI) scintillating crystal married to the appropriate electronics to create a gamma detector. They collected data at three locations in Niger and found a strong correlation between gamma counts produced by radon and its daughters and the percentage of water in the top soil ***(Figure 2-9)***. These studies concluded that determining that radioactivity of area requires a thorough understanding of the geology. As I noted previously, most personnel conducting subsequent surveys based on results of the research reported in this paper will be non-geologists. As such, recognition of specific lithologies and developing sampling protocols based on my results will be critical for accurate assessments. They also found that water significantly decreased the level of background radiation caused by the radon and its daughters that occurs naturally in the soil. This is indicated by fewer counts of gamma rays per second recorded by their gamma detector. They conclude "that as the water

content of the top soil increases, the radiation level at the air ground interface (ground level) decreases, and this supports the observation…. that the water in the overburden is a major factor governing the movement of radon through it". This conclusion again reinforces the importance of water in producing the attenuation of gamma radiation in soils.

While each of the previous papers have either discussed soils, or soils with added water, none of them have focused on the water type itself. Gainon et al. (2007) found that natural radionuclides of the uranium and thorium series are present at millibecquerel per liter (mBq/l) concentrations up to Becquerels per liter (Bq/l) concentrations in aquifers worldwide. Analysis of Swiss spring water and various European and international groundwaters, determined that radionuclides of uranium and thorium, as well as many of their daughter products, are found in measurable concentrations (Gainon 2007). Two of their analyses are illustrated in ***Figure 2-10***. This includes isotopes of uranium, as well as radon and radium, as they are especially soluble. This work suggests that water itself will emit gamma radiation and paired with the work by Ajayi et al. (2015), suggests that can cause additional gamma emissions from the soil to a greater extent than would occur in a dry soil.

Other researchers have analyzed a range of ground and surface waters, including boreholes, rivers, lakes, streams, and oceans. (Drndarski et al., 1988; Stralberg et al., 2003; Avwiri et al., 2007; Hamzah et al., 2015; IAEA, 2016). They found measurable concentrations of naturally occurring radionuclides, specifically uranium, thorium, and potassium, as well as their daughter products, present in the waters. In addition, many

sampled waters contain radionuclides that derive from industrial processes, energy generation, even nuclear tests, all albeit in lower concentrations than the naturally occurring radionuclides.

Ibikunle et al. (2016) performed direct measurements of rainwater using a gamma spectrometer (NaI) to survey the radionuclides present in the water, either during rainfall, or of the water collected in puddles shortly after a rainfall. Duenas et al. (2010) analyzed monthly samples collected from a rooftop collector. Koike et al. (2013) collected rain samples and performed analysis shortly thereafter, although no time period was specified. Like the three prior studies noted above they also used a gamma spectrometer, however, rather than a using an NaI rystal, they used a HP-Ge crystal, which allowed for increased resolution. This came at a greatly increased cost and necessitated that samples be analyzed in a lab. These studies provide a baseline of what radionuclides will be present in rainwater and may facilitate differentiating rainwater's gamma signature from other gamma signatures that may be present.

Muramatsu et al (1987) analyzed rainwater following the Chernobyl accident. They focused on identifying the presence of radionuclides and the analyses identified both anthropogenic radionuclides and radionuclides that are natural to rainwater. Shortly after the Fukushima reactor meltdown, the U.S. Environmental Protection Agency (EPA) reported finding elevated levels of iodine-131, a product of nuclear fission, in rainwater in Pennsylvania and Massachusetts (EPA, 2011). The levels exceed the maximum contaminant level (MCL) permitted in drinking water (EPA, 2011).

While the above reports detailed the contamination due to a nuclear reactor meltdown, other articles, (Water Research Foundation (WRF), 2011; Ohio EPA, 2015), detail radionuclides in rain water from soils which contain minute concentrations of uranium and thorium. These elements undergo decay to radium, with further decay to radon- a colorless, odorless, and tasteless noble gas (Web 5, 2017). It is suggested that the highest level of radon in rainwater occurs during thunderstorms (Wilkening, 2015) due to the atom's positive electrical charge (Web 4, 2017; Greenfield, 2006). During a rainstorm, and especially during a thunderstorm, the concentrations of these radioisotopes can be high enough to seriously disrupt radiation monitoring at nuclear power plants (Yamazawa, 2008; Web 4, 2017). These disruptions at nuclear power plants suggest that rainfall during gamma radiation surveying may be more difficult since their contribution to background radiation increased significantly both due to the radioisotopes present in the rain and their continued presence in the soil after rainfall.

Application of Previous Work to Research Problem

The radioactive emissions of soils and sediments have been characterized, and the reason for the radioactivity has been explained. The degree of attenuation by these same sediments has been discussed, and the reasons for that attenuation. A few types of water have also been examined for their emissions, and the degree of attenuation by those waters and others has been quantified. The attenuation of a combination of unspecified waters and soils/sediments has been quantified as well, along with an explanation for what leads to that degree of attenuation. Data on the transport of

radionuclides in rainfall after a radioactive disaster has also been gathered, notably by Wheeler (1987) and Puhakka (1990).

While there is a robust research literature and data is available to base the expected emissions of lithologies, the attenuation of radiation caused by water, and the transport of radionuclides in rainwater, there is very little data concerning the impact of those radionuclides on the background emissions at ground level. While Duenas et al. (2010), Kagaku (2013), Koike et al. (2013), and Ibikunle (2016) collected rain samples and analyzed the samples to record their gamma radiation emissions, none of them performed the analysis during the rain event. In addition, they sought to examine the water by itself, not in combination with the locale's soils/sediment, nor did they examine other local waters, and whether those waters show any evidence of the same radionuclides found in the rainfall. Therefore, I intend to examine the emissions of rainfall, and other local waters, in concert with the background emissions of various New Mexico locales and to determine whether rainfall will have a measureable impact on the gamma emissions for those locales, which I expect to be the case.

I began this research with information concerning the attenuation of water, the attenuation of gamma radiation by soils/sediments, the attenuation of water alone and in soils/sediments, and the knowledge that rainwater may contain radionuclides. There is robust data for radioisotope decay and what radioisotopes are present naturally in soils/sediments, as well as their emissions spectrum. From the onset, it was also clear that an enclosure would be necessary to attenuate background radiation to help determine whether there are differences between the surveyed waters. This was from

earlier work that I had performed where I had examined different shielding methods using a natural, low emissions radiation source. However, the size of the enclosure would be limited due to cost and the initial desire for it to be mobile, which meant that the quantity of material that could be surveyed would be limited by the enclosure's dimensions. This would limit the volume of the surveyed materials (soil, sediment, water), which meant that the gamma contribution from those materials may be small which would make it difficult to discern any changes from one survey to the next. Another issue was the lack of information about the concentration of radionuclides in rainwater, groundwater, river water, etc. This included whether radionuclides would be present at all, and if so, would there be changes as a rain event continues or through different periods of the year, or if any change occurs at all. Perhaps the most significant issue that arose during this study was the complete lack of rainfall. Rainfall and its radioactivity are the focus of this study, so a lack of rainfall means the hypothesis cannot be proven or disproven.

Study area

The study focused on eleven locales in New Mexico. ***(Figure 2-11)*** Multiple sites from Los Alamos County were sampled and surveyed due to their proximity to LANL. White Mesa was also sampled and surveyed, and acted as a control, due to the lithology's low background emissions. I sampled near Abiquiu, the top of Sandia Peak, and in the Sandia Foothills. Sampling and surveying were also performed in the Rio Grande Valley in Albuquerque as well as the volcanos to the west of Albuquerque. Other sites that were

sampled and surveyed were locations in the Sangre de Christo Mountains north of Santa Fe, a site near Lamy, and a site near Galisteo.

Study population and Sampling Approach

The study sites were chosen primarily due to their unique nature, which allowed for detailed comparisons between lithologies before and after the introduction of fluids. While each area may be somewhat similar in chemical composition, there is variability. This was ideal for comparing how different rock chemistries interact with water. To capture this potential variability each site was represented by a minimum of three samples if the site appeared to be homogeneous over a large area (e.g. White Mesa). In contrast, sites that have a varied mineralogy (e.g. Sangre de Christo Mountains), and where compositional change can occur over very short distances, required additional sampling and surveying to characterize the locality and to produce results that determine the effect of water on different lithologies. The goal of the sampling strategy was to sample at a density that quantifies the overall geology in each area, producing a result that is representative of large portions of the study area, while also gathering samples in an area that have a different chemistry compared to a sample in its near vicinity. Because of this, the final number of samples varied from one site to the next, depending on whether I felt it was necessary. This was determined after the initial surveying of crushed samples, and meant that additional samples were gathered, rather than fewer.

Sites were selected to capture the range of mineralogies within New Mexico, with an emphasis on characterizing as many different lithologies as possible. This allowed for the exploration of different radioisotopic signatures and their interactions with water. Having a varied sample population may also allow for comparison to sites outside of New Mexico that have similar lithologies without needing a prior characterization of background radiation.

Figure 2-1: FUSRAP sites designated prior or during the most recent waste site evaluation (Vine 2016)

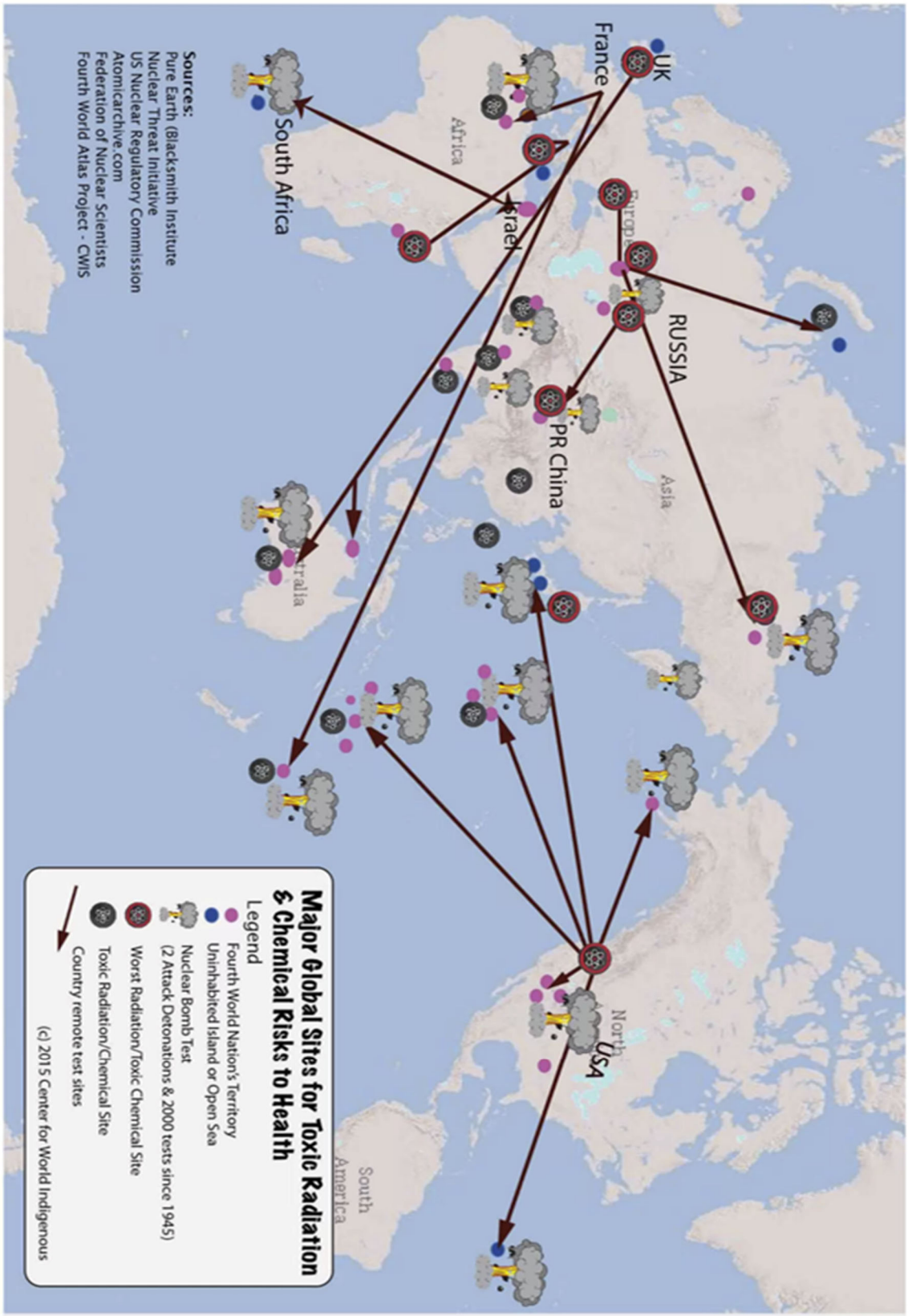


Figure 2-2: Detonation Sites & waste sites throughout the world. (Ryser, 2016)

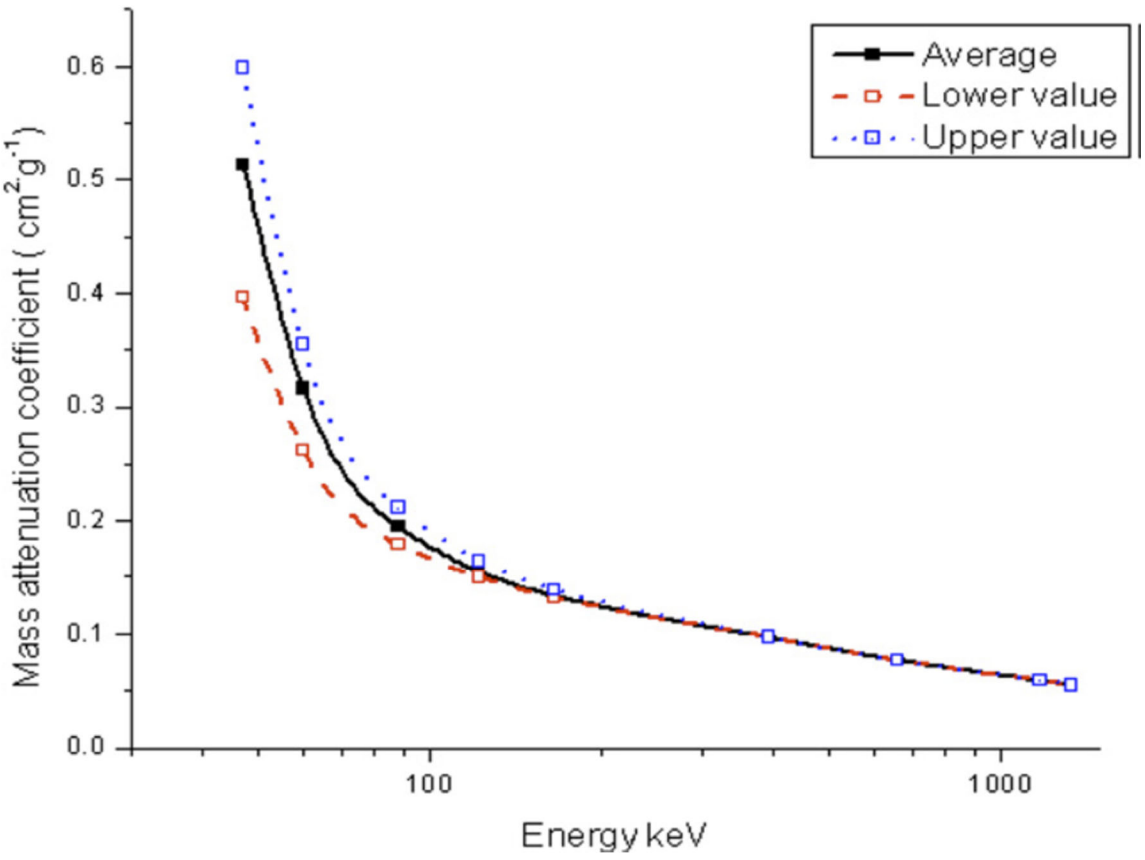


Figure 2-3: Correlation Between Mass Attenuation Coefficient & Energy. (Al Masri, 2012)

Table 2
Mass attenuation coefficient calculated using X-com program.

Mass attenuation coefficient (cm^2 g^{-1})			
Energy keV	Range	Average	STD %
47	0.397–0.599	0.513	7.5
60	0.262–0.355	0.316	5.7
88	0.179–0.212	0.195	2.9
122	0.150–0.164	0.155	1.4
165	0.132–0.139	0.134	0.9
392	0.097–0.097	0.097	0.3
661	0.077–0.077	0.077	0.2
1173	0.059–0.059	0.059	0.2
1332	0.055–0.055	0.055	0.2

Figure 2-4: Mass Attenuation Coefficient Using X-Com Program – (Al Masri, 2012)

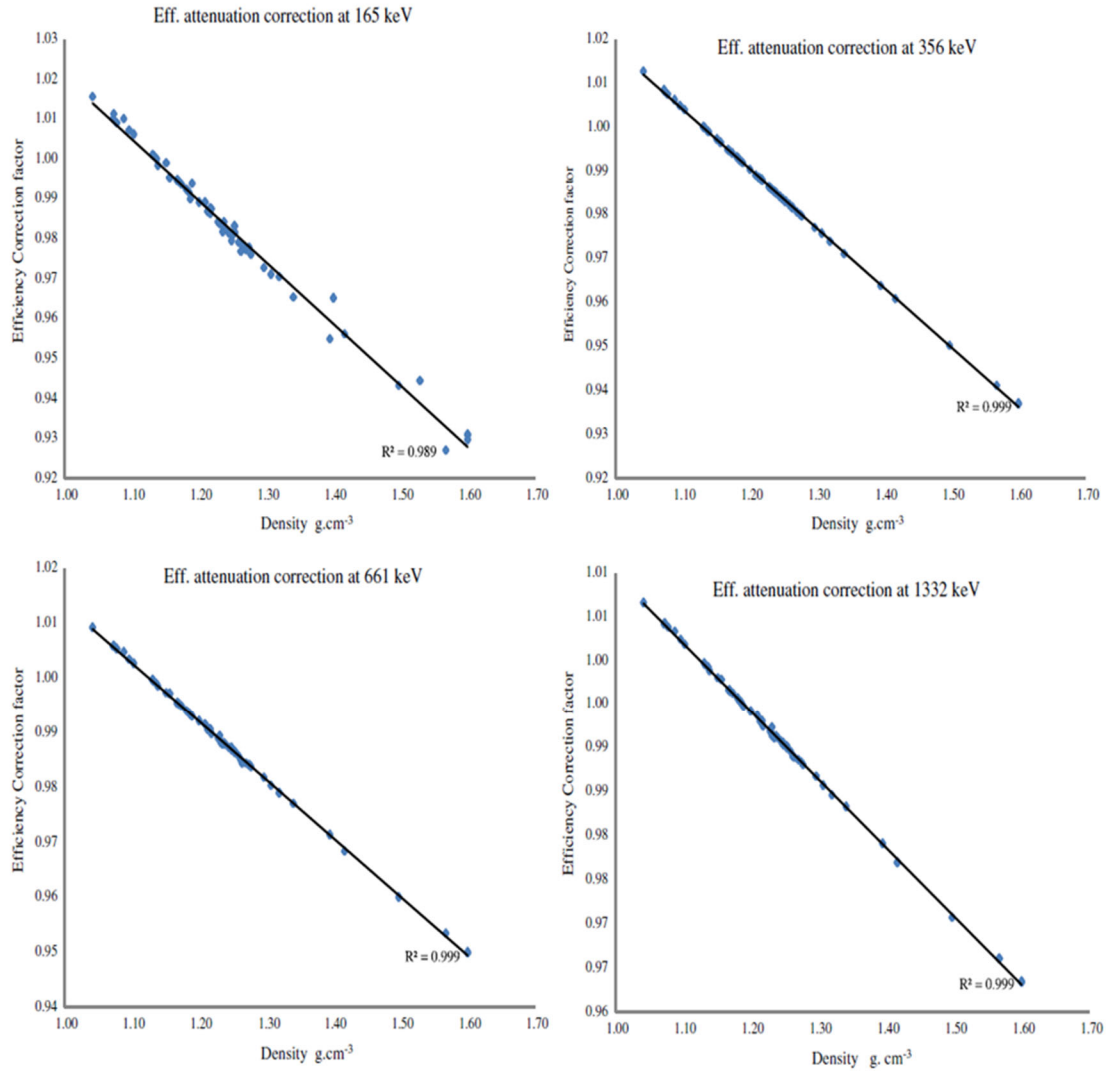


Figure 1-5: Correlation between efficiency correction factor (due to attenuation) and material density – (Al-Masri, 2012)

Table 1

EDXRF analysis results of the dry soil samples studied.

Soil	Chemical components (weight)							
	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	SO_3	CaO	MnO	Others
Sandy	628.0	300.3	34.7	19.3	13.9	2.3	0.3	1.2
Clayey	441.9	328.4	167.4	34.9	17.1	1.6	3.2	5.5

Figure 2-6: *Analysis of Soil Samples – (Costa, 2013).*

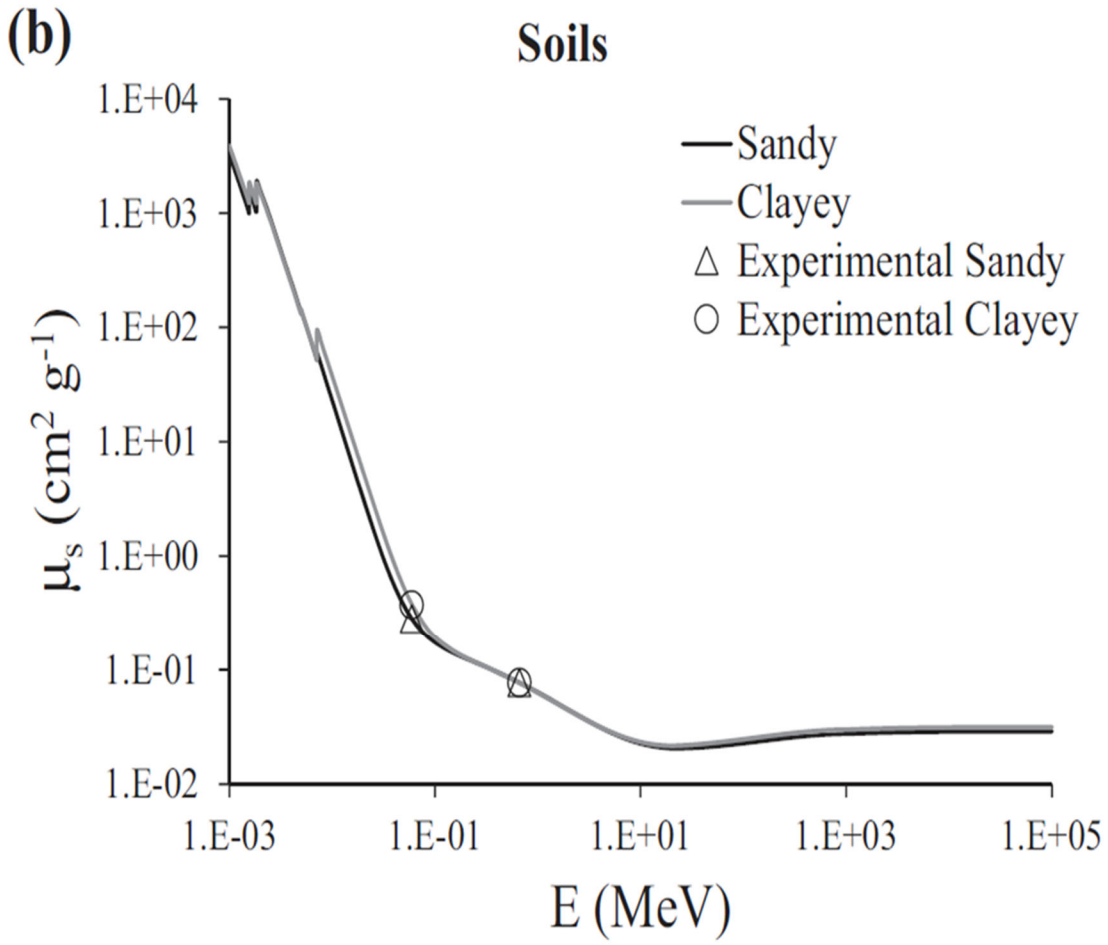
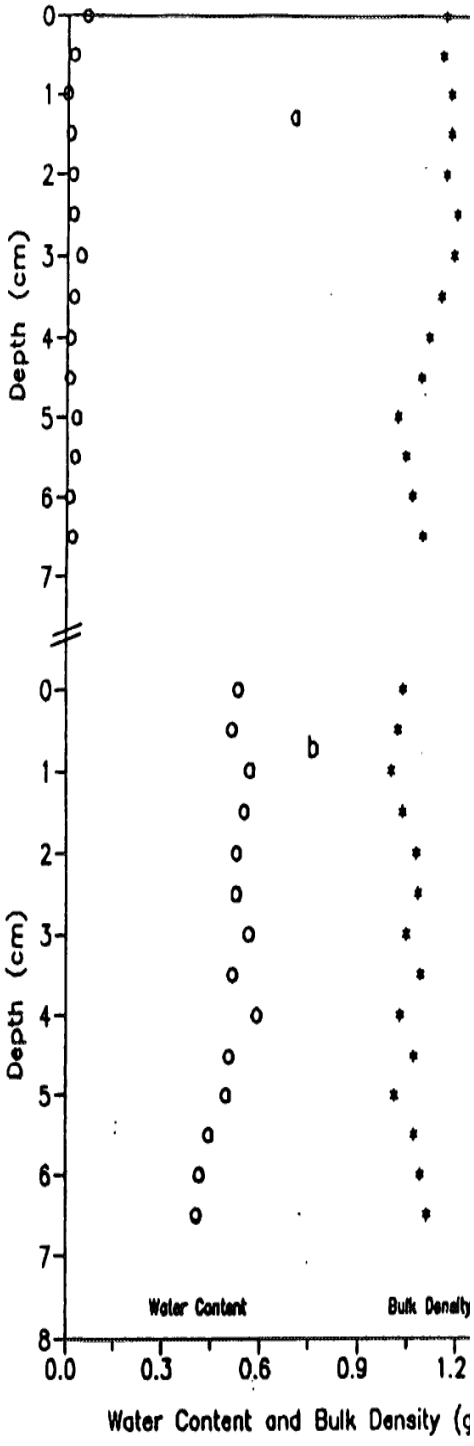
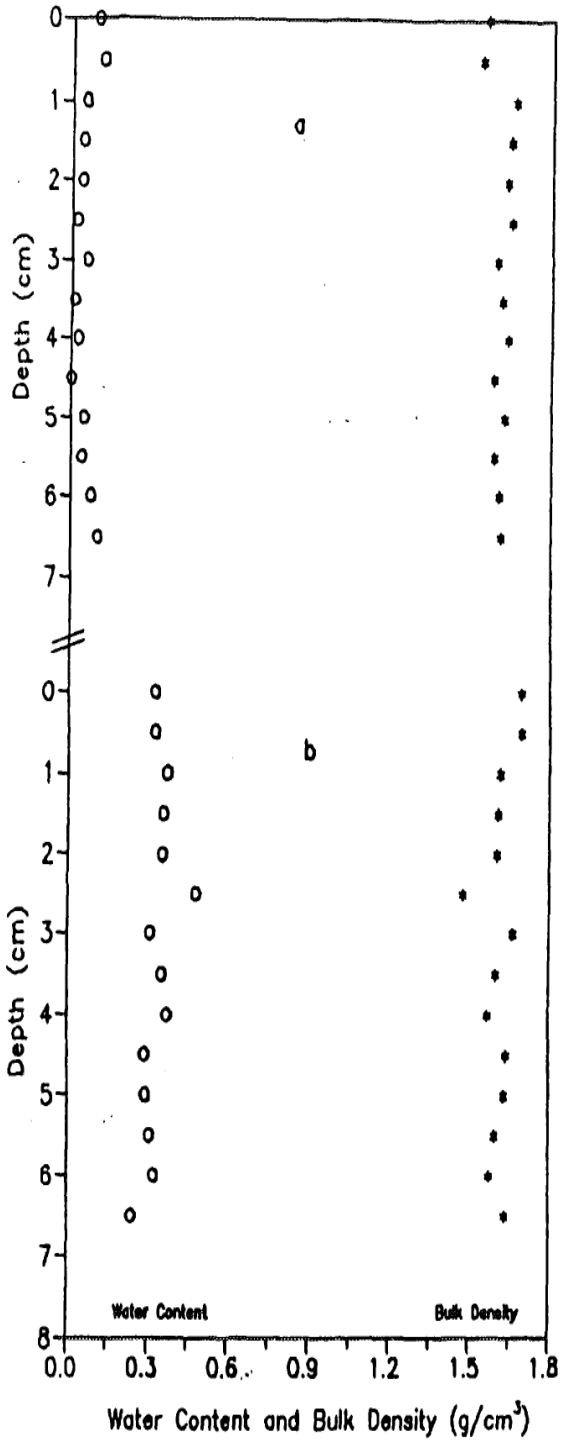


Figure 2-7: *Calculated (solid line) and experimental (symbols) mass attenuation coefficients of Soils – (Costa 2013).*



Water content and Bulk density determined by the dual gamma method for Clayey soil

a) before wetting, b) after wotting



Water content and Bulk density determined by the dual gamma method for
Sandy soil a) before wetting b) after wetting

Figure 2-8: (Ajayi, 2015)

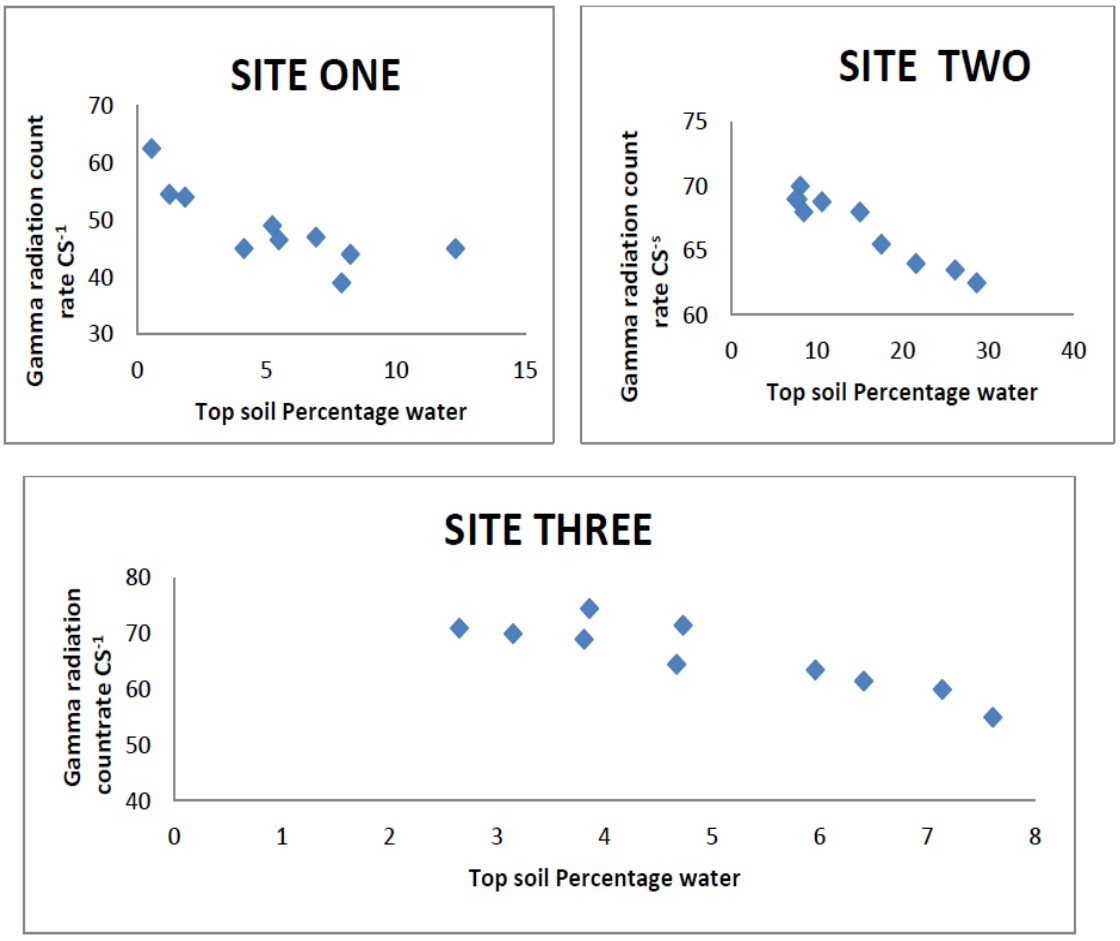


Figure 2-9: The variation of top soil % water with gamma radiation level at the air ground interface at Sites 1, 2 and 3 – (Ajayi, 2015)

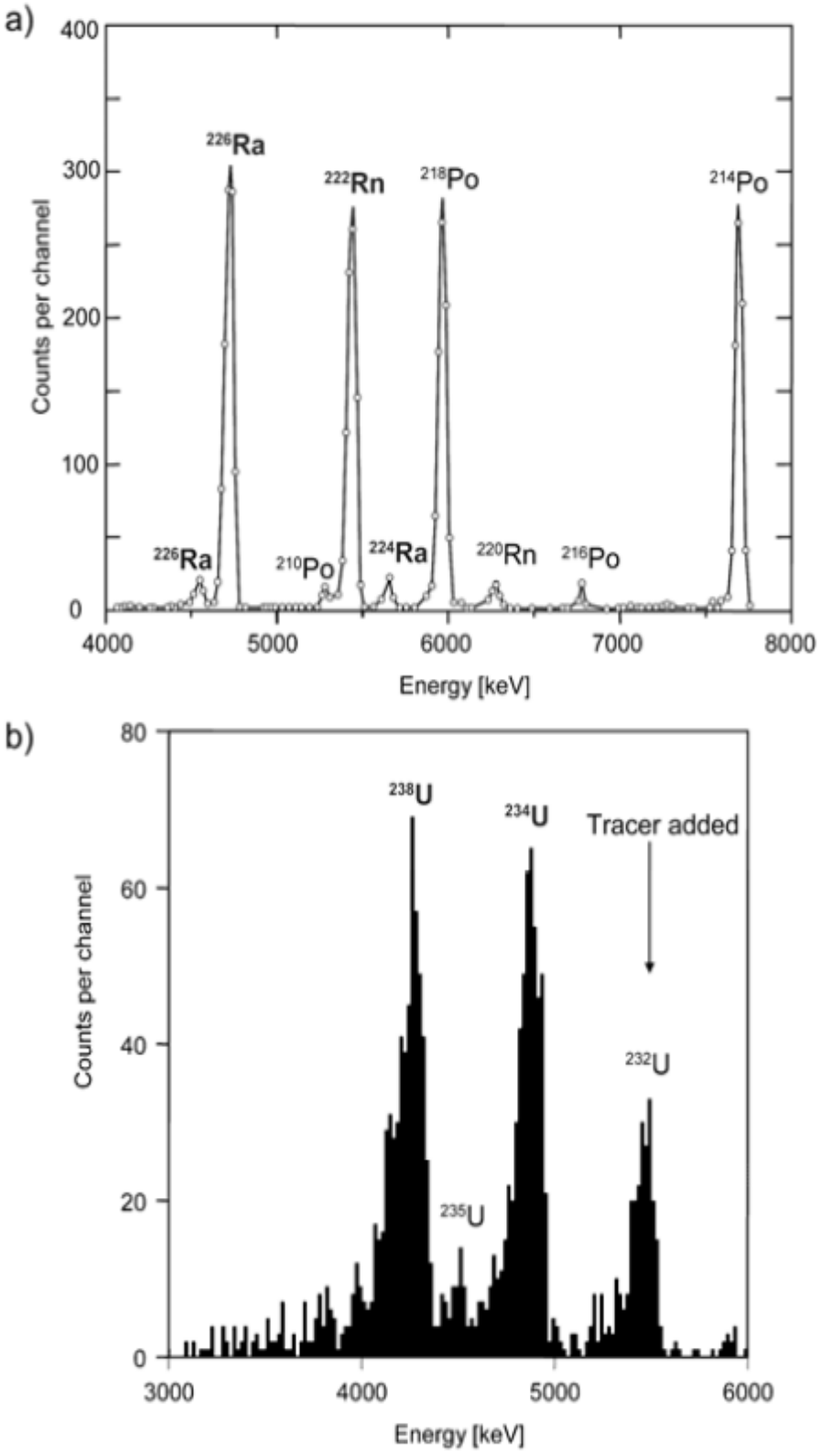


Figure 2-10: a) Mineral water "Pedras Salgadas" (Portugal), approx 1500 mBq/l 226Ra b) mineral water "Aproz Ancienne" (Switzerland), approx. 500 mBq/l. Both spectra measured with a 900mm2 Si alpha detector for 80'000 - (Gainon, 2007)

Figure 2-11: Approximate Locations of All Sampling Sites

Chapter 3: Methodology:

Data Collection Methods

The gamma spectrometer is the foundation of this research and was used to determine gamma emission spectra of the rocks, soils, and waters during the gamma surveys. The spectrometer is composed of a scintillating crystal, a photomultiplier, and associated electronics that allow a computer to read the signal from the crystal and photomultiplier. The spectrometer used in the analyses utilizes a sodium iodide crystal attached to electronics that allow for counting the number and energy of gamma rays that pass through the detector every second. The spectrometer utilized in this research can be seen in ***Figure 3-1.***

There are several factors that determine a gamma spectrometer's detection efficiency, which is defined as the percentage of radiation that a detector detects from the overall yield emitted from the gamma source (Akkurrt et al., 2014). This includes absolute efficiency, intrinsic efficiency, and full-energy peak (FEP) efficiency. Absolute efficiency is detector's ability to recognize a gamma ray that is passing through the crystal. The absolute efficiency of the detector used is ~25% and is typical of a 2x2x2 cubic Sodium Iodide (NaI) crystal (Web 3, 2016). Intrinsic efficiency is the ratio of the number of pulses recorded by the detector to the actual number of gamma rays hitting the detector, and the FEP efficiency is the ability of the detector to accurately determine the full peak of a pulse, rather than simply a pulse of arbitrary size. The combined efficiency of the detector used, at a source distance of 10cm, is 21%. There is also the detector's ability

to discern the difference between two gamma rays' energies, or resolution, and is defined as the "full width at half maximum" (FWHM) (Web 1, 2017). This measure is the width of the gamma ray peak at the middle of the highest point on the peak distribution. It is often expressed as a percentage, by taking the FWHM in keV or MeV, and dividing that by the energy of the gamma ray (Web 3, 2017). For the detector used in this research, the FWHM is 8% at 125 keV, and is 13.5% at 700 keV (Web 3, 2017).

Being able to discern one energy from the other is key to determining what radioisotopes the energies represent, as each radioisotope that emits gamma radiation does so at a specific energy level. The data produced by the detector will be treated as usable and true data that is representative of the actual gamma radiation being emitted by a sediment, water, or source, as the detector conforms to the standards set forth by the IAEA and is in accordance with the guidance set forth by the United States Nuclear Regulatory Commission (Smith et al. 1985; IAEA, 1991; Reilly et al 1991) Since each sample was analyzed by the same detector, calibrated with the same sources, and measured under the same conditions, the efficiency of the detector for this research can be neglected.

Analysis of preliminary surveys suggested that using time-dependent surveys, rather than a count-dependent surveys, would be a better choice. A time-dependent survey analyzes a sample for a given period, resulting in a survey that will vary in the total number of gamma counts, while the period of the survey is constant. A count-dependent survey continues until a predetermined number of gamma rays have been counted by the spectrometer, and as such the period varies. A time-dependent survey

was selected since in practical applications it is the most common method used. Since this research has a strong practical application, it was best to maintain continuity with the bulk of prior research. The selection of a time-dependent survey also makes it easy to compare the relative radioactivity of two sites by comparing the total number of gamma rays recorded after a given period. A higher number of gammas counted in each period between two samples means the sample with the higher count is more radioactive. A time-dependent count also simplifies comparison of differences between surveys of the same sample but with different waters or water quantity. It was also invaluable for comparison of individual surveys taken during a rainfall, because it was possible to observe any time dependent change in emissions for a given interval or versus a prior interval. Lastly, it is simpler to program in a time-dependent survey method that would take surveys at a given time interval, rather than a given count interval, and continue to take surveys for a given period for specified intervals.

To avoid unwanted gamma ray contamination, I had initially intended to build a lead enclosure. Lead was chosen due to its high attenuation of gamma radiation, while being inexpensive and readily available. However, due to restrictions placed on lead use at Los Alamos National Laboratory, I was unable to purchase the inexpensive variety, and the accepted types of lead were prohibitively expensive. I instead chose to utilize a large storage container filled with distilled water and placed the original enclosure inside which was sealed from the surrounding water by a thick plastic bag. Water's attenuation value is significantly lower than steel and lead, but this can be compensated for by

placing much more of it between the enclosure and the outside environment. The large storage container utilized produced this compensation (McAlister, 2018).

To further minimize gamma radiation entering from below the enclosure a 10mm thick piece of steel was affixed to the bottom of the enclosure. Samples were then placed in a 25cm steel tube, which was sealed on the bottom with another 5mm thick plate, which was then put into the enclosure itself. The whole apparatus is illustrated in ***Figure 3-2.*** The samples, where applicable, were crushed and sieved to a uniform grain size, of less than 1mm, which was determined by sieving the crushed materials using a number 18 sieve. The results are illustrated in ***Figure 3-3.*** This was done to eliminate mechanical variability among the samples, which allowed for more consistent coverage of the samples for any given water volume. Prior sample surveys in the enclosure, numerous initial surveys were performed to gather the background emissions of the enclosure and to determine how well the water and steel were attenuating the background radiation at the survey site. These surveys were very important as the emissions spectrum gathered in them will provide a baseline to identify any changes in gamma emissions caused by the introduction of the soil or rocks, and then by the introduction of the various fluids.

At each sample site, at least 3 samples of the rock/soil/sediment were taken if possible. Each of the samples were crushed and sieved, placed in the steel tube, which was then placed in the enclosure. Three surveys were taken of each sample for a period of five minutes (300 seconds). Surveys of the waters were also taken, including distilled, mineral, tap, and hose water. All sediment samples were a standardized volume

(800cm^3), while the volume of water varied between a minimum of 200cm^3 and a maximum of 800cm^3 to determine the attenuation, if any, caused by the water, and to what degree for a given volume of water. This methodology worked well for all sampling of bare rocks, waters, and combinations of rock and water, excepting rainfall, which will be discussed below. In the original proposed research, I proposed introducing a radioactive source to determine variability of attenuation between the different rocks. However, due to variability from survey to survey, and sample relative to background, calculation of attenuation due to the sample was not feasible.

The radiation that was expected in rainfall would be very short lived. Therefore, the most difficult aspect of gathering this data was being in the right place and the right time, as the collection and measurement of rainfall needed to take place over a very short time frame. From January to July, collection and measurement was impossible, due to the near complete lack of rainfall in New Mexico. It was made even more impossible by the change with the enclosure. Carrying a large storage container filled with water, the enclosure, a 10mm x 25cm x 25cm thick piece of steel, a 25cm by 10cm steel tube, along with 60 different rock samples, a large funnel for collecting the rain water, a detector, computer, etc., and deploying and surveying it all quickly enough did not work well, even when there was abundant rainfall.

Thankfully, ingenuity and a trip to Hawaii helped to solve those issues. On May 3rd, 2018, Kilauea volcano began erupting violently, and in more locations, than it had been since it began eruptions in 1983. I was given the opportunity to travel to Hawaii to gather rainfall data for my project, which I did on May 19th. As I was flying, I was unable

to bring any of the apparatus I had been using to take my surveys up to this point, and I was not going to be able to create a gamma-free box like I have done in Los Alamos. I instead chose to do *in-situ* surveys, utilizing the five-gallon plastic buckets common at any hardware store, along with collecting the rainfall that was running off the largest surface I had available; the rental car. The rental car was itself surveyed by rinsing the car following all surveys, to determine if it contributed any measurable gamma radiation. It was not found to do so. ***Figure 3-4*** and ***Figure 3-5*** illustrate the water collection and surveying methods used on the trip. A five-minute survey was taken with the bucket empty, in a location that was either dry, or relatively dry. The bucket was then filled with 2 gallons of rainwater, and a survey was taken while the detector was suspended above the water. This was done at nine rainfall locations, often with multiple surveys per site, with a completely dry site on the western coast of Kona also being surveyed to serve as a control, as the site had been rain-free for nearly three weeks. Two final surveys were taken, one of the waters used to rinse the rental car after I had returned it, and one of the waters used to rinse the car after it had been washed. This was done to determine if the car is inherently radioactive and would contaminate the tested rainwater.

Upon returning from Hawaii, I analyzed my results and determined that it would be appropriate to add an additional method moving forward for analysis of the rainfall. The bucket method, which proved successful in Hawaii would be used going forward, making it much easier to perform *in-situ* surveying, albeit with less control of the variables and possibly more unknowns than I had at the control site. To do so, I utilized

two methods to collect the water, one being a large pop-up shelter, the other being my own vehicle, with the pop-up being utilized whenever possible as it could be kept clean and free from dust and other particulates more easily than the vehicle. For later surveys close to Los Alamos, I also began taking multiple surveys at the same location for extended periods of time to capture the decay and the decreasing concentration of the radioactive particles that the rainwater contained.

Freed from having to transport the enclosure and accoutrement, I decided to make it a permanent fixture in an appropriate space. I would then wait for it to rain where I had the enclosure setup, collect the rainfall, and then add it to the different samples to take my surveys. This made it easy to quickly analyze multiple samples with the rainfall, although it was impossible to analyze all samples in the same rain event within a short timeframe. In addition, having two methods of data collection also enabled me to determine if there were any differences between the *in-situ* surveys and enclosure surveys and attempt to adjust my survey methods accordingly.

While initial storms were brief and intense, raining a few minutes at most, later storms would last for considerably longer, with some having multiple rain events. Three storms had extended periods of rainfall, lasting hours, along with periods of no rainfall, followed by more rainfall. After the first such event, I began taking surveys at five-minute intervals for extended periods of time. In these cases, I would use a five gallon or fifty-gallon drum, as seen in ***Figure 3-6,*** to collect rainfall from a collector that would continue to fill the bucket or drum continuously through the rain event. I used two different collectors, one being the pop-up tent, the other being my home's roof. Both

the bucket and the drum drained at the bottom, typically at slower rate than the rainfall that filled them.

Computational Analysis

The number of surveys of each sample taken during the data gathering phase of the research was three. While additional surveys could be taken, and were at the beginning, due to the nature of gamma detection the values were always slightly different, but well within a statistical average that was representative of the sample's actual gamma emissions, as explained below. Averaging the three surveys together, and then averaging the three surveys from each sample from a given location was shown to, accurately represents the gamma emissions from the samples of sediment and water. In support of this approach I note that each survey represents a Poisson distribution, as radiation is considered a random phenomenon characterized by a Poisson distribution (MIT, 2009). A Poisson distribution is a discrete probability distribution that expresses the probability of a given number of events occurring in a fixed interval of time and/or space if these events occur with a known constant rate and independently of the time since the last event (Haight, 1967). Each survey taken represents an average of the number of gamma rays per energy received every second and even brief surveys typically count at least 20,000 gamma rays before concluding. The combined average of 3 surveys of each of at least 3 samples represents a population of more than 180,000 data points for low emissions lithologies, to more than 300,000 data points for a high emissions lithology and is nearly an hour of survey time. The results can be considered accurate and representative for another reason. The surveys produce a number that

reflects the mean of the number of counts per gamma energy per second. This is also known as the standard error of the mean, or SEM. SEM can be expressed as $N_x = \frac{\sqrt{N}}{\sqrt{T}}$, where $\sqrt{N}$ is the standard deviation of the population, and $\sqrt{T}$ is the number of data points. The standard deviation is the square root of the average rate of the Poisson probability density function that describes the number of radioactive events per time. Therefore, given an adequate period of time, the standard error becomes small, and it becomes likely that small changes are representative of actual change and not the randomness between surveys (Dowell 2018; Wiki, 2018). The chosen survey interval is also a quarter of the half-life of the radionuclides likely to be present in the rainwater and will help in detecting the decay of those expected radionuclides. Due to the large number of gammas rays counted, the surveys themselves are statistically representative of the actual emissions of the sediments or rocks and have a very small statistical uncertainty. To average the survey data, I used a smoothing function, known as the "Average of 30" method, which is similar to a Kernel Smoother, and more specifically, a Kernel Average Smoother. (Web 2, 2017). ***Figure 3-7*** shows the difference between an unsmoothed, relatively low gamma count result, and the lines resulting from different "Average of" methods. The "Average of 30" method was chosen because it results in a graph that has much of the noise from extraneous data removed yet retains recognizable peaks. As those peaks represent emissions from a specific radioisotope, it is desirable to retain them.

Another reason why each sample was surveyed only three times, is that there are at least three additional samples from each site. If there were large variations between the

individual samples from each site or the combination of sample and waters, or even statistically significant small differences, then further surveys may have been necessary, but this was not the case. Each sample's survey was "smoothed" and averaged. Individual "smoothed" surveys were compared with the other "smoothed" surveys taken of the same sample. Qualitatively comparing the surveys allowed an estimate of "measurement uncertainty", which then allowed for the determination of what constitutes important variations in the recorded data. These surveys were graphed and compared to emissions graphs for known radionuclides using the spectrum analysis program PeakEasy (PeakEasy, 2017). If there is a statistically significant disagreement from one survey of a sample to the next survey of a sample with the same conditions, it may have indicated a problem in the data gathering method and would have necessitated a change in that method and resampling. However, even with additional surveying performed outside the original scope and methodology, the data that were collected continued to pass the tests for "good, useable data".

Following data collection and initial statistical analysis, I compared the baseline data, which are the results from surveys of the sediment samples only, to the data collected after a sample has had water added to it. Statistically significant differences were identified and assessed. These 'statistically significant' differences are defined as a difference that falls outside the range of statistical uncertainty. The differences, such as an increase or decrease in the counts per second, were then used to determine the attenuation, if any, of radiation caused by the sediments and the water. Any additional

peaks that were not present on the baseline survey would indicate that a new radionuclide was introduced to the sample.

The statistical analysis of the samples with similar conditions were used to determine the degree of attenuation caused by the water. Once a single site's samples were compared and the degree of attenuation estimated, the data were compared to samples from the same area. Again, an assessment of statistically significant differences was made, and again, the percentage of attenuation, if any, was calculated. Finally, one area's data were compared to another area's data, which was instrumental in determining if water consistently attenuates radiation regardless of what the sediment chemistry may be, and what percentage of attenuation occurred, if any. The data can also be used to determine what water has the most effect on the attenuation of radiation.

Mechanisms to Assure the Quality of the Study

To assure quality results, the sampling method and study population must be representative of the study area, while avoiding sampling bias. While I initially believed that more than 3 surveys of each representative sample would be needed to help minimize sampling error and facilitate identifying statistically significant differences, this proved not be necessary, as noted above. To minimize sampling bias, I specified areas that samples should be taken from, while another student performed the actual sample gathering with each sample assigned a number, rather than a site location name or geologic identifier.

To avoid instrument bias, the detector was calibrated to known gamma sources, under conditions similar to where and how it will be used, assuring that the collected spectrums are representative of the actual emitted spectrums. In this research, the high-quality data that gathered was somewhat dependent on the number of samples and the number of surveys taken, as well as appropriate computational analysis of the samples. Maintaining the same volume of the samples and waters, same container type for samples, sample gathering and preparation, etc., proved crucial to gathering data that could then be compared. The most important factor in gathering comparable data was maintaining consistency and repeatability.

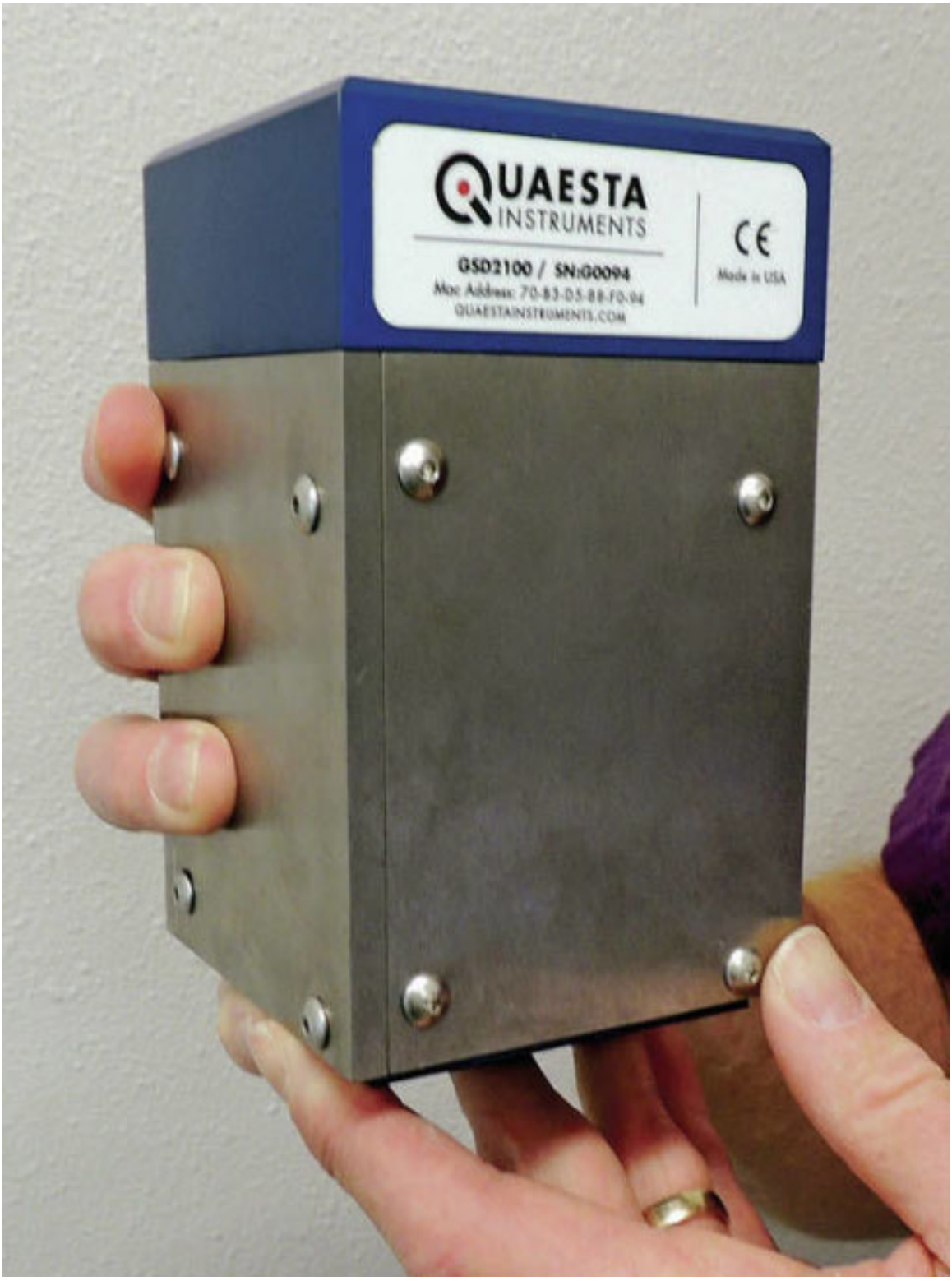


Figure 3-3: Quaesta Instruments Gamma Spectrometer

Figure 3-4: 80/20 Enclosure w/o Water Drum

Figure 5-3: Crushed Granite Sieved Through #16

Figure 3-6: Rain Collection and Surveying technique during heightened Kilauea volcano eruptions

Figure 3-7: Gamma spectrometer and accoutrement used for surveying of Kona basalt and rainfall during heightened Kilauea eruptions

Figure 3-6: Rain Collection Drum

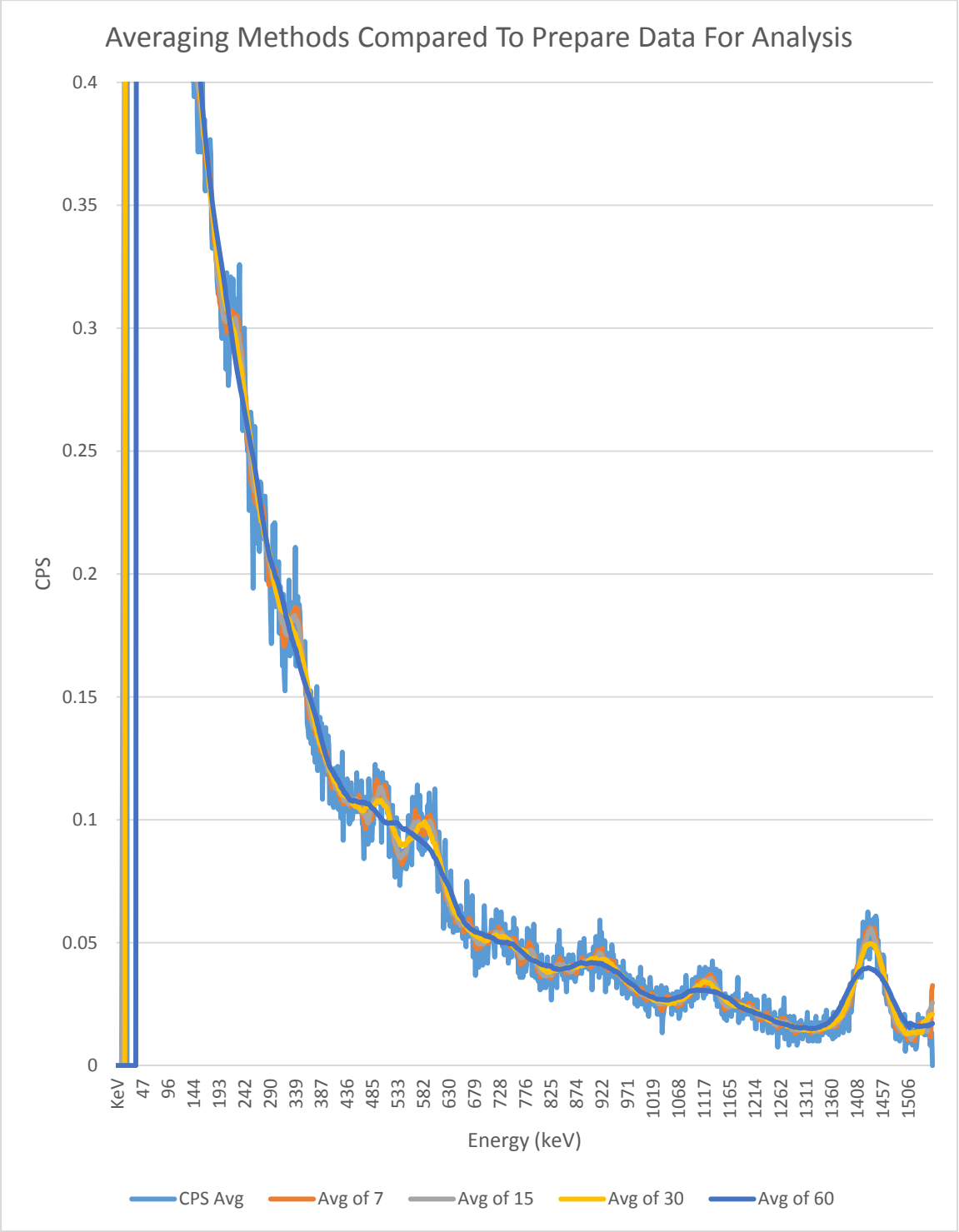


Figure 3-7: Examples of Smoothing Methods - Average of 7 Method results in distinguishable peaks without elimination of valuable data. Average of 30 Method results in distinguishable peaks while eliminating extraneous data. Average of 15 or 60 are not adequate, and either do not do enough, or do too much.

Chapter 4: Results

Introduction

This section documents the results of the surveys, including those measured within the experimental apparatus and in-situ surveys measured in the field. The results are grouped by sample site, with each sample site being represented by a graph that shows the differences that result from different survey and sampling conditions. Each sample line on a graph is the average of three surveys of three samples, or nine total surveys, except for the samples combined with rainfall, which are an average of three surveys performed on a chosen sample at each sample site. This section begins results from measurements in the experimental apparatus and its environment. This is followed by the results of the surveys from each of the sample sites including the samples, the samples combined with the different waters, and finally the samples combined with rainwater. Finally, the results of the *in-situ* surveys are presented. These include rainwater data from Hawaii, followed by rainwater data from surveys taken in New Mexico.

Enclosure Surveys

The enclosure apparatus consists of a cube built with an aluminum channel called 80/20. Affixed to the bottom of the enclosure is a 10mm thick piece of steel. The samples were placed inside a steel tube sealed with a steel plug on the bottom. To prevent water from entering the enclosure, it was placed in a heavy-duty plastic bag

that was then secured around the enclosure. The bagged enclosure was then placed in a large storage drum, which was filled with distilled water (***Figure 3-2***)

Figure 4-1 incudes the initial background survey data taken of the enclosure environment, the survey of the enclosure, and finally the whole experimental apparatus, including the water filled drum. The background is the gamma emissions present in the environment in which the enclosure will be placed. The background emissions in the survey area were averaged from five surveys, resulting in a cumulative count per second (CPS) of 128. The interior of the enclosure itself, consisting of the aluminum 80/20, the steel plate, the steel sample tube, and the plastic bag was surveyed, resulting in a cumulative 98 CPS. This was done by placing the detector on top of the enclosure and steel sample tube and performing five surveys in this manner. Finally, the enclosure was placed in the container, which was then filled with distilled water, with results showing a background emission of a cumulative CPS of about 65 CPS. The degree of attenuation is greater at gamma energies less than ~650 keV. After that, the degree of attenuation decreases significantly, until the spectral lines are relatively equal in CPS. There are four recognizable peaks in the averaged survey data, at 100, 490, 605, and 1460 keV. These peaks, as well as all others, correspond to gamma rays emitted by naturally occurring radionuclides, will explained further in the paper.

The first surveys that were taken following the survey of the enclosure apparatus were of the various waters, including tap water, Rio Grande water sampled near the Montano Bridge, and distilled/spring water that originated from a source in Arizona that was purchased from a grocery store. The source's location was not specified further. ***Figure***

4-2 shows the averaged emission spectrum of three surveys of each water. There are five emission spectrum lines, representing each of the waters as compared to the enclosure environment. Each line is the averaged gamma ray counts for a given energy (keV). There are three distinct peaks in this survey, at 100, 490, and around 1460 keV. There is also the suggestion of a peak at 600 keV. There is a slight difference in the energy of the peak at 1460 keV. ***Figure 4-3*** shows the average of the total CPS for each of the given waters. The enclosure has the highest CPS of nearly 0.0612 CPS, while the waters have lower CPS, the lowest being Springwater, with a CPS of 0.0605.

Figure 4-4 through ***Figure 4-14*** follow the same outline as ***Figure 4-1*** and ***Figure 4-2***. Each figure represents one sample site and the averaged survey values of all samples taken from that location, relative to the enclosure's values. Each line represents the averaged CPS per gamma energy for an experimental condition. This includes the samples with the waters and rainwater introduced. Every figure has four recognizable peaks, at about 100, 490, 600, and 1460. There is also a downward trend of CPS evident in all figures, including ***Figure 4-1*** and ***Figure 4-2***, from left to right. With the introduction of the sample to the enclosure, there is an increase in CPS that is dependent on the sample. When the White Mesa gypsum is added, there is an average 1 percent increase in CPS, while the Abiquiu tuff increases the average CPS by 7 percent. The other samples lie between the 1 percent average increase of the gypsum and the 7 percent average increase of the tuff.

With the addition of water to the samples, there is a small decrease in CPS that varies with energy, and an overall decrease in average CPS. The decreases were as small as a

tenth of a percent, to slightly more than 1%. The average decrease across all samples was slightly more than 0.5%. The decreases were not consistent from sample to sample, nor water to water. With the addition of rainwater to all samples, there is an increase relative to the bare sample, and the samples with the other waters. The increase varies with energy and sample and decreases as the rainwater ages after collection. The increase was as much as 9% greater than the bare sample, as seen with the White Mesa gypsum samples, and as little as 3% more than a bare sample, as seen with several samples. ***Figure 4-15*** provides the average CPS of the samples relative to the same sample site and to other sample locations, including the bare enclosure.

For all figures, the CPS varies depending on the energy of the gamma ray. With the introduction of the enclosure, there is a decrease in CPS at all energy levels, with a larger relative decrease at the energy levels below roughly 650 keV. There is also a decrease in average CPS. When the enclosure is placed in the water-filled container, there is another decrease in energy, like what is seen when the enclosure itself is surveyed versus the bare background. Again, there is a larger decrease in lower energy gamma ray CPS counts compared to higher energy gamma rays, with a decrease in overall CPS. When the samples are introduced, there is an increase in average CPS and CPS at any given energy level. When the waters are surveyed with the sample, there is a small decrease in average CPS and generally a decrease in CPS at any given energy level. When the rainwater samples are combined with the samples, there is a increase in average CPS, and an increase in the CPS for each energy level. This increase decreases as the rainwater ages.

In-Situ Surveys

The first *in-situ* surveys were conducted in Hawaii. The surveys began on the Kona, known as the Big Island (***Figure 4-16, 4-17, 4-18)***. Following the trip to Hawaii, and the commencement of rainfall in New Mexico, *in-situ* surveys were begun in New Mexico. Surveys were taken near Hyde Park Rd in the Sangre de Christos, multiple locations of the Bandelier tuff in Los Alamos County, and in the gypsum of White Mesa. The conditions of the surveys varied, dependent on conditions such as quantity of rainfall and the amount of lightning striking nearby. In general, more than 3 surveys were taken, and in two locations in Los Alamos, the surveys were continuously running for more than 8 hours.

Surveys in Hawaii began on May 19, 2018, in the city of Kona. To perform the studies, two five-gallon buckets were used, along with small metal cans to hold the gamma spectrometer at a given distance from the ground and above the rainfall collected in the bucket. The rainfall was collected off the car into small containers, and then transferred into the larger 5-gallon bucket, until there were roughly two gallons of water in the bucket. The car was utilized because it was an effective large surface to capture and channel the water into the metal receptacles. The surveys were then performed, each with a period of 300 seconds. ***Figure 3-4*** and ***Figure 3-5*** show the tools and methods used to perform this work.

There were ten survey locations, and the first location chosen was a national historical park called Kaloko-Honokohau. The location had experienced multiple lava flows in the

past, and two of the five flows were surveyed. The location had also not experienced any rainfall for more than two weeks, assuring a dry, relatively moisture free survey location. Following the first survey, a circuit of the island was done, with the survey locations being decided upon based on the quantity of rainfall occurring at the time. Surveys were performed in the middle of the island along the Saddle Road, continued down the eastern coast, and finished near the most southerly tip of the island. Rainfall that was adequate for collection began near the Ka'Ohe Game Management Area and continued into Hilo. Four stops were made before Hilo, with rainfall collected and surveyed, as well as a dry site surveyed near the rainfall collection site. The dry sites were the location I had parked the vehicle and remained dry due to the vehicle acting as an umbrella. After Hilo, there were five sample sites along the Hawaii Belt Road. The first site was near Keaau, another just before Hawai'i Volcanoes National Park, another in the middle of the park, one near the very end of the park, and the final location just after Ocean View near the southernmost tip of the island. The approximate locations of the survey are identified in ***Figure 4-16***.

The surveys of Hawaii are presented in two figures, ***Figure 4-17*** and ***Figure 4-18***. ***Figure 4-17*** presents the results in a similar manner to the figures with the results from the enclosure surveys. The lines represent the average CPS per gamma ray energy after being averaged with a moving average of 30 data points. The nine survey site locations are combined into four sites, each with a dry and wet graphed line. This is true for all locations excepting the surveys performed at the two locations at Kaloko Honokohau, those surveys are presented individually. Every site surveyed exhibits several

recognizable peaks at around 225, 490, 600, and 1460 keV. These peaks are the result from the gamma emissions of the naturally occurring radionuclides, including uranium, potassium, thorium. The locations after Hilo but before the eruption sites, grouped as the Before Volcano line, exhibits peaks at about 1275, 1325, and 1530 keV, as well as most of the other peaks from the other surveys. While not obvious in ***Figure 4-17***, each location saw an increase in average cumulative CPS and CPS per gamma energy with the inclusion of rainfall. This is more obvious in ***Figure 4-18***, which shows the average CPS of the wet/dry surveys relative to each other with the average CPS of all wet/dry surveys. Each survey site's dry CPS was within 1% of the averaged CPS of all the survey sites' dry surveys. The wet surveys averaged CPS change relative to average varied from a 3% decrease to a 27% increase. All wet surveys increased relative to the dry survey at the same site. The smallest increase was 1%, recorded on the north-eastern side of Saddle Road, while the largest increase was 12%, recorded near Ocean Point near the south-western tip of the island.

With the commencement of the monsoon season in New Mexico, and using the methods from Hawaii, multiple *in-situ* surveys were performed at various locales. The surveys were performed in tuff in Los Alamos. The first surveys were taken in an area that will be referred to as "Grill Tuff', the results of which are seen in ***Figure 4-19***. The surveys were taken in while the lithology was dry, and when the lithology was saturated with 20 gallons of distilled water. The third line represents an average of four surveys, or 20 minutes of survey time, starting with the beginning of a heavy rain event. The lines represent, as with the other figures, average CPS for a given gamma energy. There are

four peaks obvious in the graphed line, at about 490, 575, 900, 1100, and 1440. There is a decrease in average CPS with the introduction of the distilled water, and an increase with the introduction of the rainfall. The increase and decrease are greater at lower energies (below 650) and are negligible at higher energies. The overall change in CPS with the introduction of the distilled water is 1.5%, while the average increase in CPS with the introduction of rainfall is 5% ***(Figure 4-20)***.

Further analysis in Los Alamos was performed near my residence in another outcrop of Bandelier tuff, and will be referred to as "House Tuff". This enabled an extended period of analysis prior to, and during, a rainfall event. The results are presented in ***Figure 4-21*** and ***Figure 4-22. Figure 4-21*** is again a graph showing the average CPS per gamma energy for the test conditions. This includes the dry survey area, the survey area saturated with distilled water, and the survey area during a rainfall event surveyed in 5-minute intervals for 75 minutes. There are noticeable peaks in all lines, at around 460, 575, 900, 1180, and 1460 keV. The peaks are less obvious in the line representing the distilled water experimental condition. There is a large decrease in gamma rays with the introduction of the distilled water into the drum. There is an increase with the first rainfall survey, and the increase persists throughout the 75-minute interval. During the testing interval, the energy at which the peaks are located changes, most noticeably with the peak that changes from 1470 to 1400. Per ***Figure 4-22***, there is a 30% reduction in average cumulative CPS with the introduction of the distilled water, and an increase of 15% during the first 5-minute survey of the rainfall, relative to background. The

increase decrease as the surveying period continues, until it is only 9% during the last 5-minute survey period.

Other surveys include an analysis of a location adjacent to Hyde Park Road in the Sangre de Christo mountains (SdC), and surveys taken in the White Mesa gypsum. ***Figure 4-23*** presents the results of the SdC surveys, while ***Figure 4.24*** presents the results from the White Mesa gypsum analysis. Both the rainfall and the dry condition surveys have several distinct peaks at around 200, 460, 550, 864, 1050, and 1350 keV. There is also an increase in the average CPS per gamma energy with the introduction of rainfall to survey site. With the introduction of the rainfall, there is an additional distinct peak at 100 keV, while there is just a suggestion of the peak at the same energy with the dry conditions. There is a 10% increase in the average cumulative CPS with the introduction of rainfall to the SDC survey site. During the White Mesa rain survey, cumulative average CPS increases by 7%, with a relatively uniform increase in CPS throughout the averaged emissions spectrum. There were three distinct peaks, at about 150, 460, and 1375 keV. There is a trough at 960 keV.

Summary of Surveys

All surveys of the dry materials exhibited peaks at similar keV. A few of the surveys saw peaks at other energy levels. With the introduction of the non-rainwater waters, the average CPS per gamma energy decreased, while with the introduction of rainwater the average CPS per gamma energy increased. The decrease with the introduction of different waters was relatively uniform, while the increase with the introduction of

rainfall was not uniform and varied from 3% up to 15% or more. Because the surveys are all presented in CPS, the average CPS per gamma energy is relatively uniform. The surveys also represent more than 2 million data points and vary per survey from 10,000 data points to more than 30,000 data points. Variability from survey to survey in total CPS given similar conditions is less than 1%, except for surveys involving rainwater. Surveys involving rainwaters and similar conditions varied in total CPS by as much as 30%, and as little as 3%.

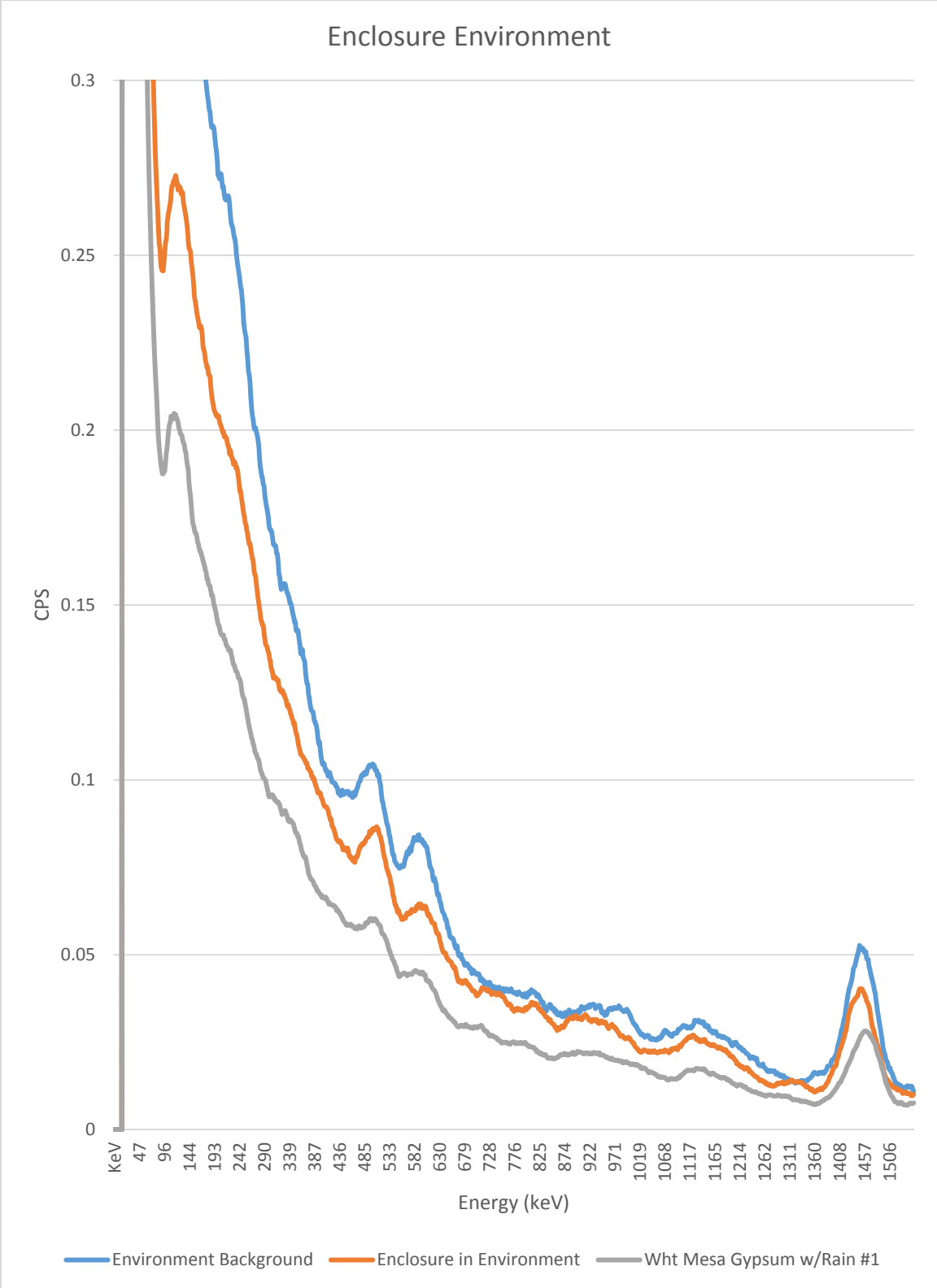


Figure 4-1a: Background Emissions of Environment, Enclosure, and Enclosure in Water-Filled Container

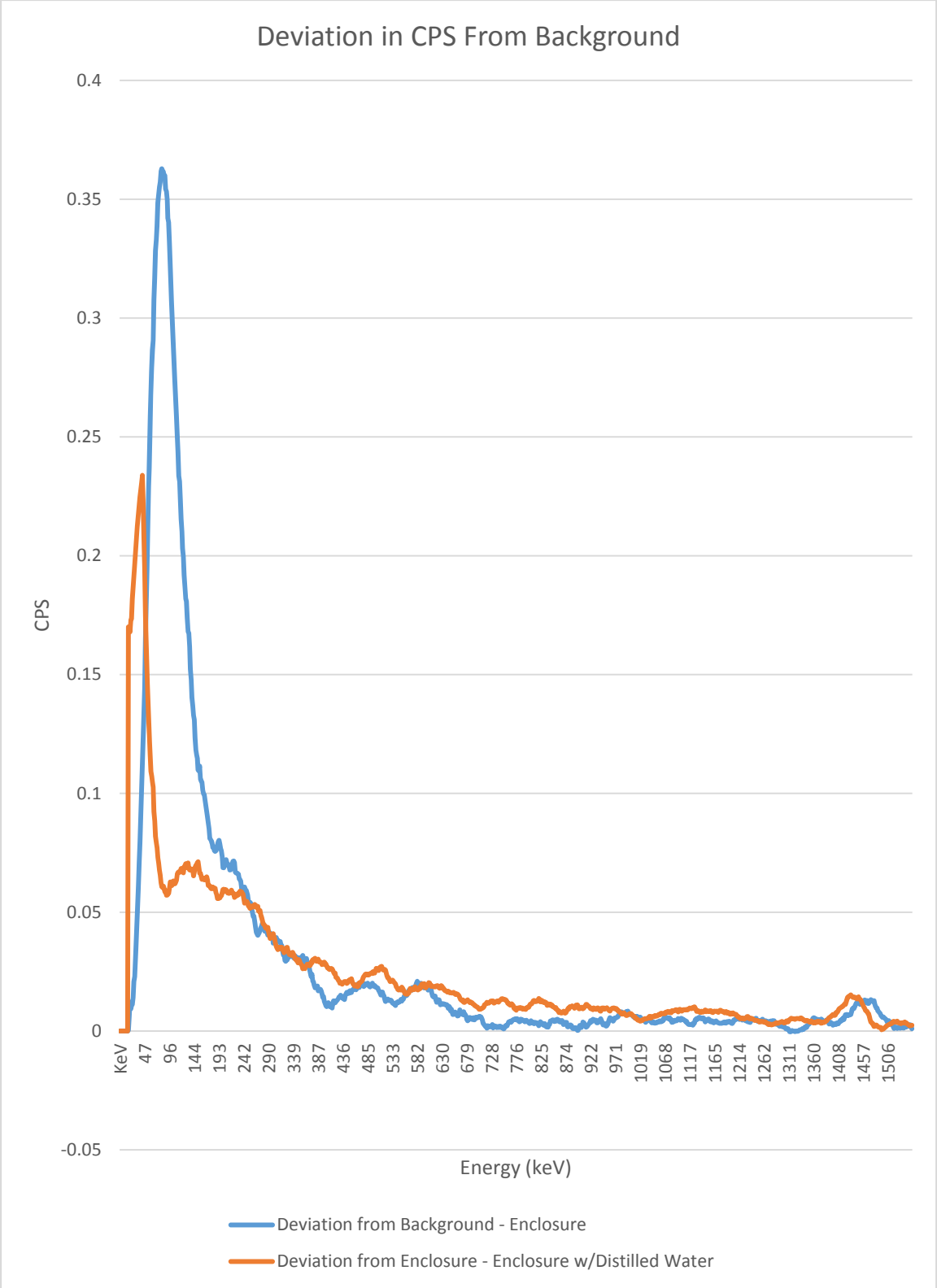


Figure 4-1b: Deviation in CPS from Background Emissions of Environment with Enclosure, and with Enclosure in Water-Filled Container

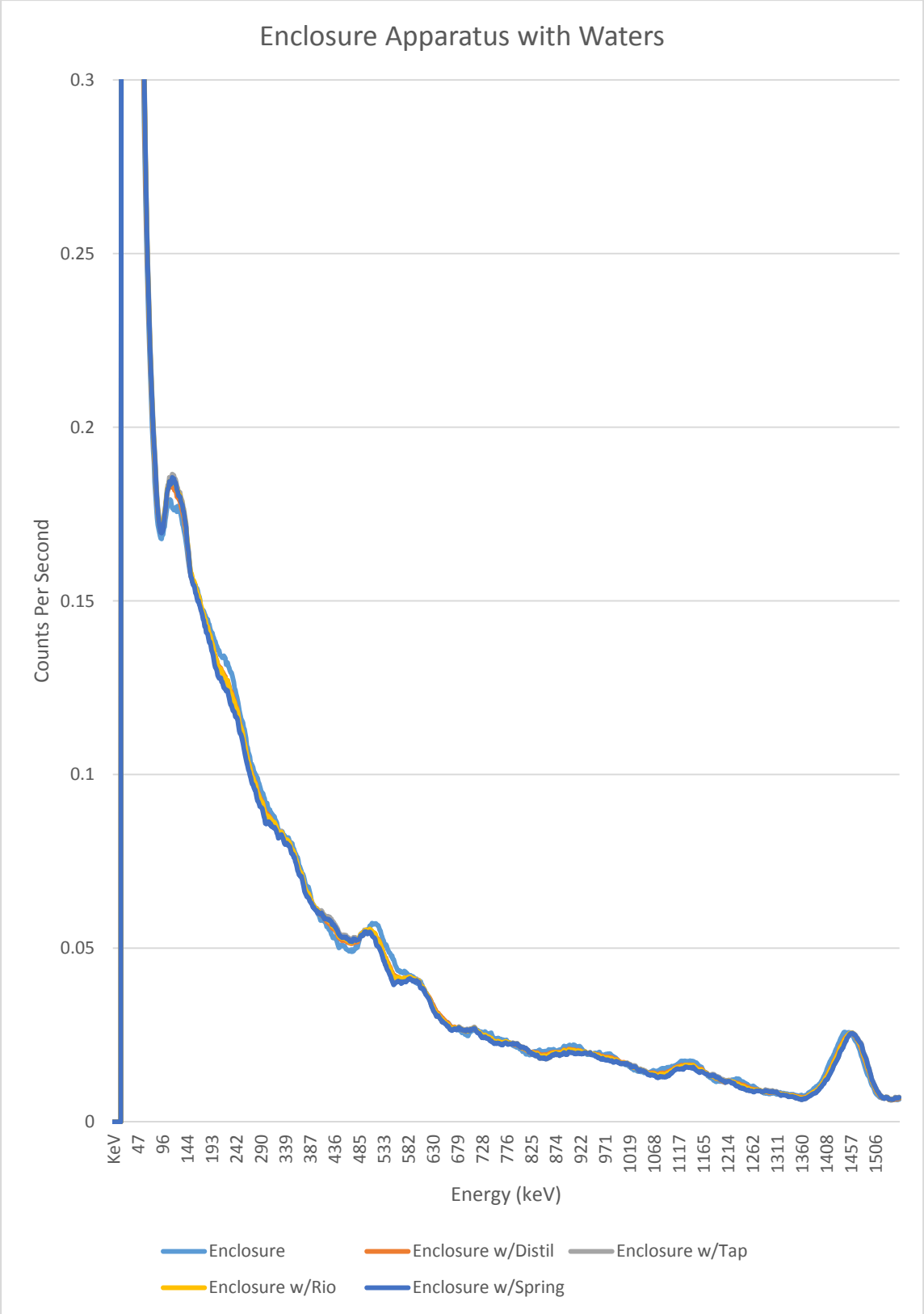


Figure 4-2a: Counts per Second of Various Waters in Enclosure Apparatus

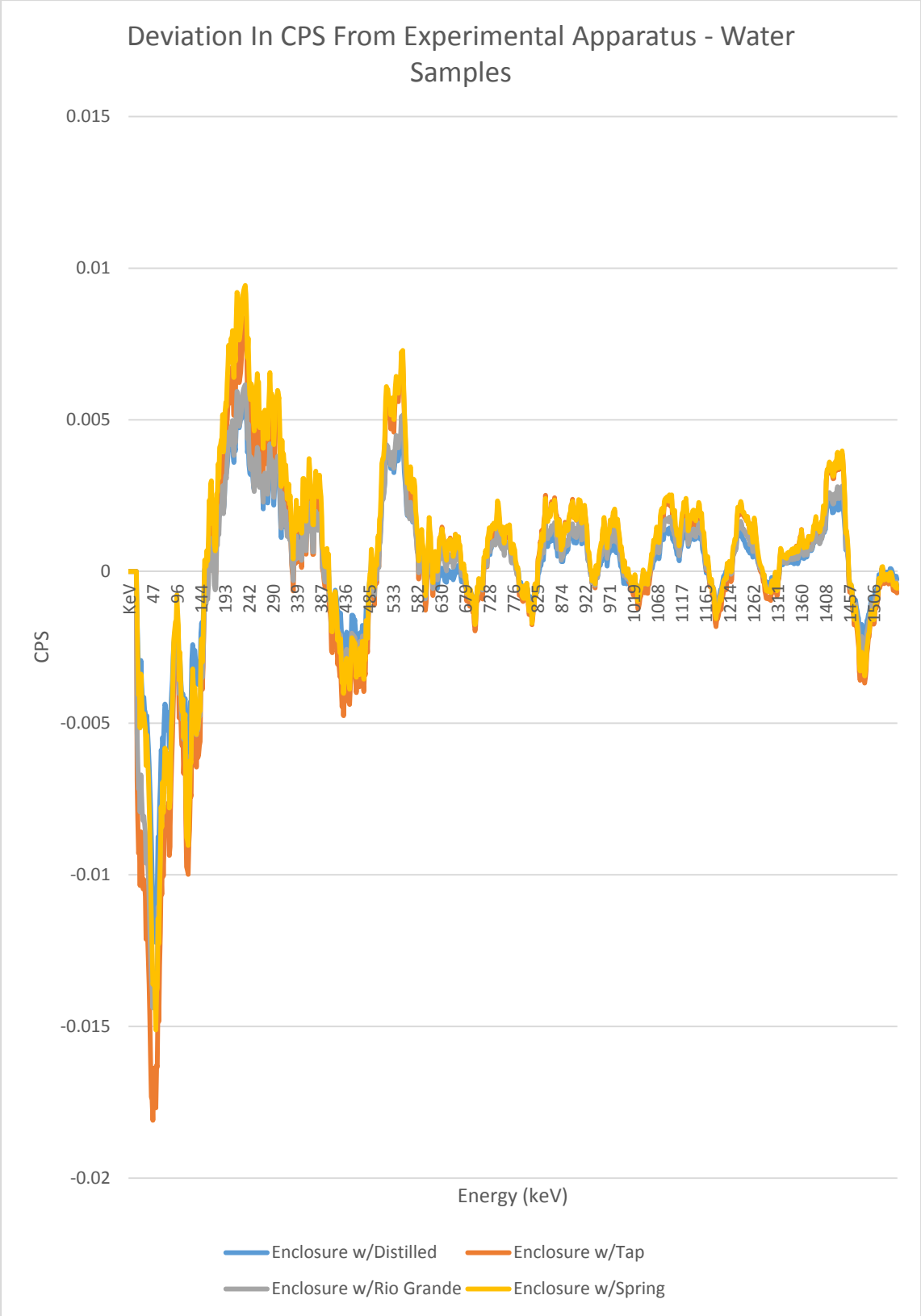


Figure 4-2b: *Deviation in CPS from Experimental Apparatus with Introduction of Water Samples into Survey Chamber*

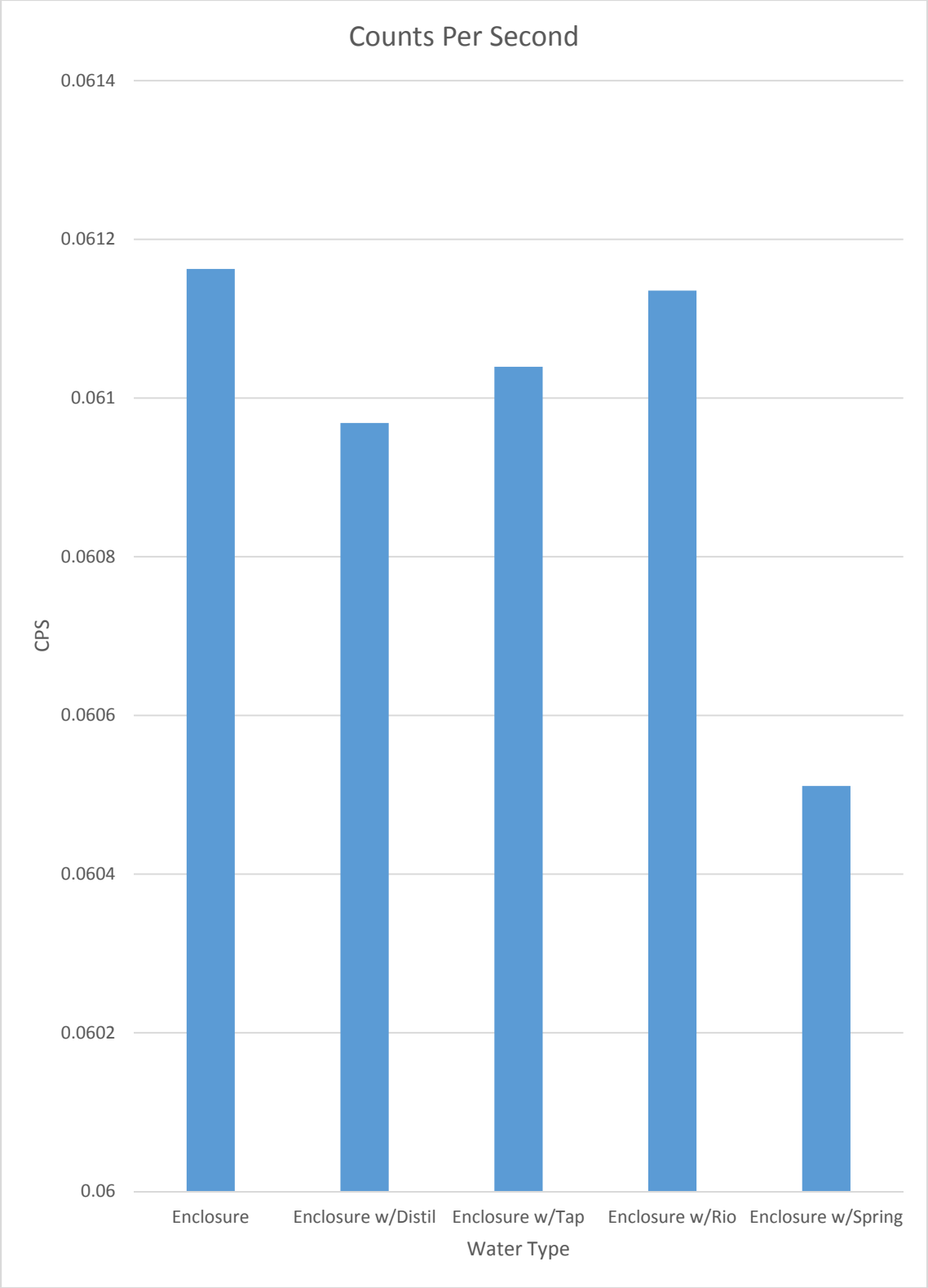


Figure 4-3: Difference in Average Counts per Second of the Waters in the Enclosure Apparatus

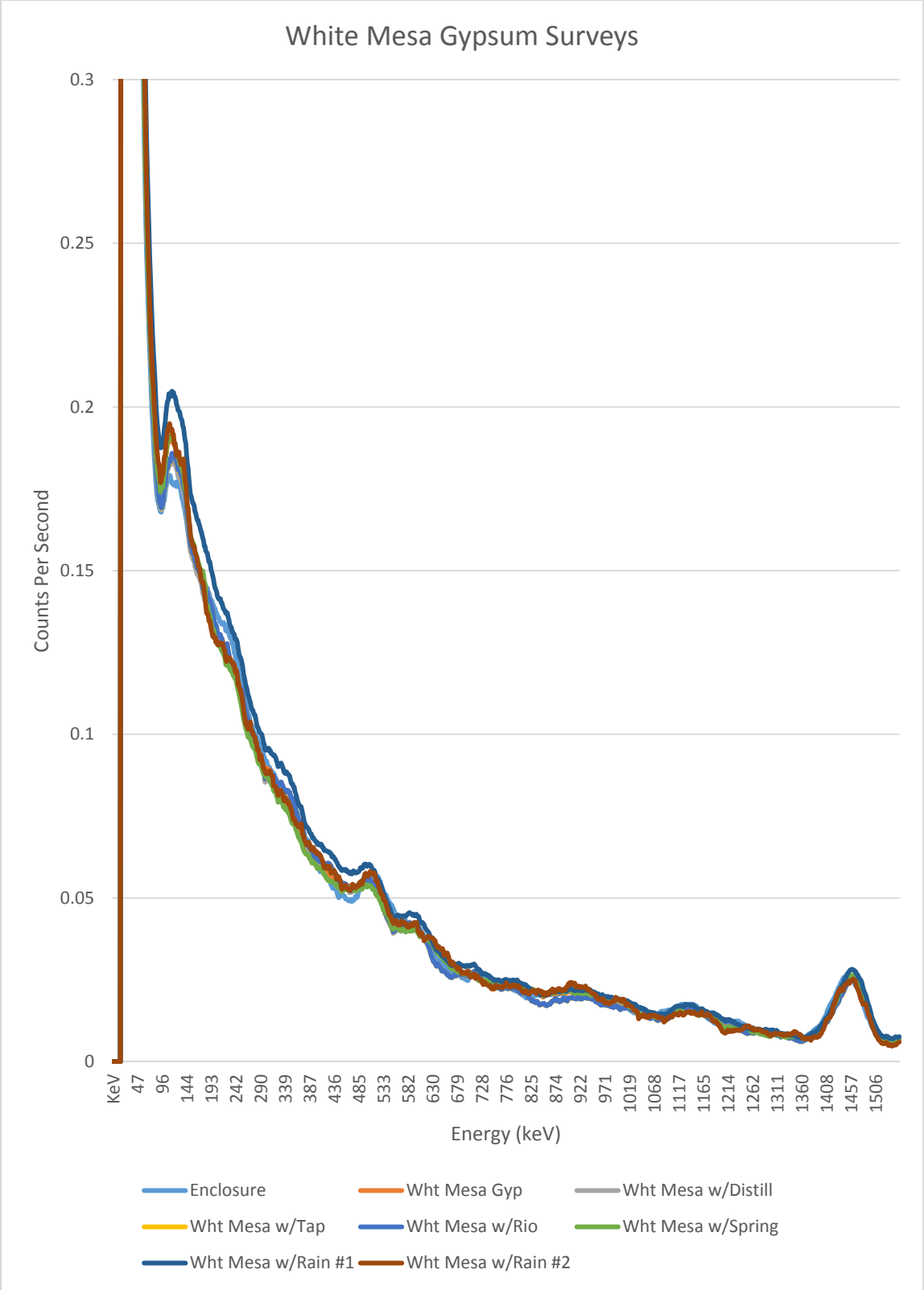


Figure 4-4a: White Mesa Gypsum Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

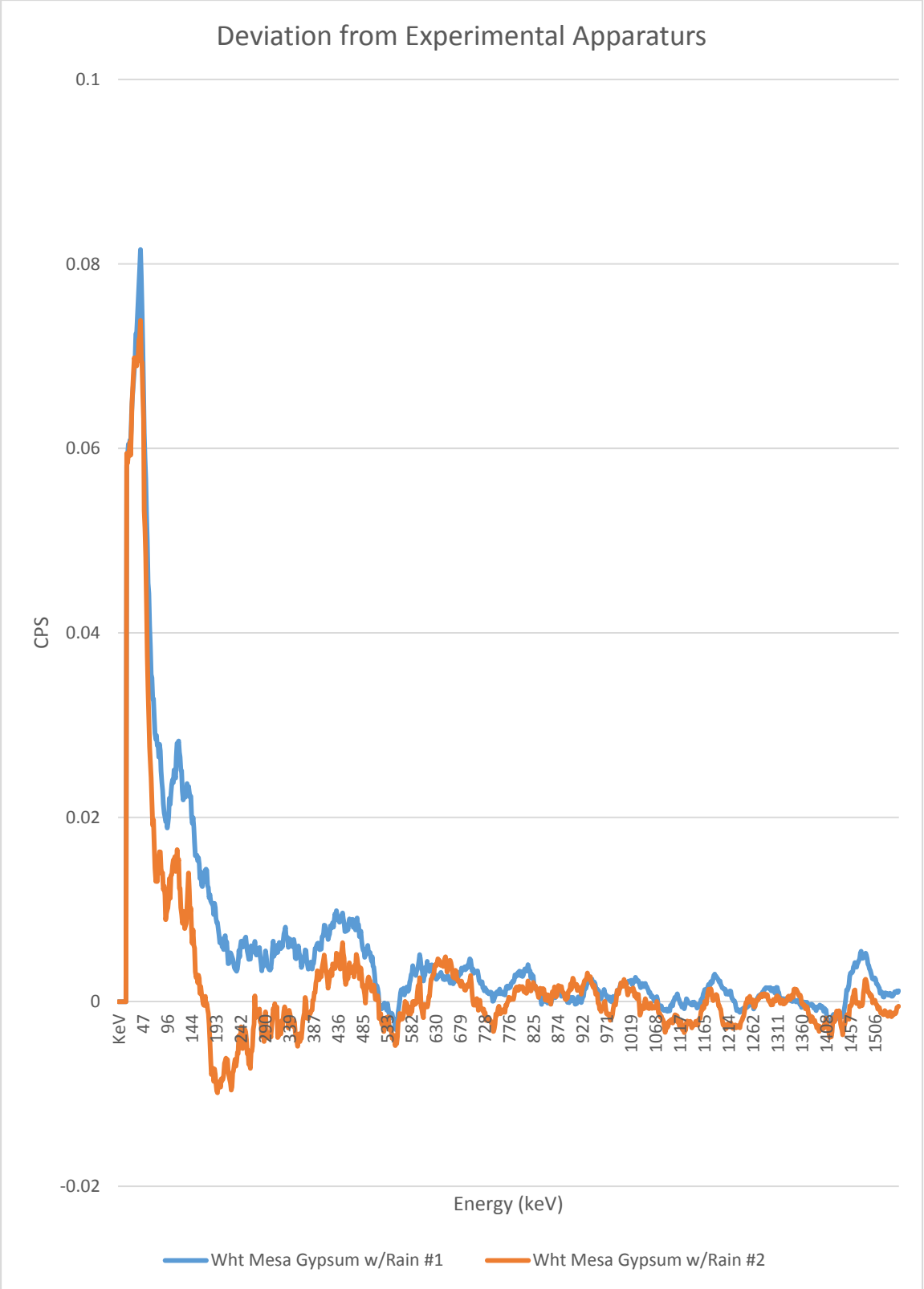


Figure 4-4b: Deviation in CPS from Experimental Apparatus with Introduction of White Mesa Gypsum Rain Samples

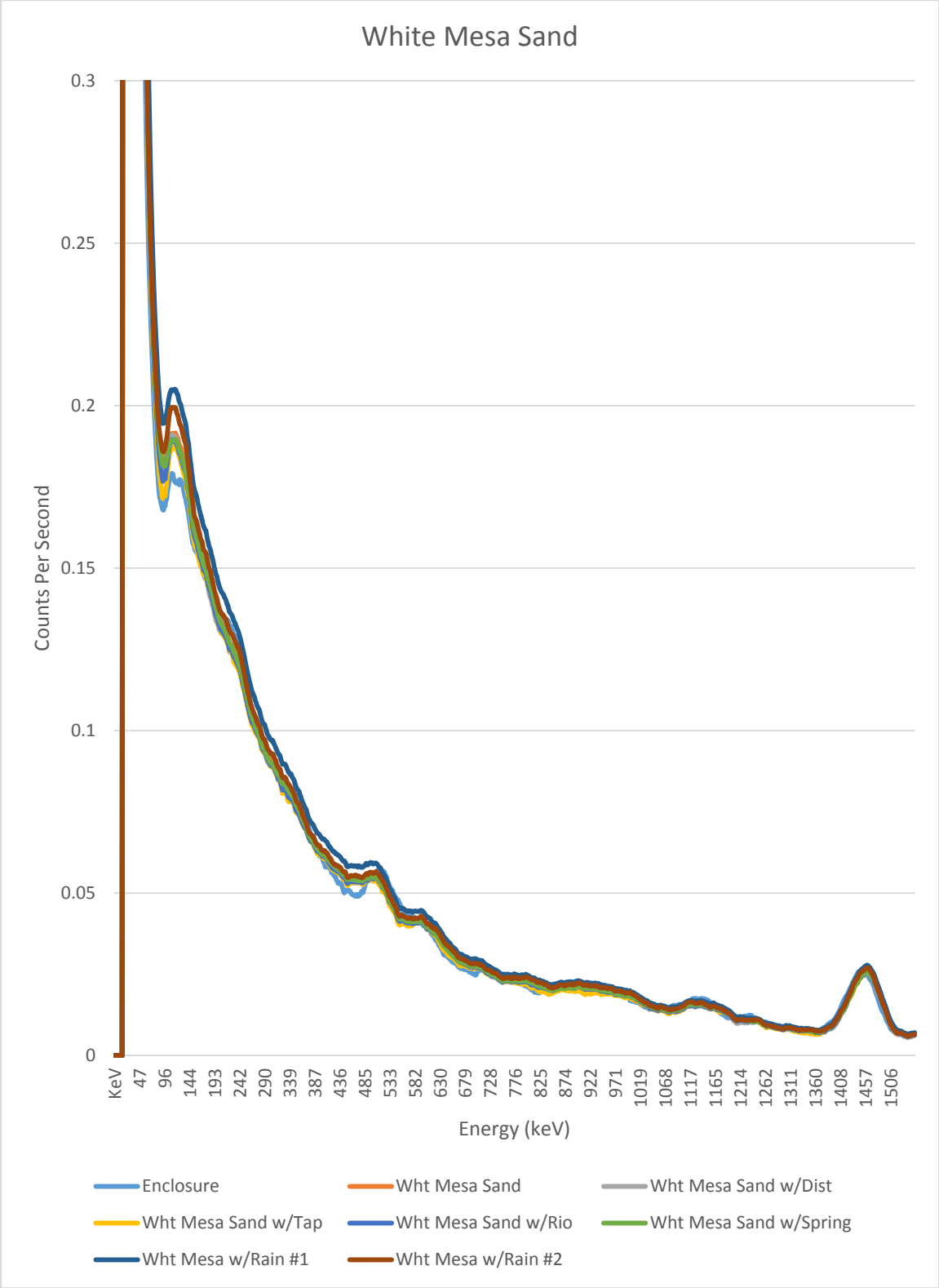


Figure 4-5a: White Mesa Sand Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

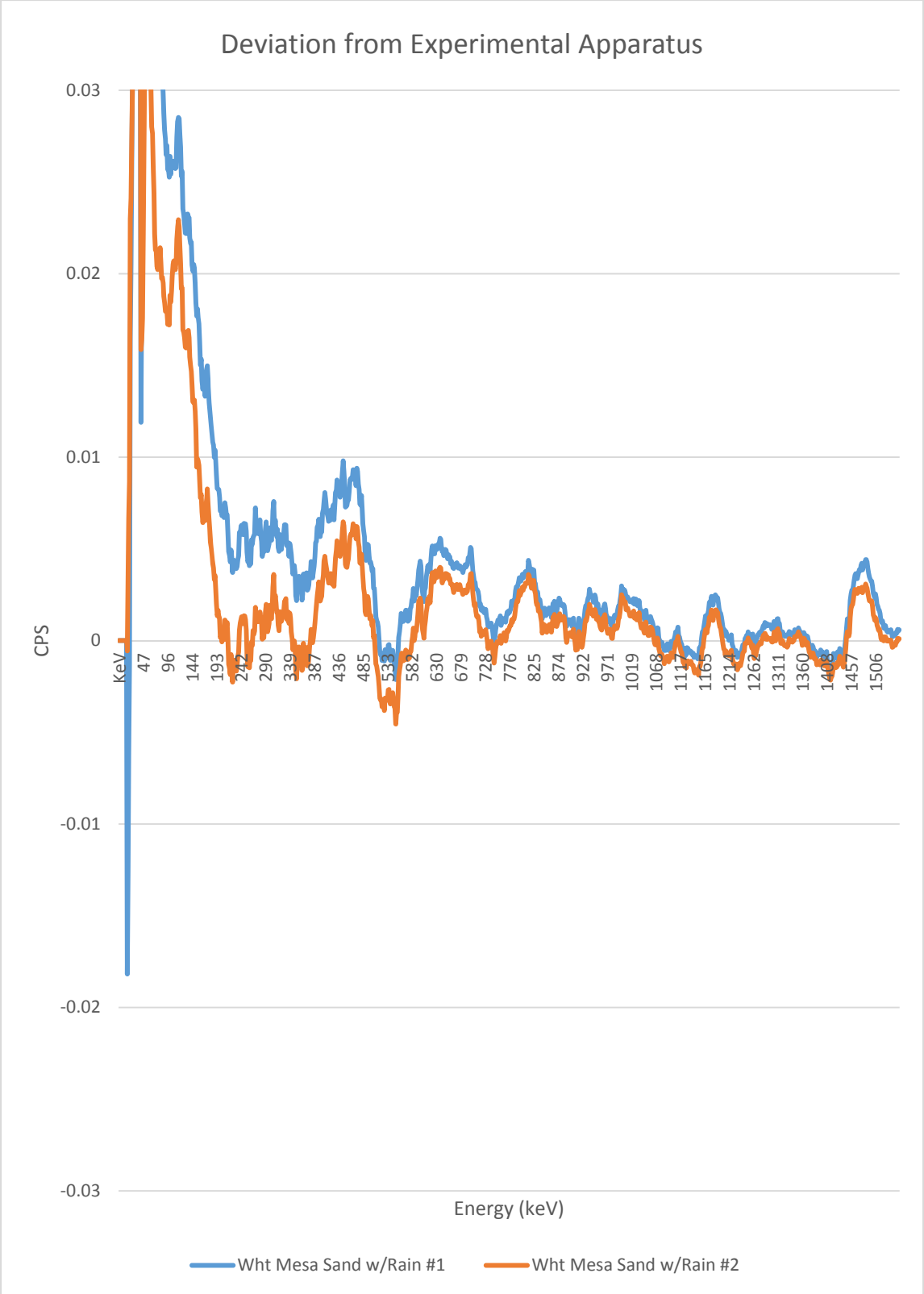


Figure 4-5b: Deviation in CPS from Experimental Apparatus with Introduction of White Mesa Sand Rain Samples

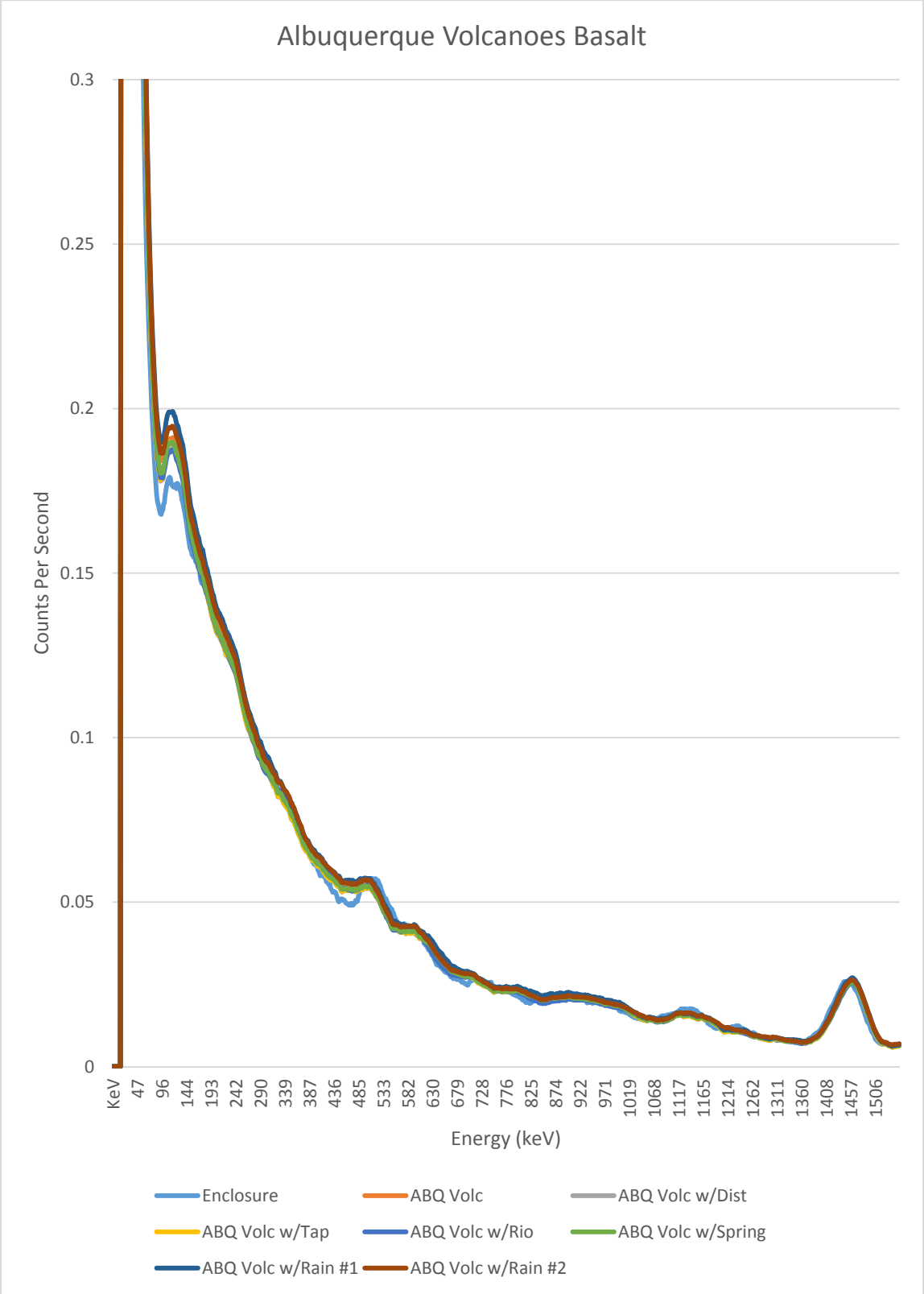


Figure 4-6a: Albuquerque Volcanoes Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

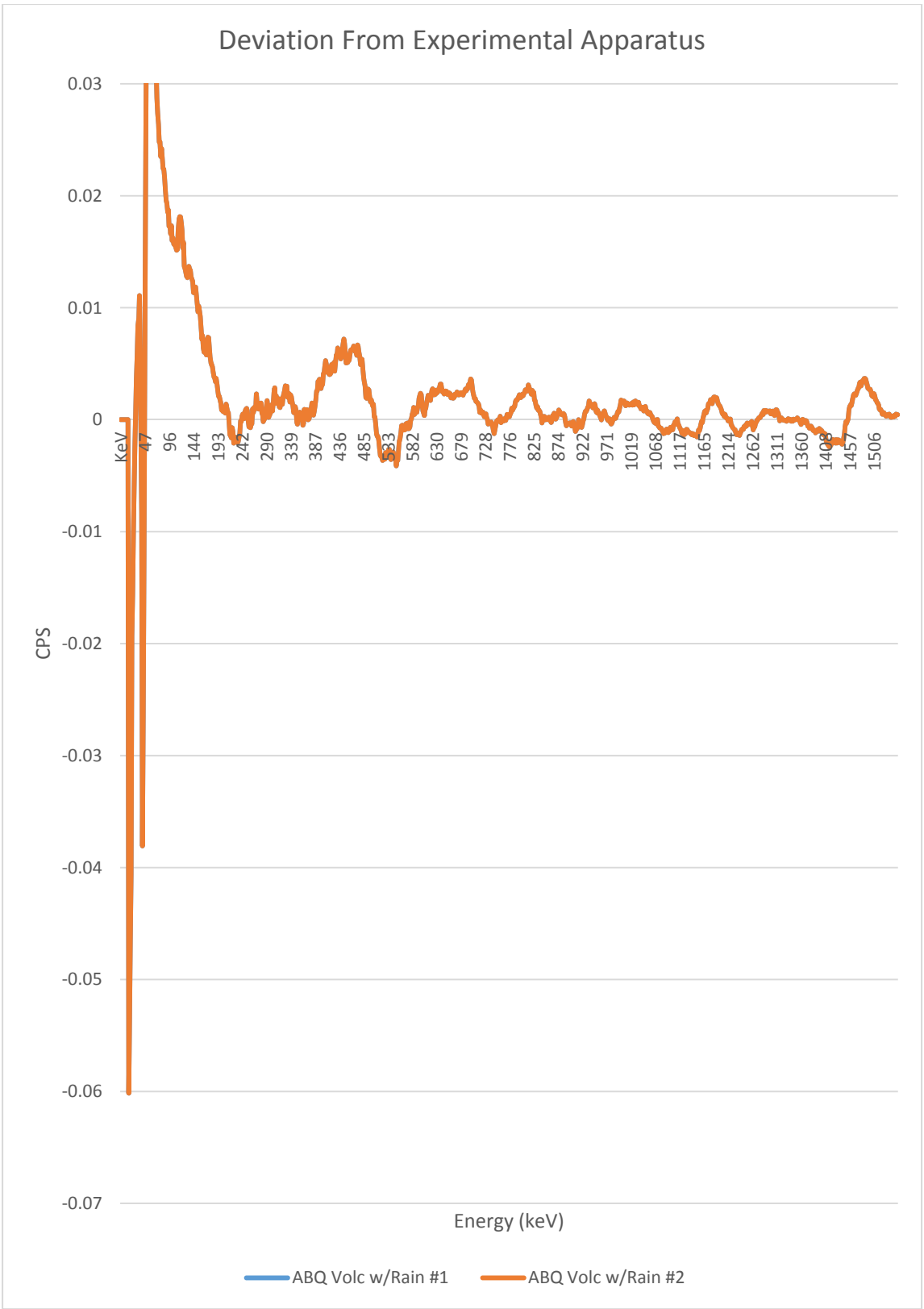


Figure 4-6b: Deviation in CPS from Experimental Apparatus with Introduction of ABQ Volcanic Basalt Rain Samples

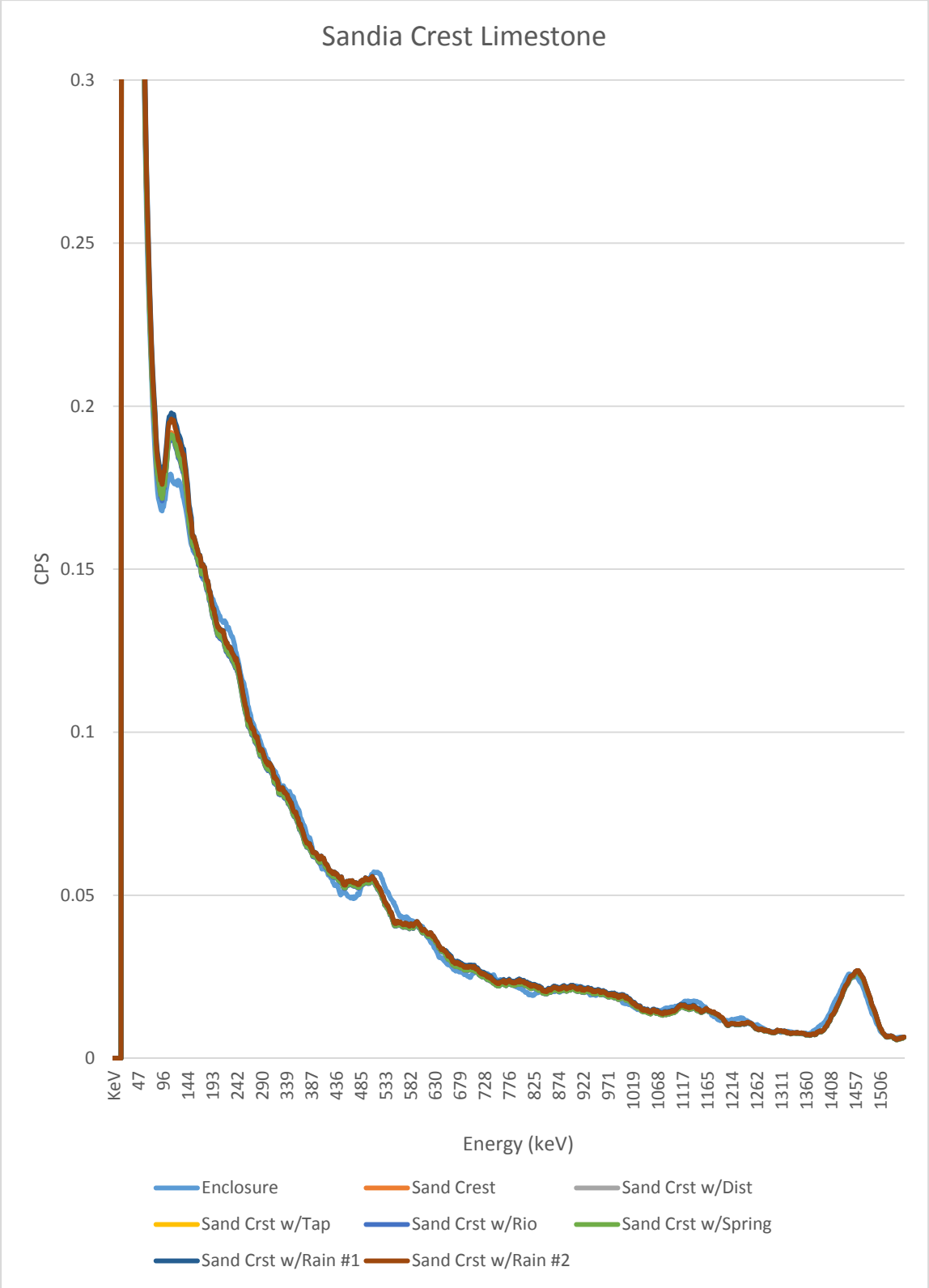


Figure 4-7a: Sandia Crest Limestone Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

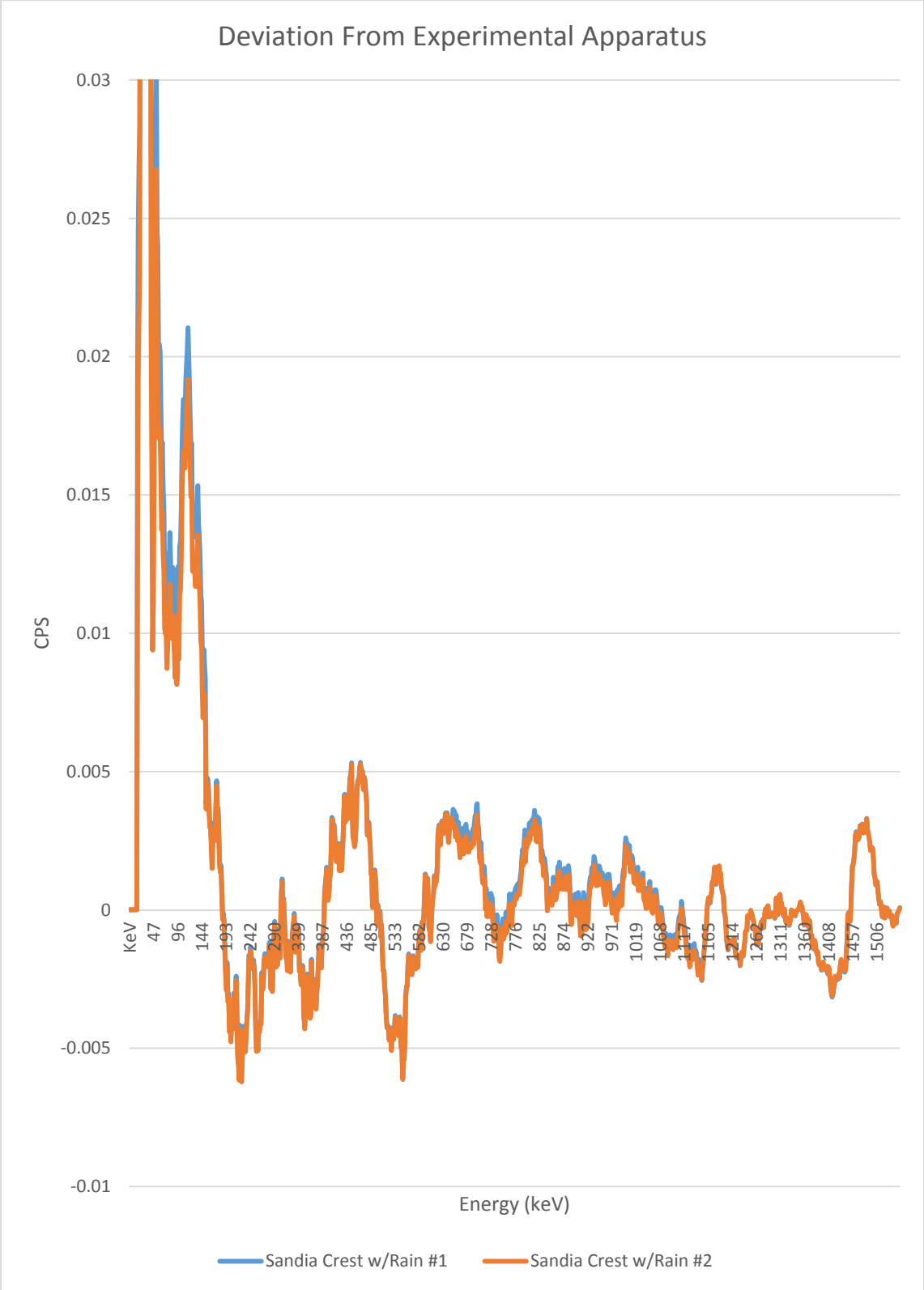


Figure 4-7b: Deviation in CPS from Experimental Apparatus with Introduction of Sandia Crest Limestone Rain Samples

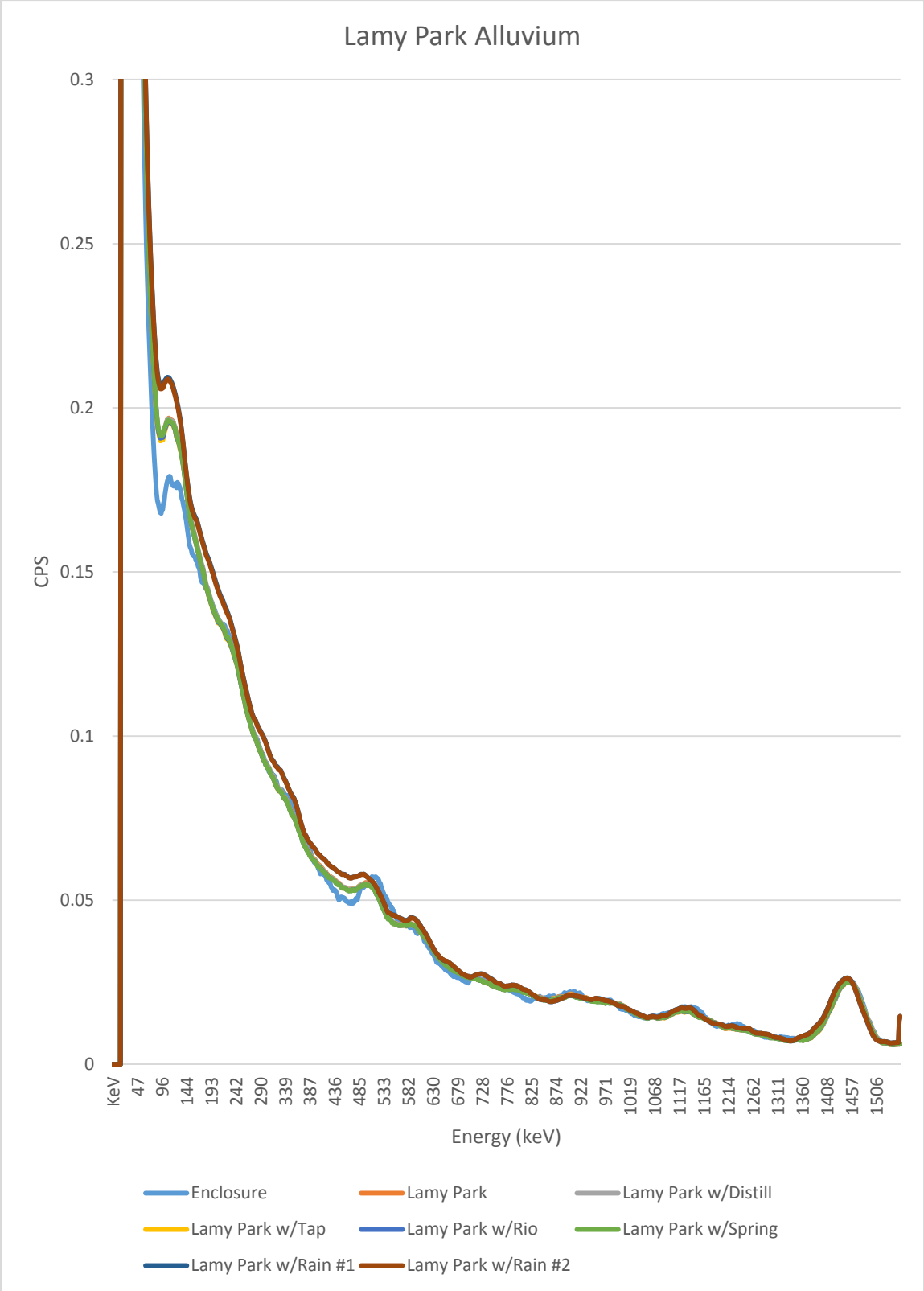


Figure 4-8a: Lamy Park Alluvium Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

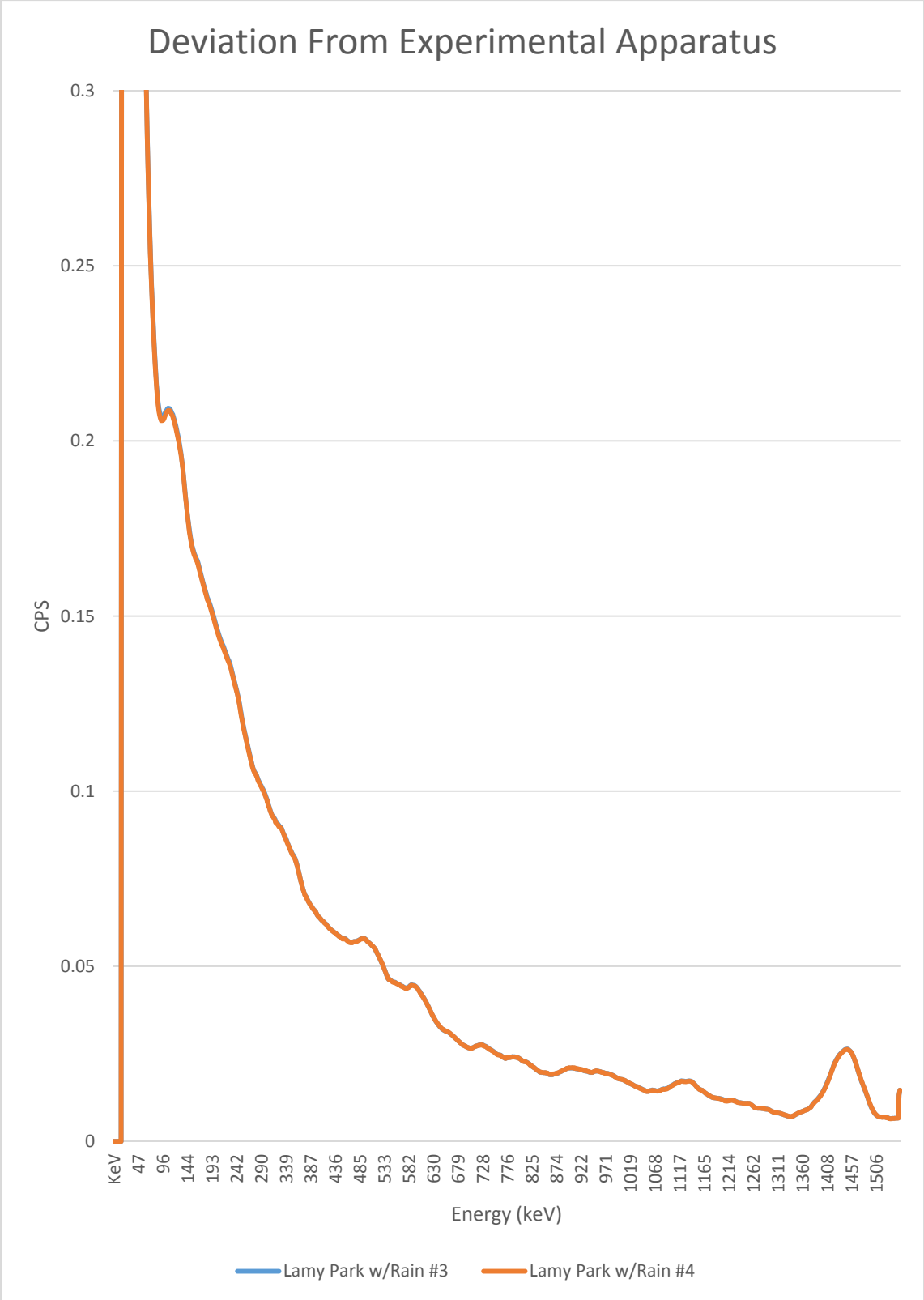


Figure 4-8b: Deviation in CPS from Experimental Apparatus with Introduction of Lamy Park Alluvium Rain Samples

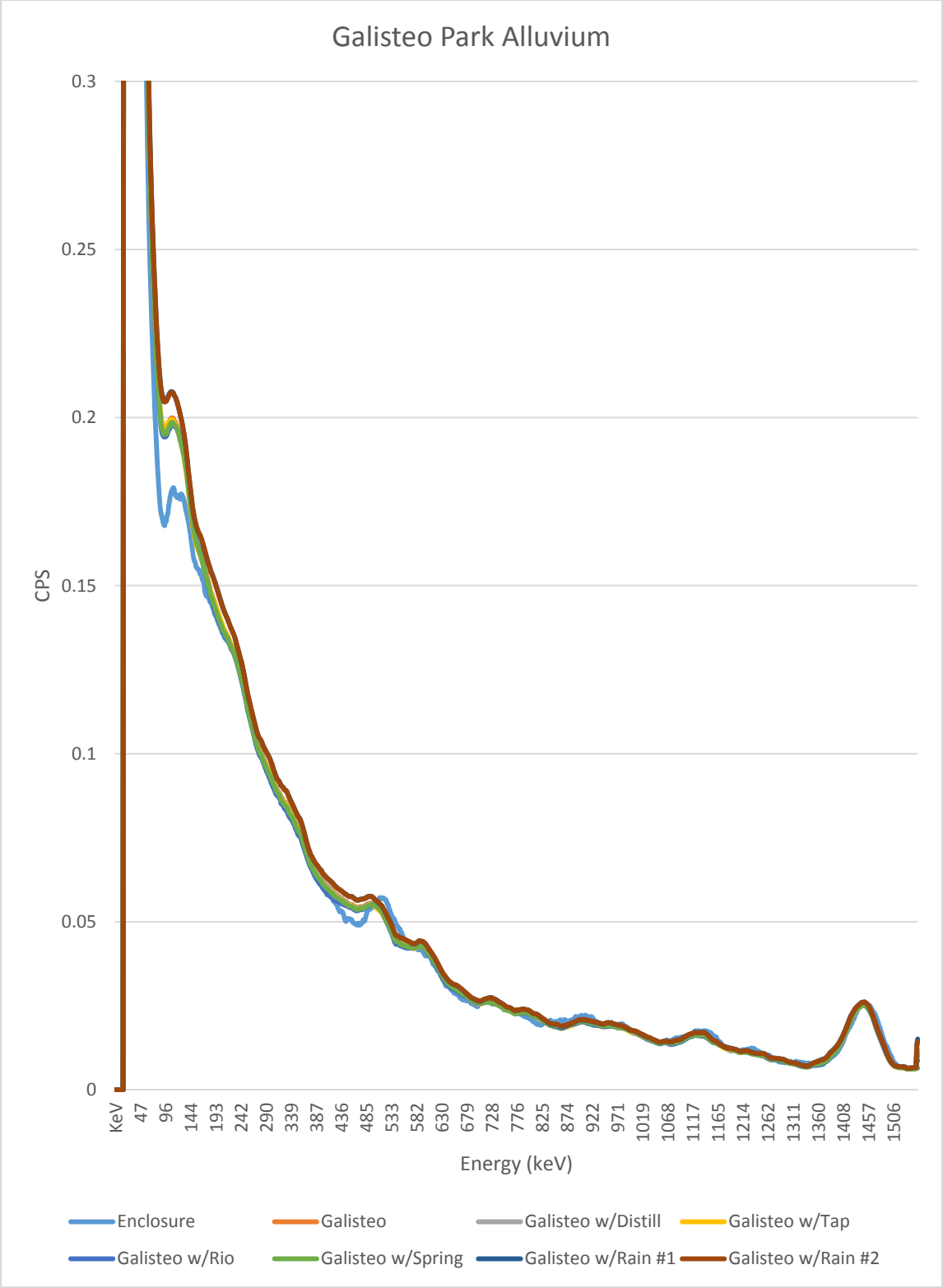


Figure 4-9a: Galisteo Park Alluvium Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

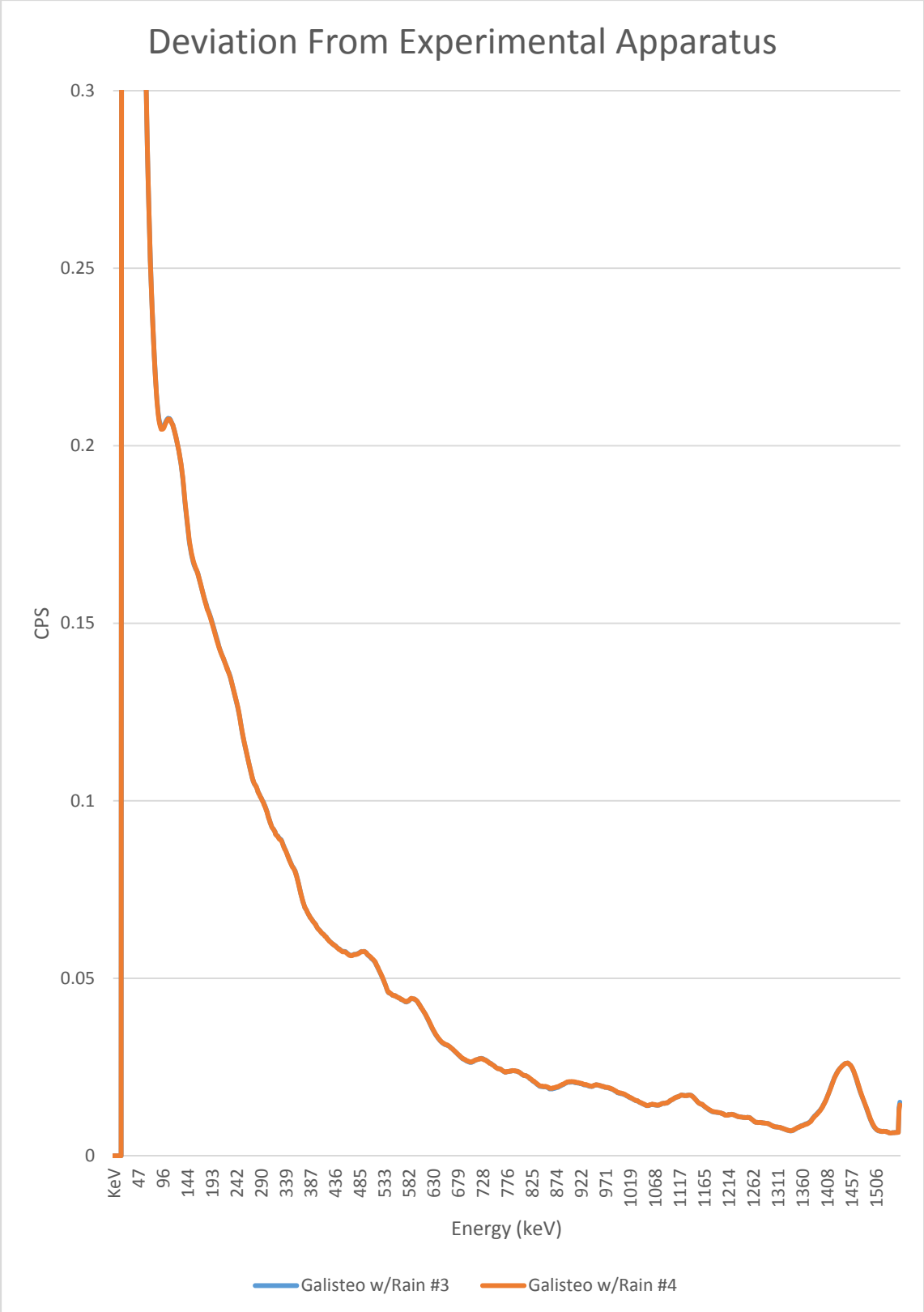


Figure 4-9b: Deviation in CPS from Experimental Apparatus with Introduction of Galisteo Park Alluvium Rain Samples

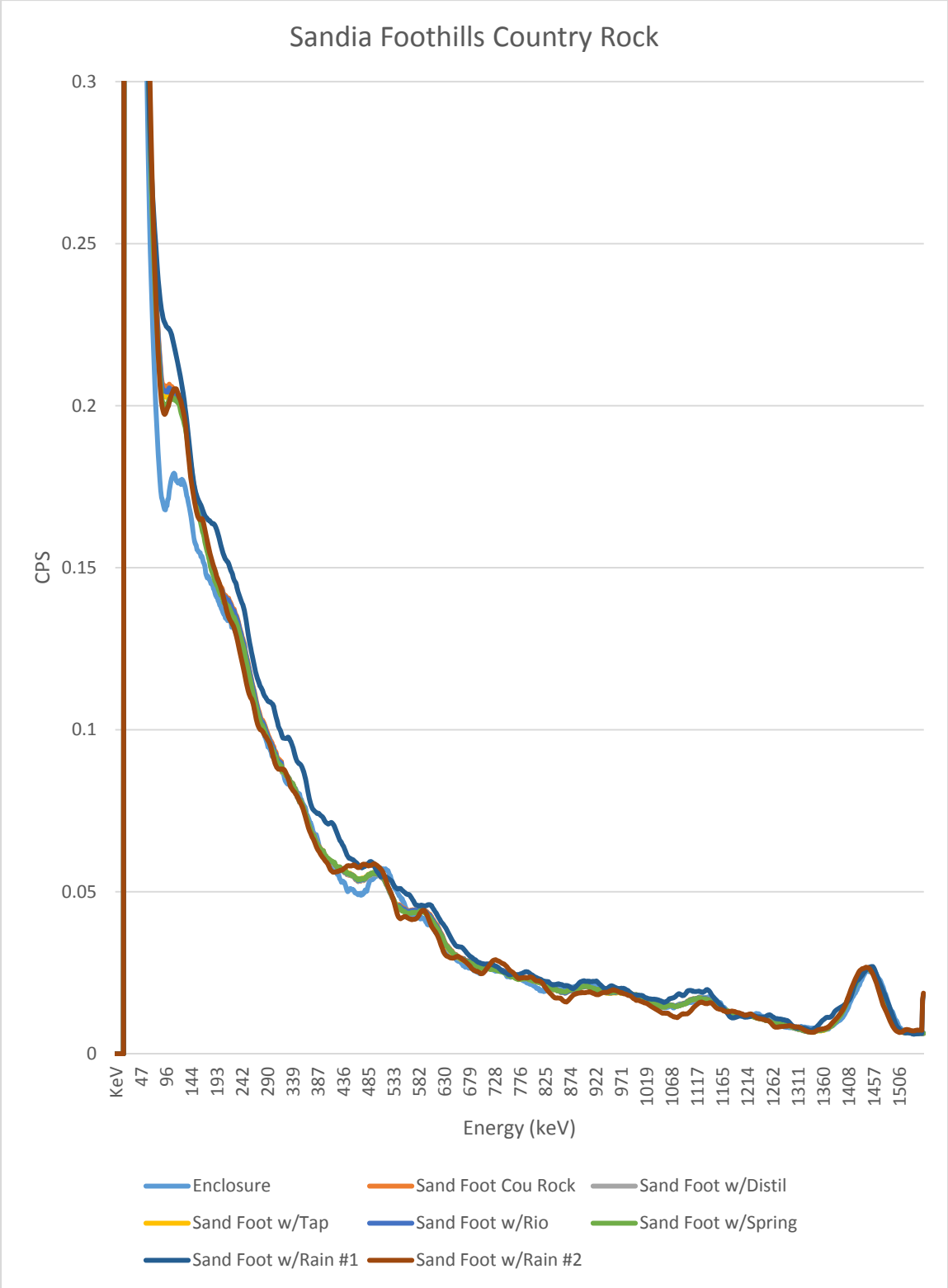


Figure 4-10a: Sandia Foothills Country Rock Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

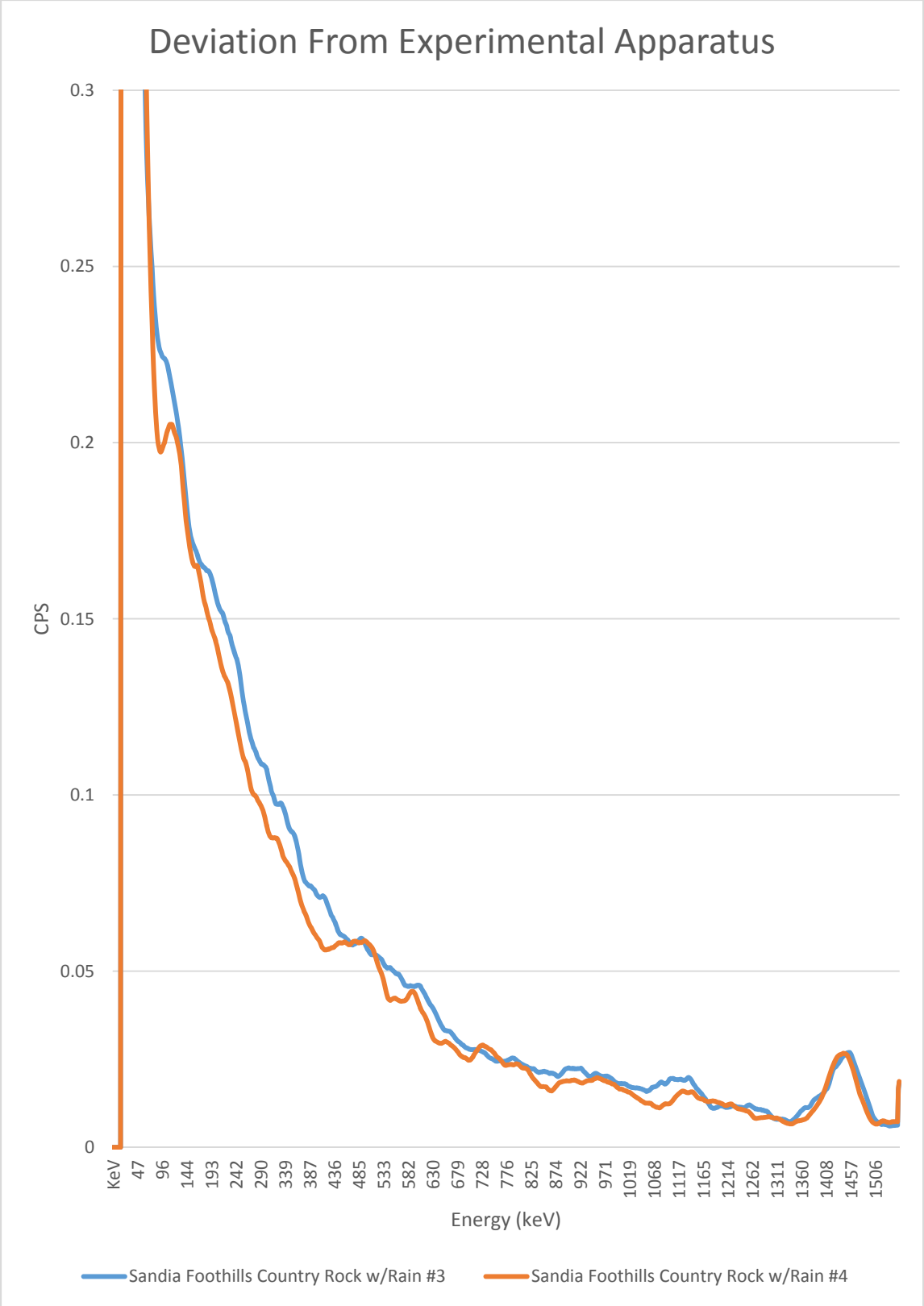


Figure 4-10b: Deviation in CPS from Experimental Apparatus with Introduction of Sandia Foothills Country Rock Rain Samples

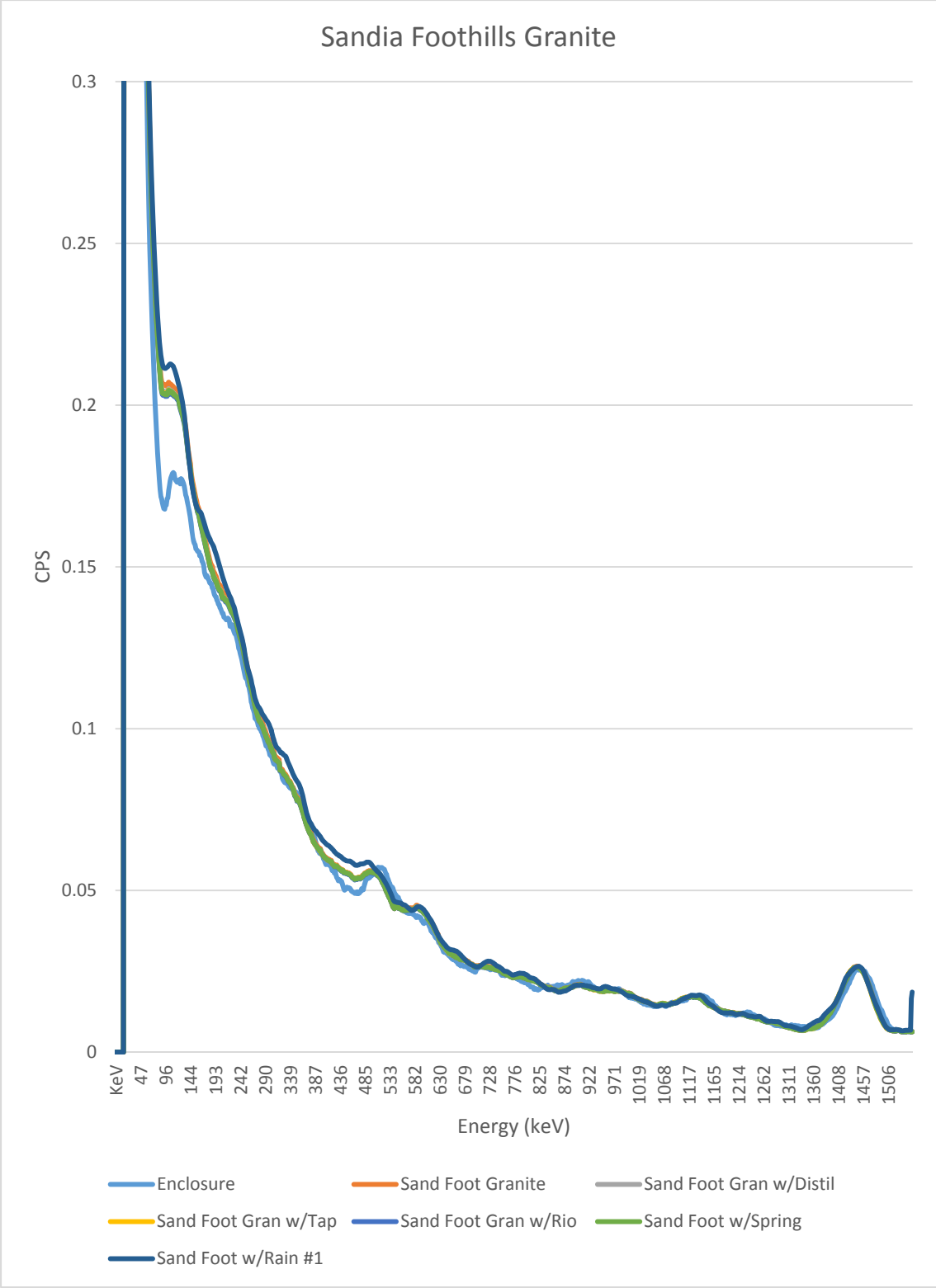


Figure 4-11a: Sandia Foothills Granite Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

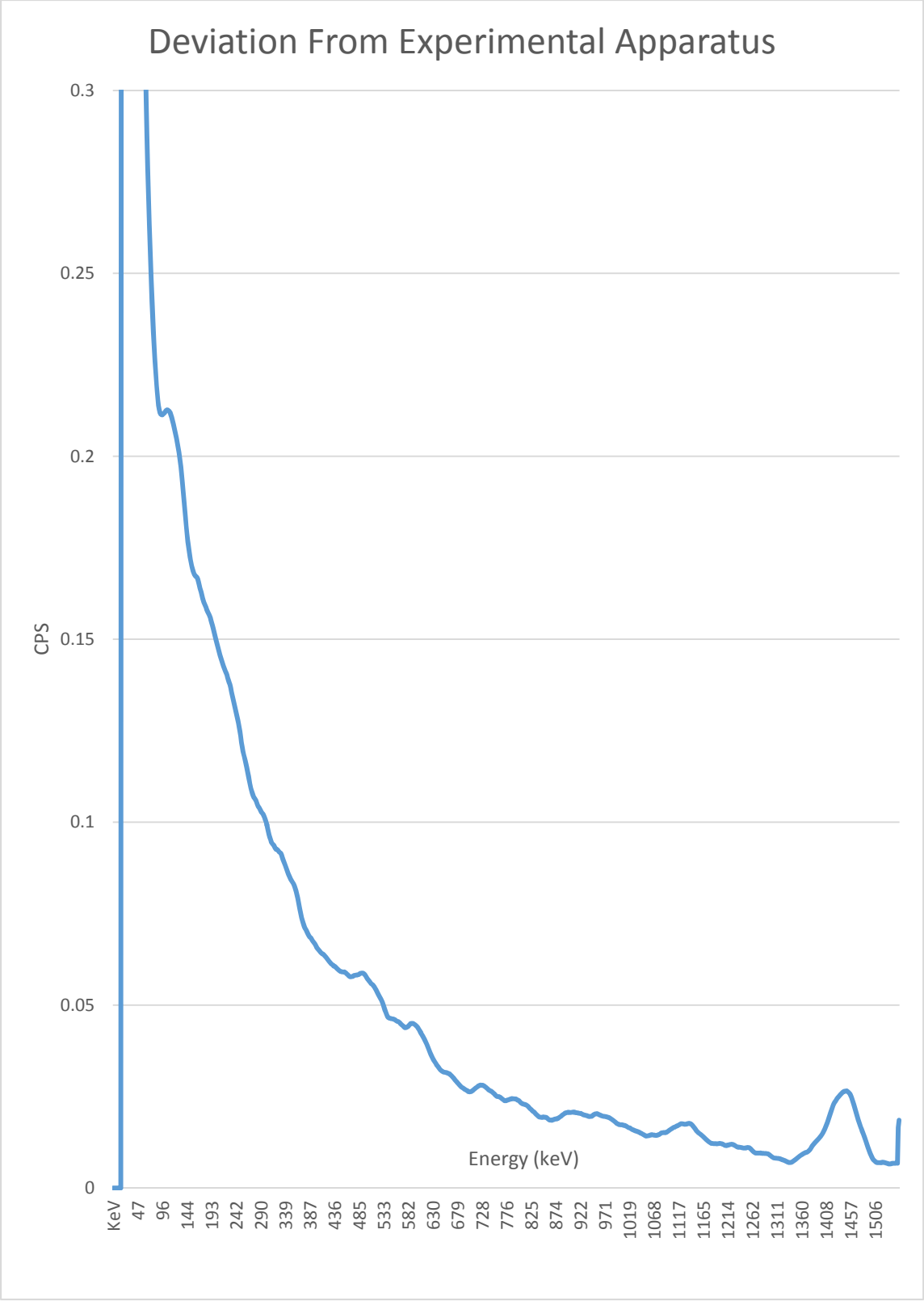


Figure 4-11b: Deviation in CPS from Experimental Apparatus with Introduction of Sandia Foothills Granite Rain Sample

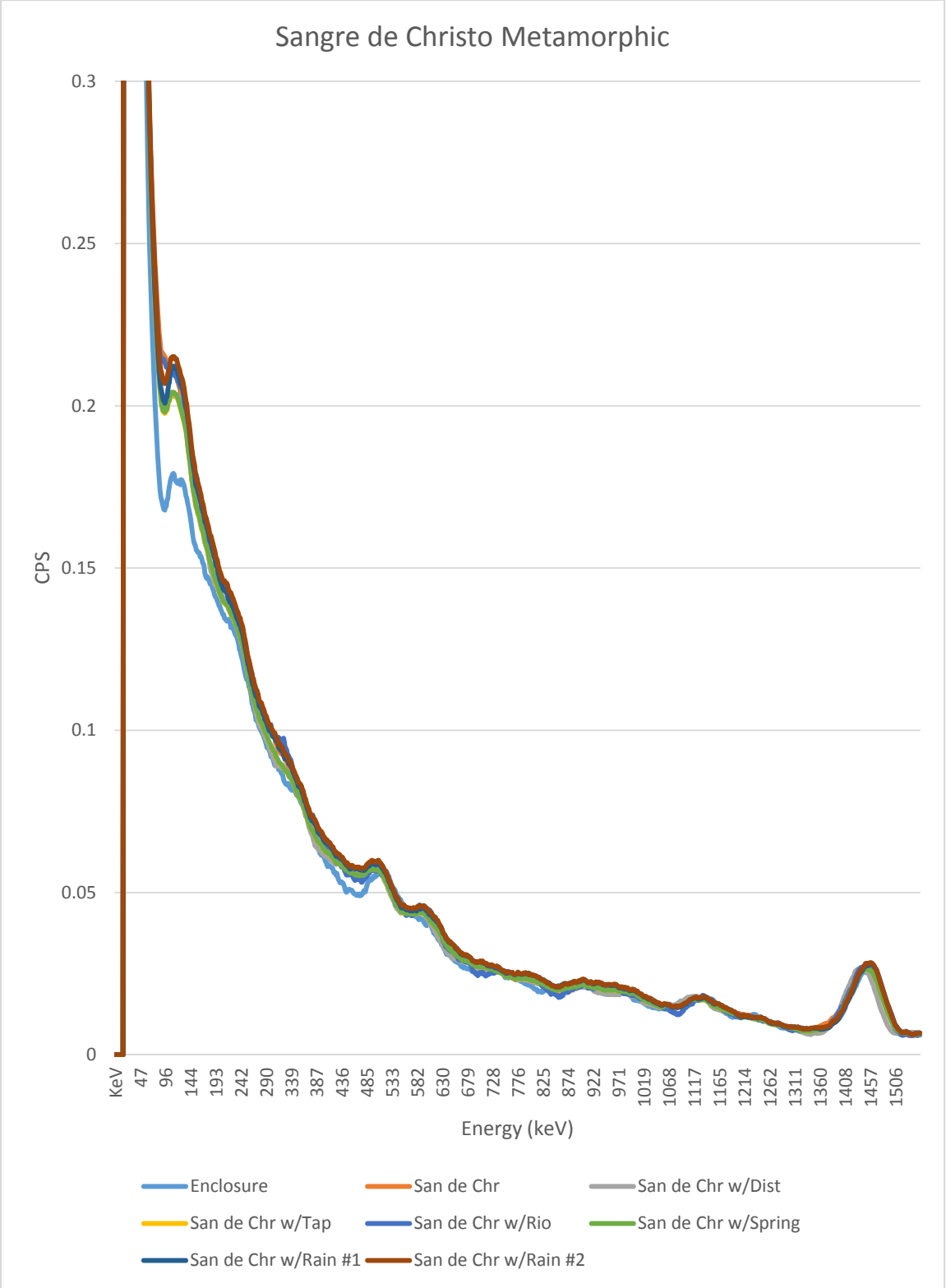


Figure 4-12a: SdC Metamorphic Rock Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

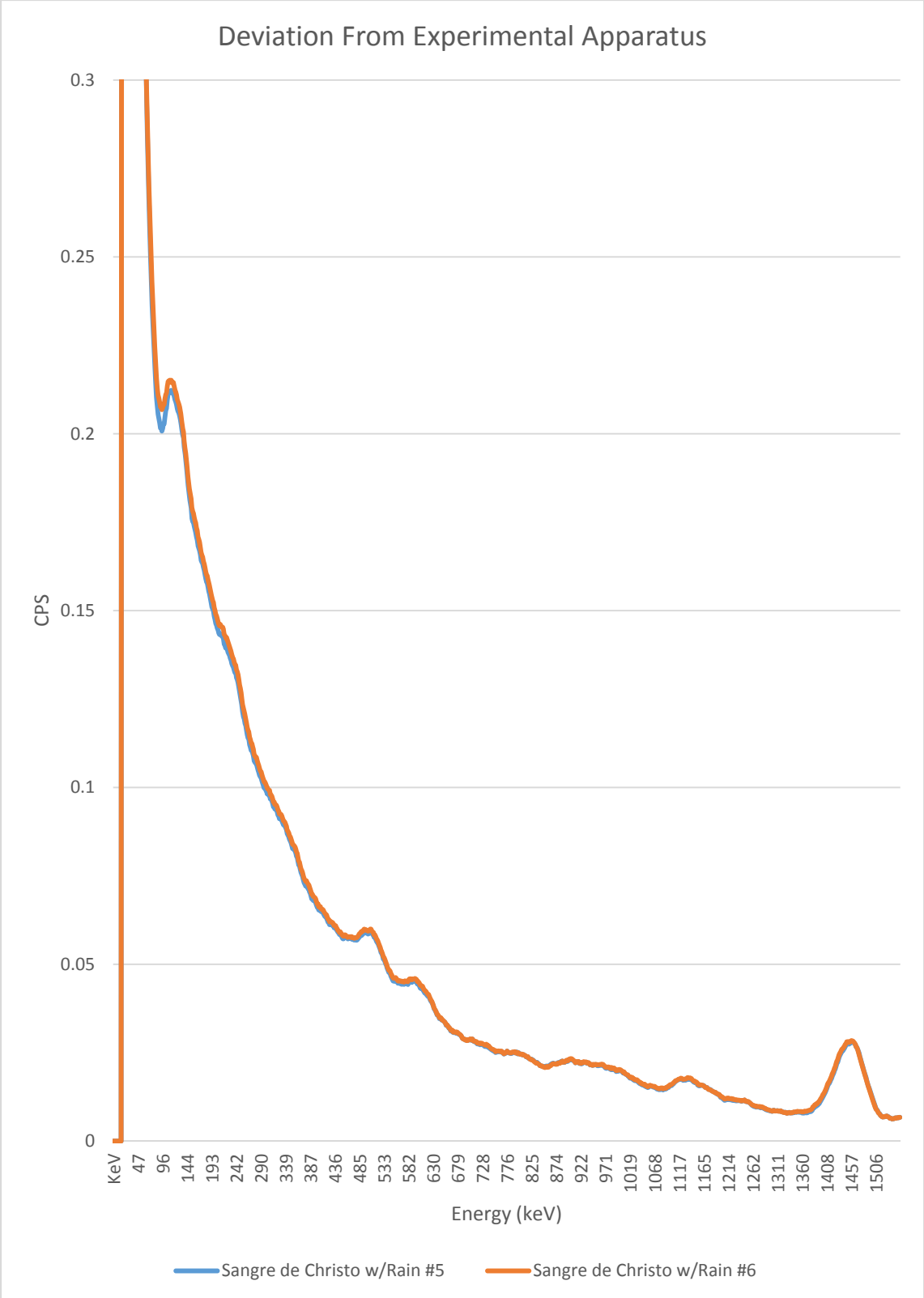


Figure 4-12b: Deviation in CPS from Experimental Apparatus with Introduction of SdC Metamorphic Rain Samples

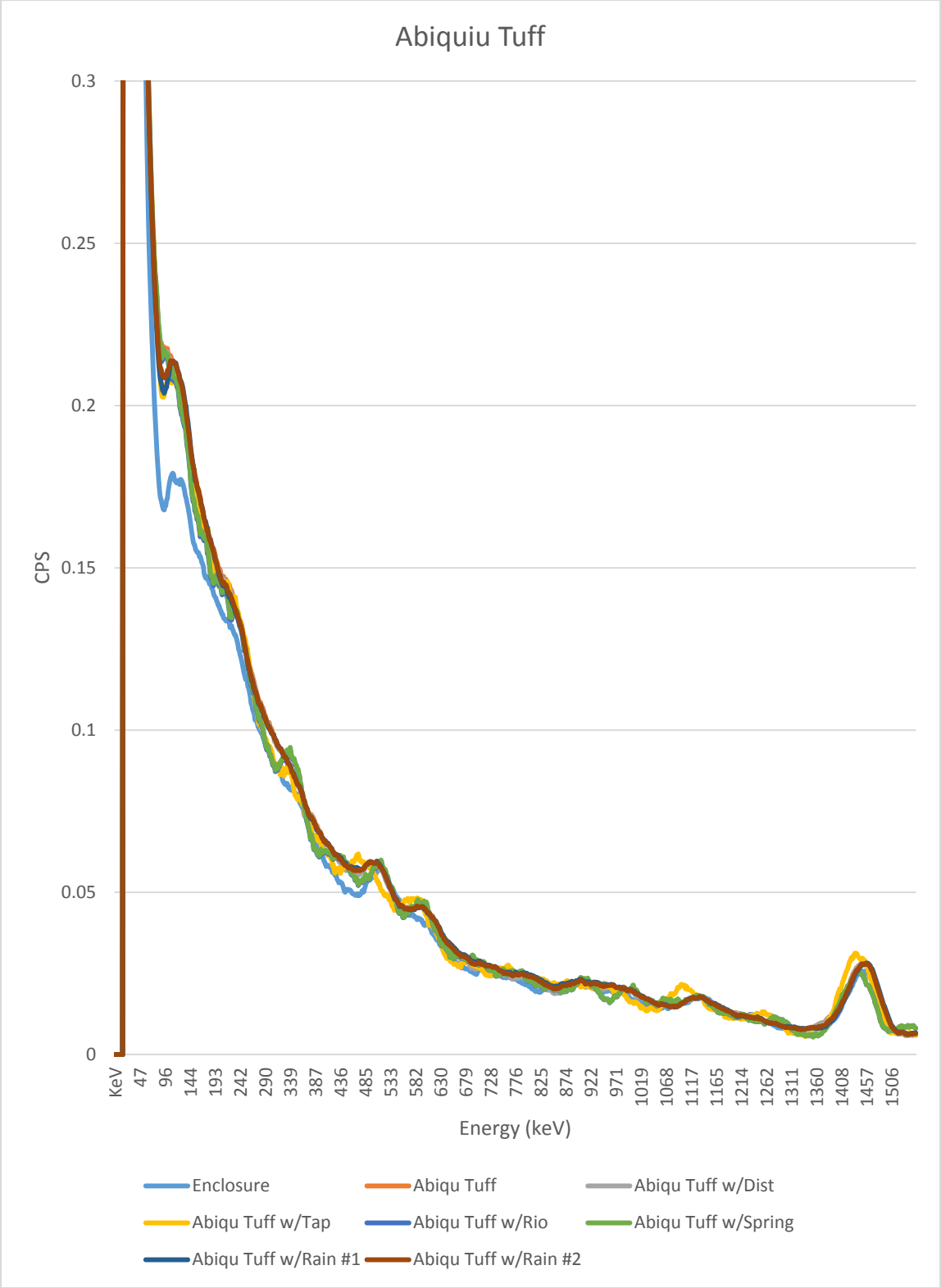


Figure 4-13a: Abiquiu Tuff Surveyed Average Counts per Second per Gamma Energy Compared to Enclosure CPS

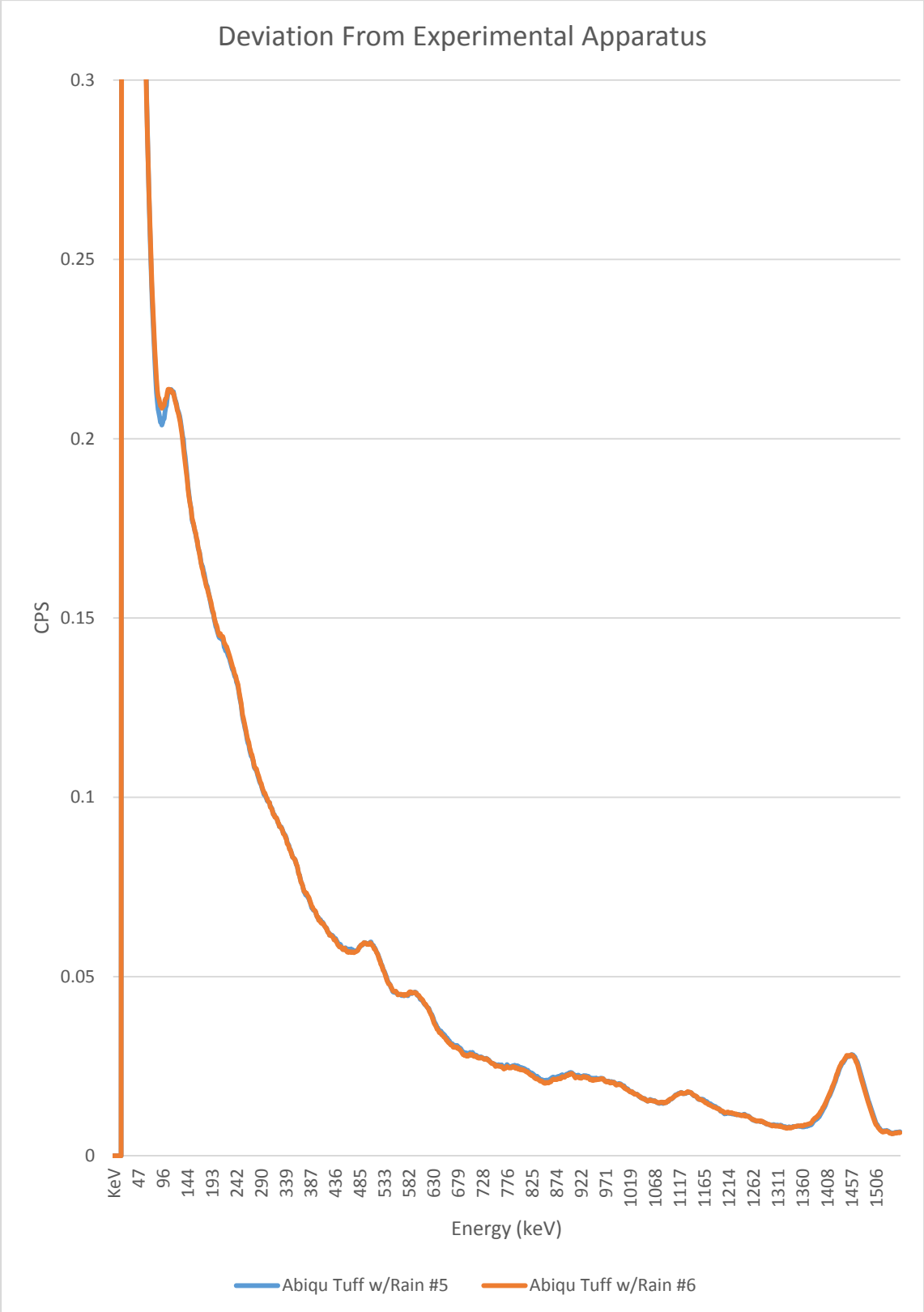


Figure 4-12b: Deviation in CPS from Experimental Apparatus with Introduction of Abiquiu Tuff Rain Samples

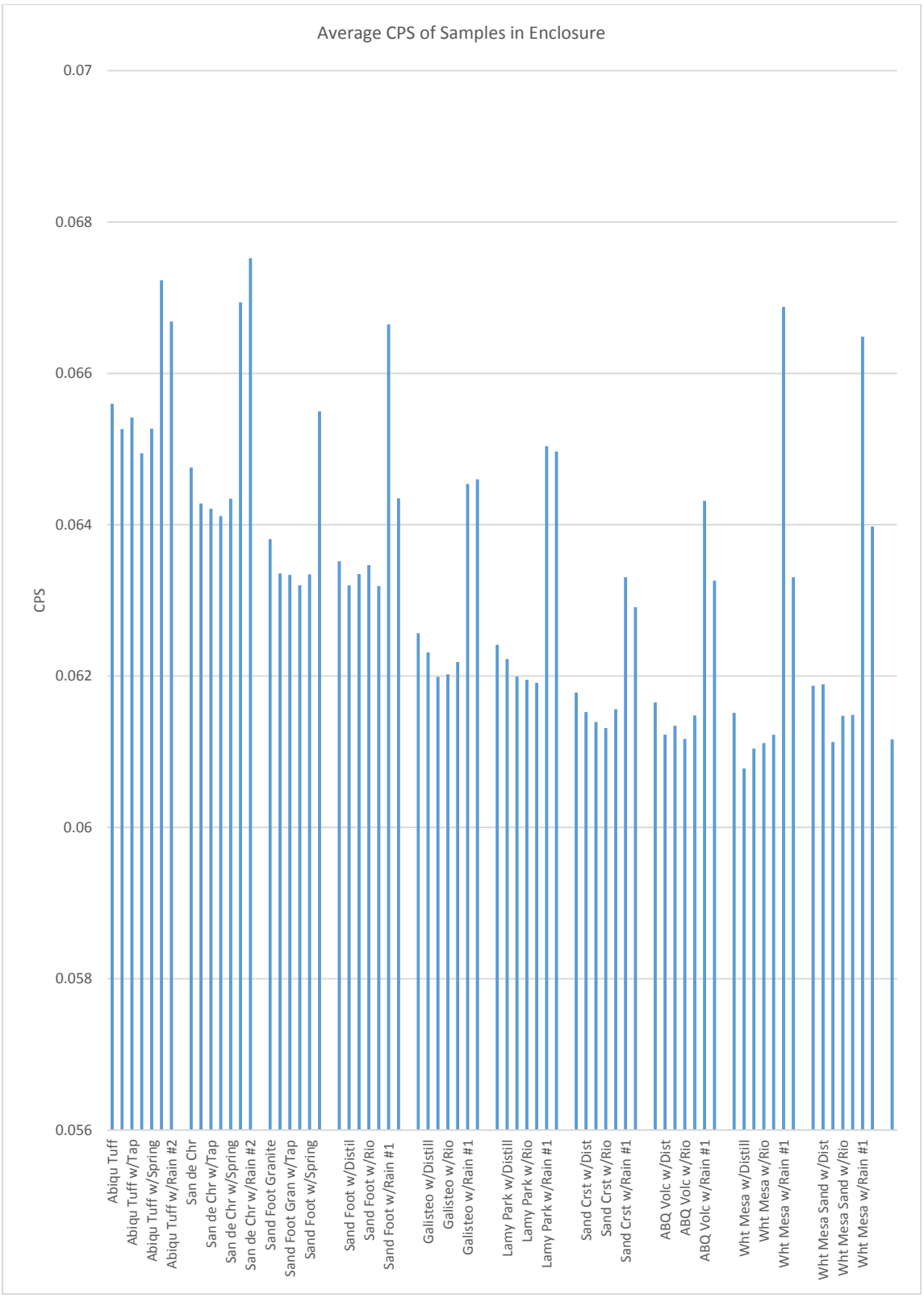


Figure 4-15: Average CPS for Each Sample with Waters and Rainwater

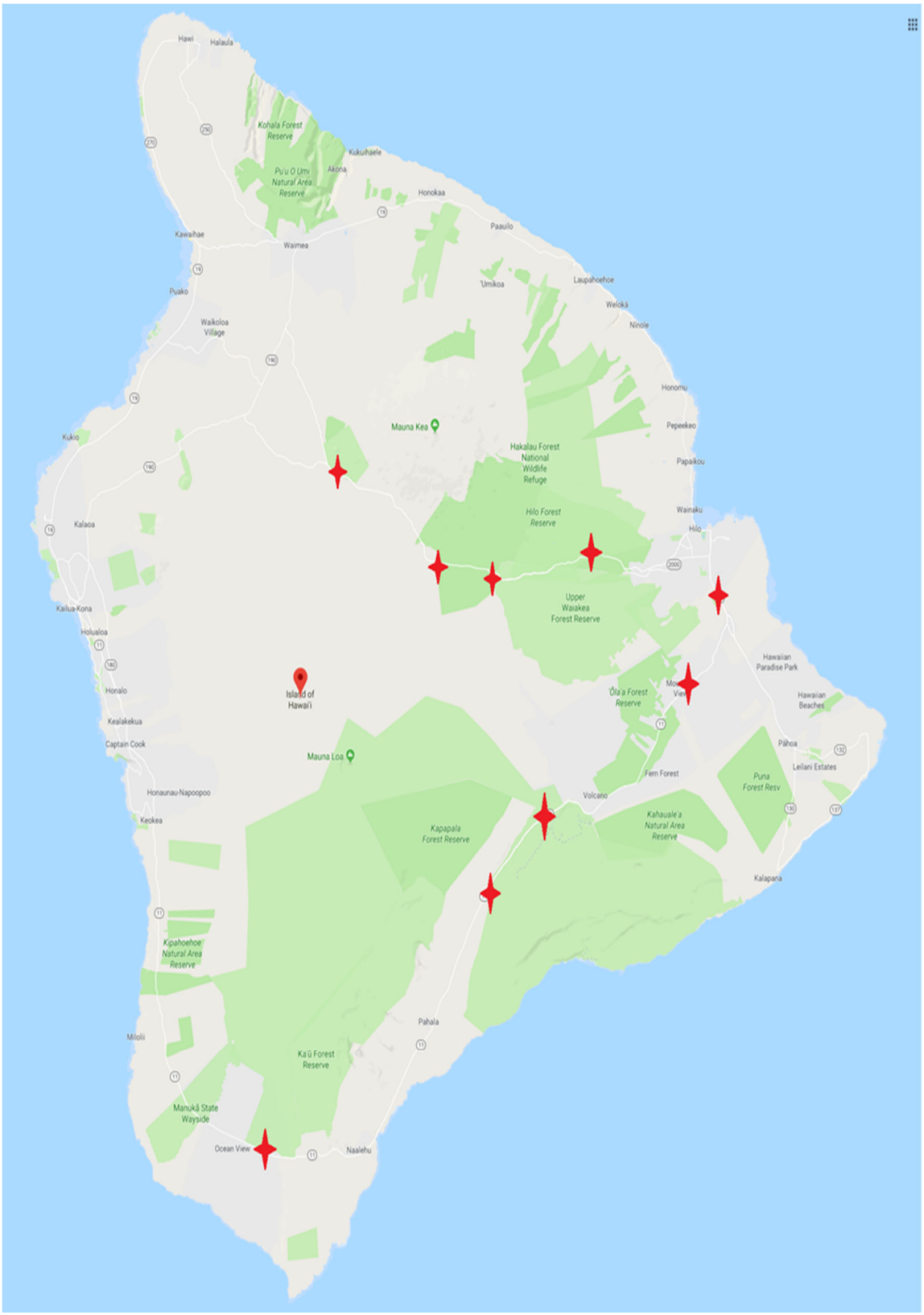


Figure 4-16: Approximate Locations of Survey Sites on Kona Island

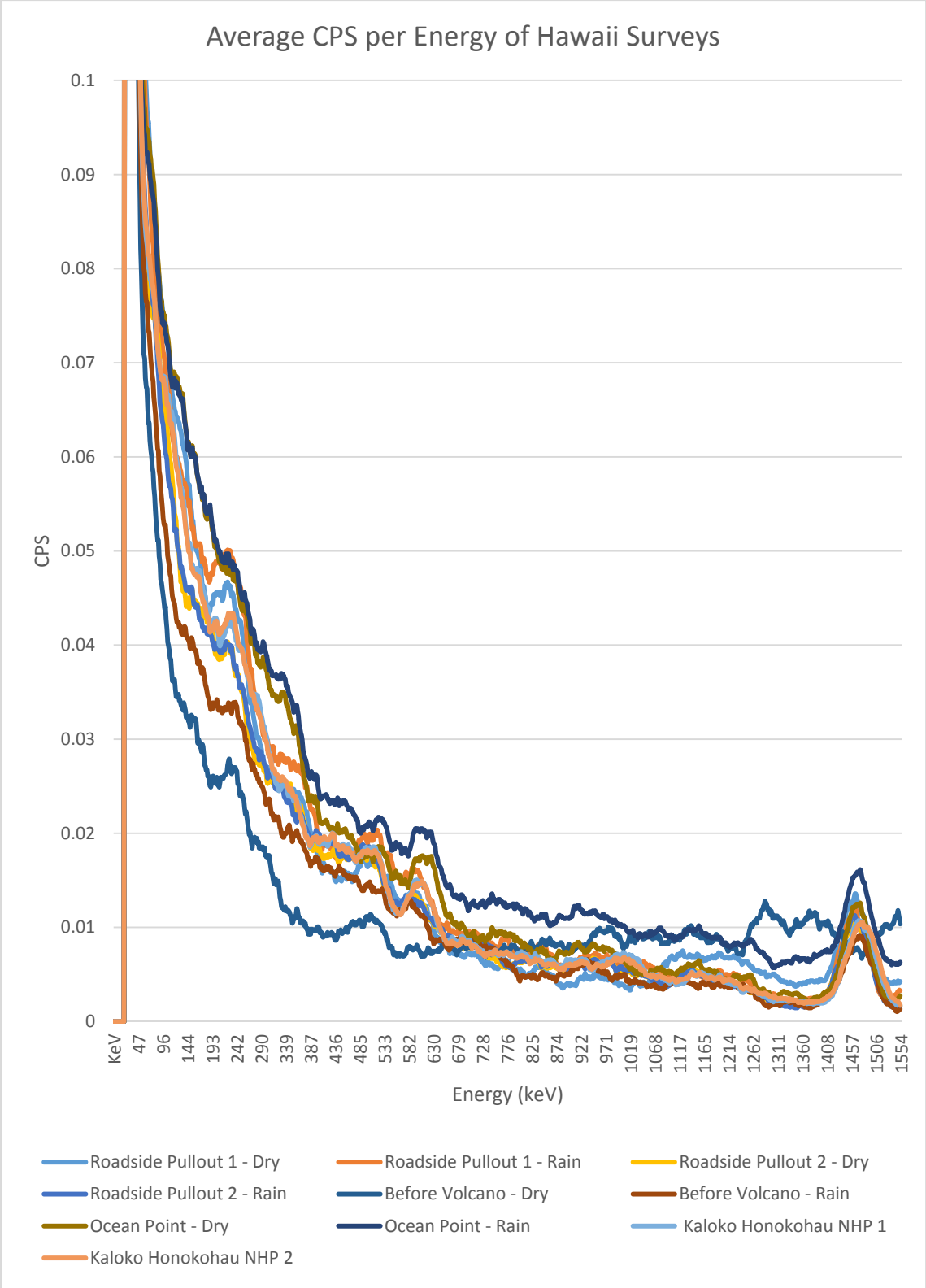


Figure 4-17a: Kona Island Surveyed Average Counts per Second Relative to Dry Conditions

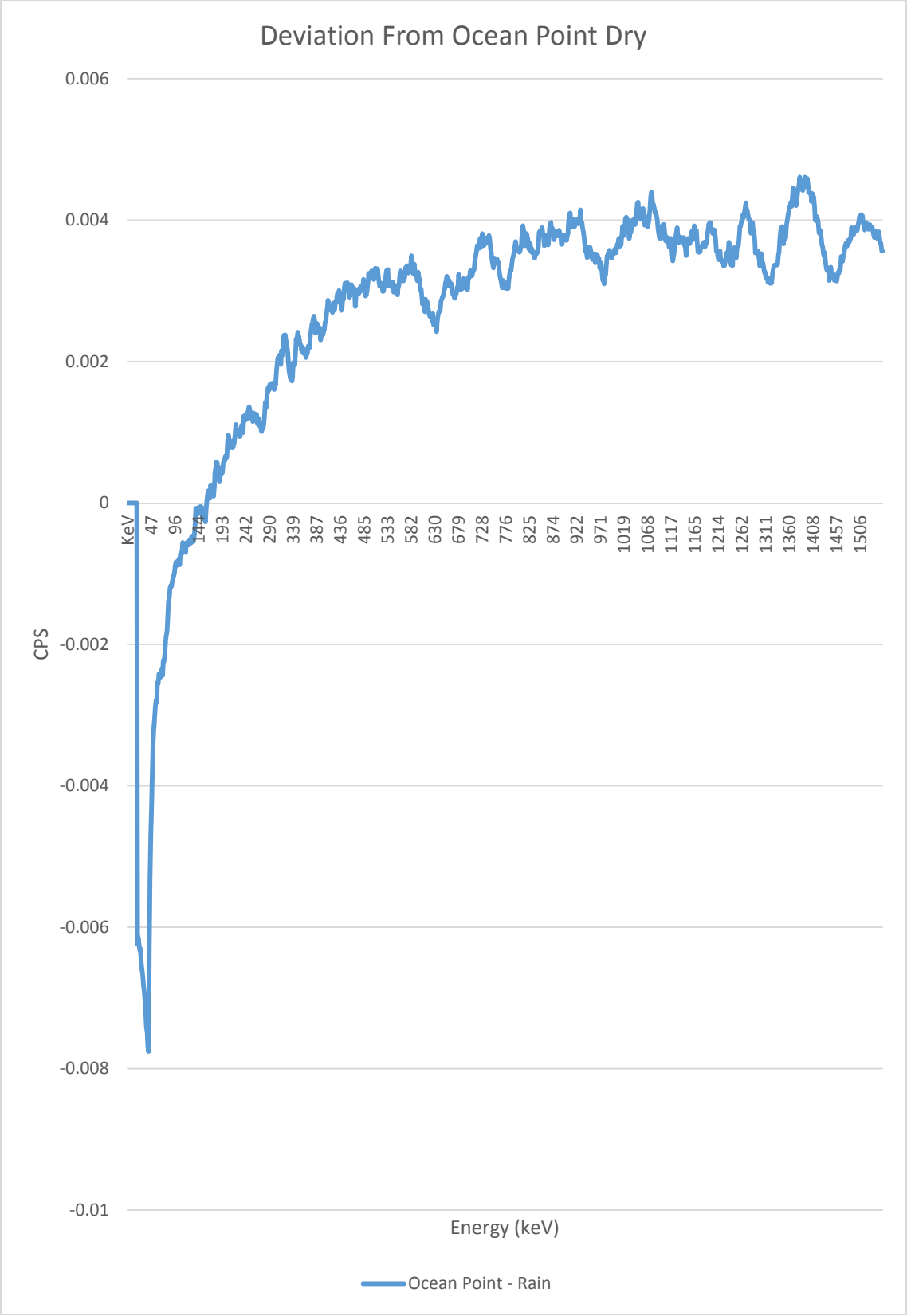


Figure 4-17b: Deviation in CPS from Dry Site to Site w/Rainfall

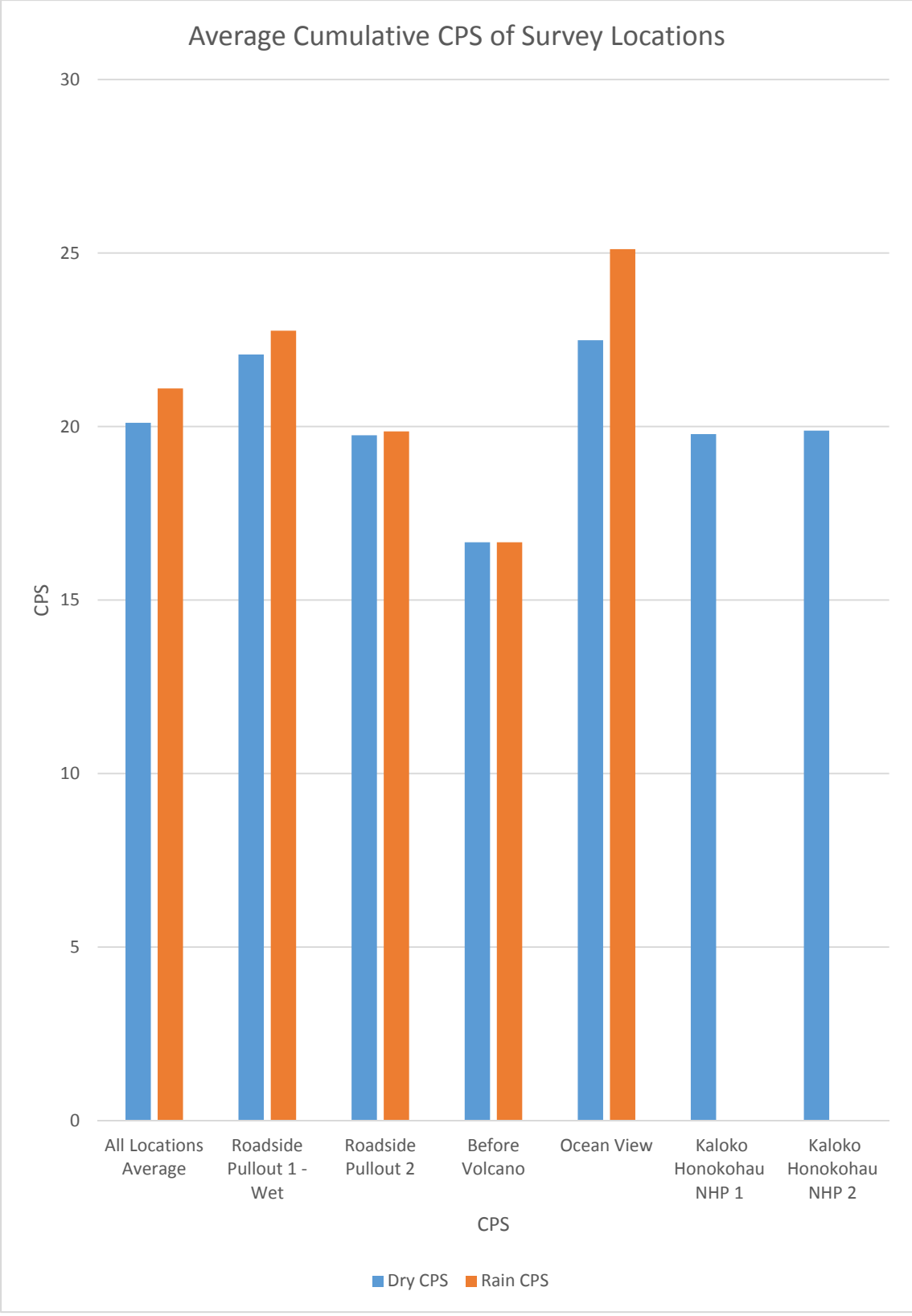


Figure 4-18: Kona Island Average Cumulative CPS With Average of All Wet/Dry Surveys

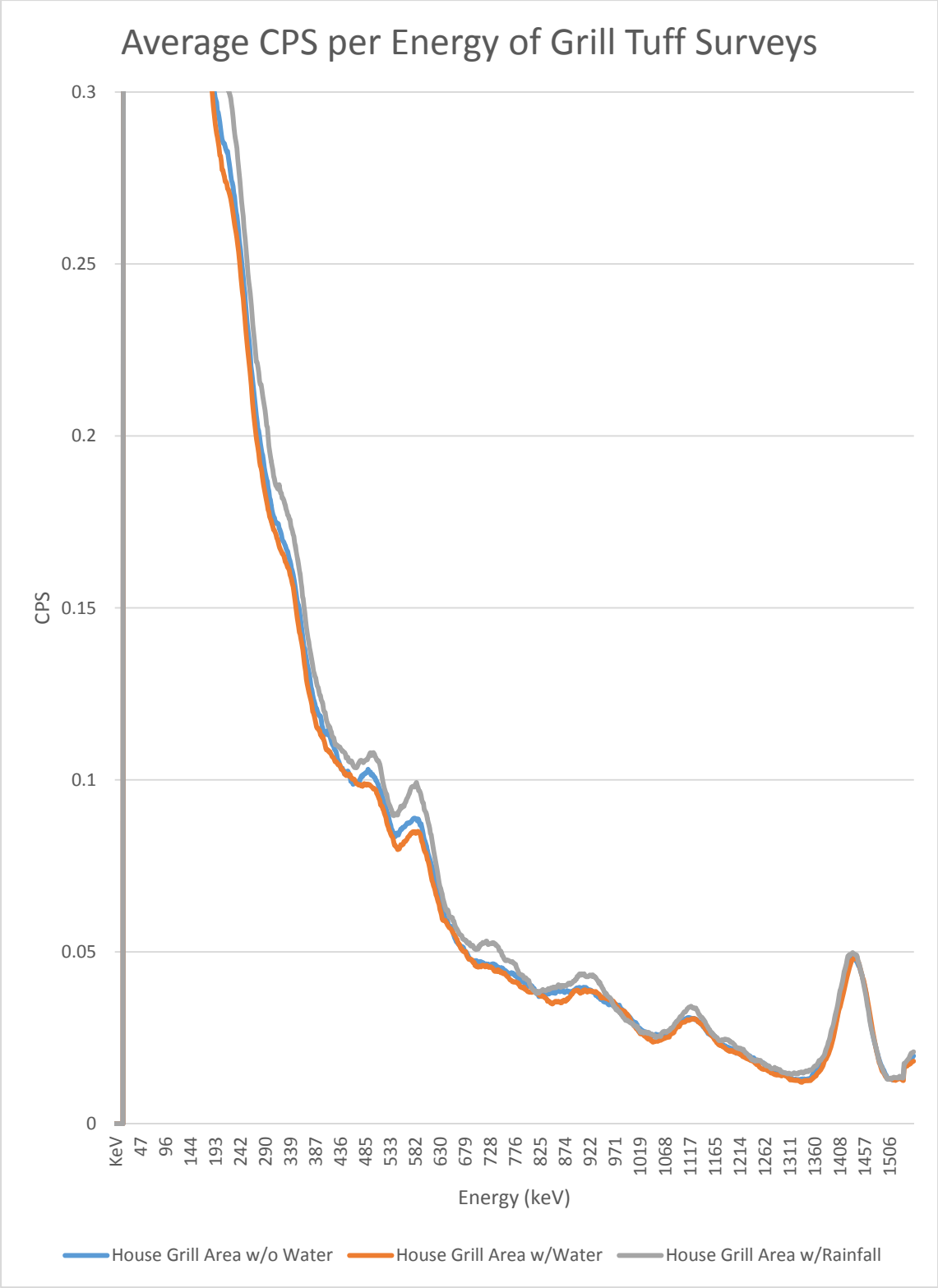


Figure 4-19a: Grill Tuff Surveyed Average Counts per Second per Gamma Energy Compared to Dry Conditions

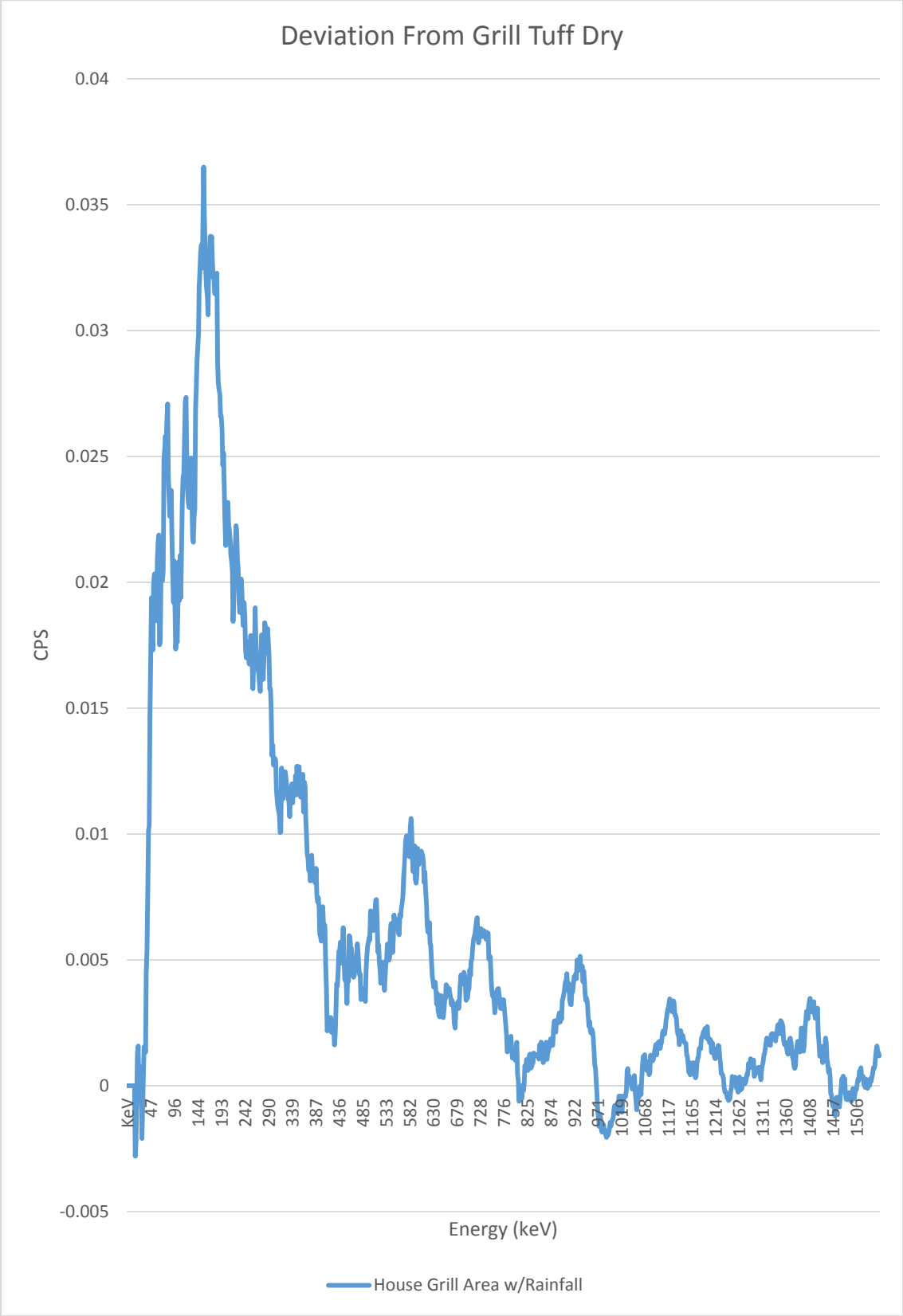


Figure 4-19b: Deviation in CPS from Dry Site to Site w/Rainfall

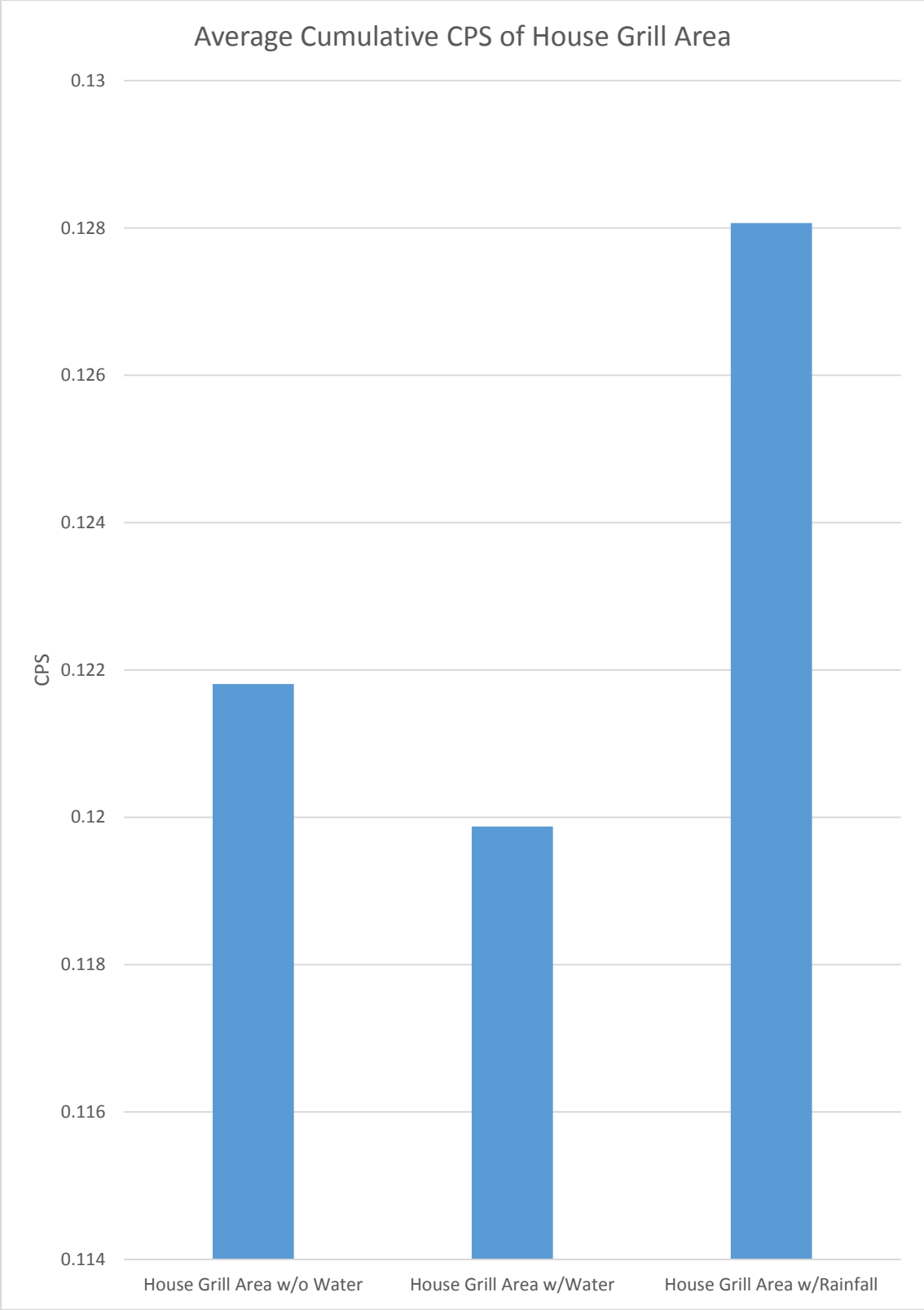


Figure 4-20: Grill Tuff Surveyed Cumulative Average CPS

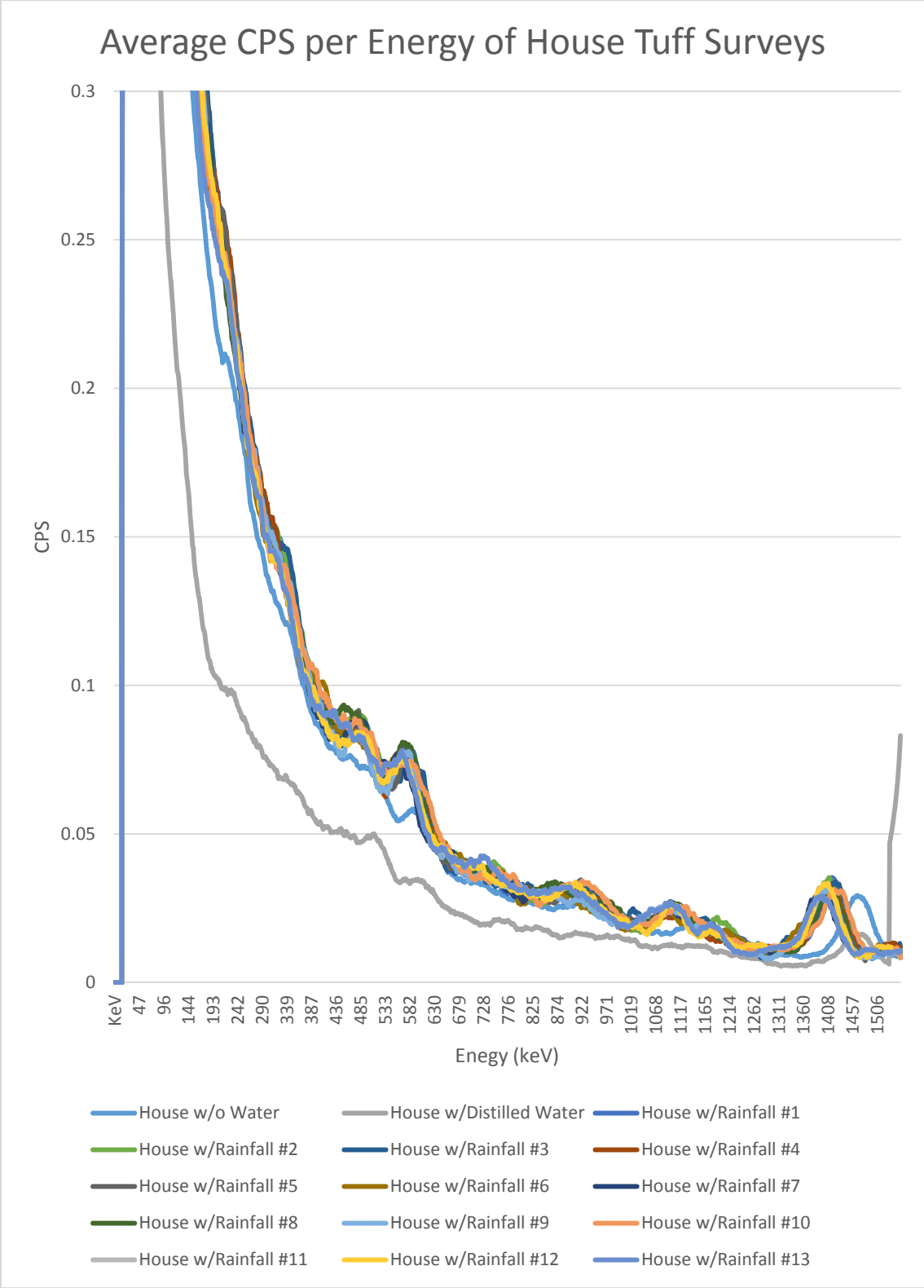


Figure 4-21a: House Tuff Surveyed Average Counts per Second per Gamma Energy Compared to Dry Conditions

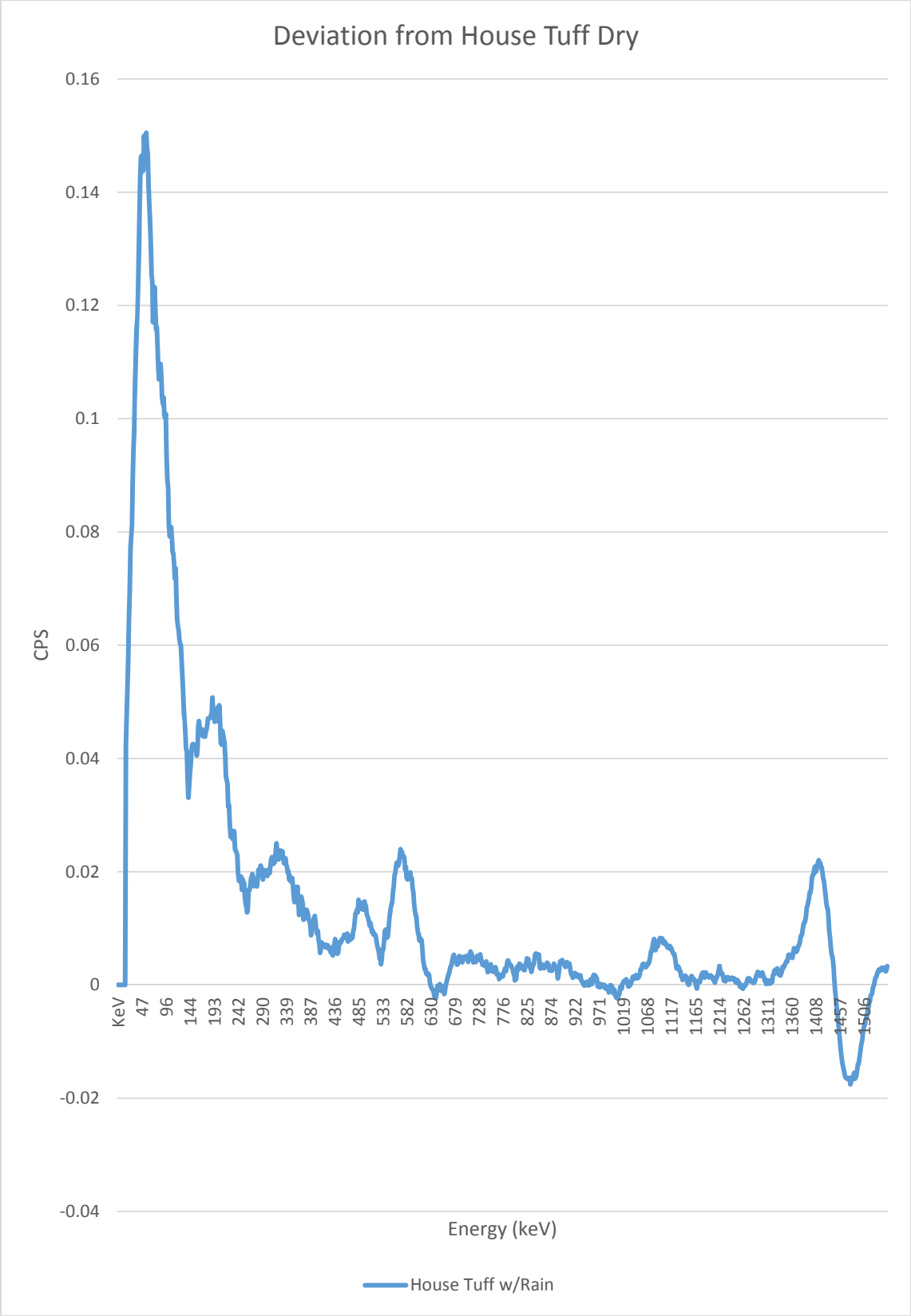


Figure 4-21b: Deviation in CPS from Dry Site to Site w/Rainfall

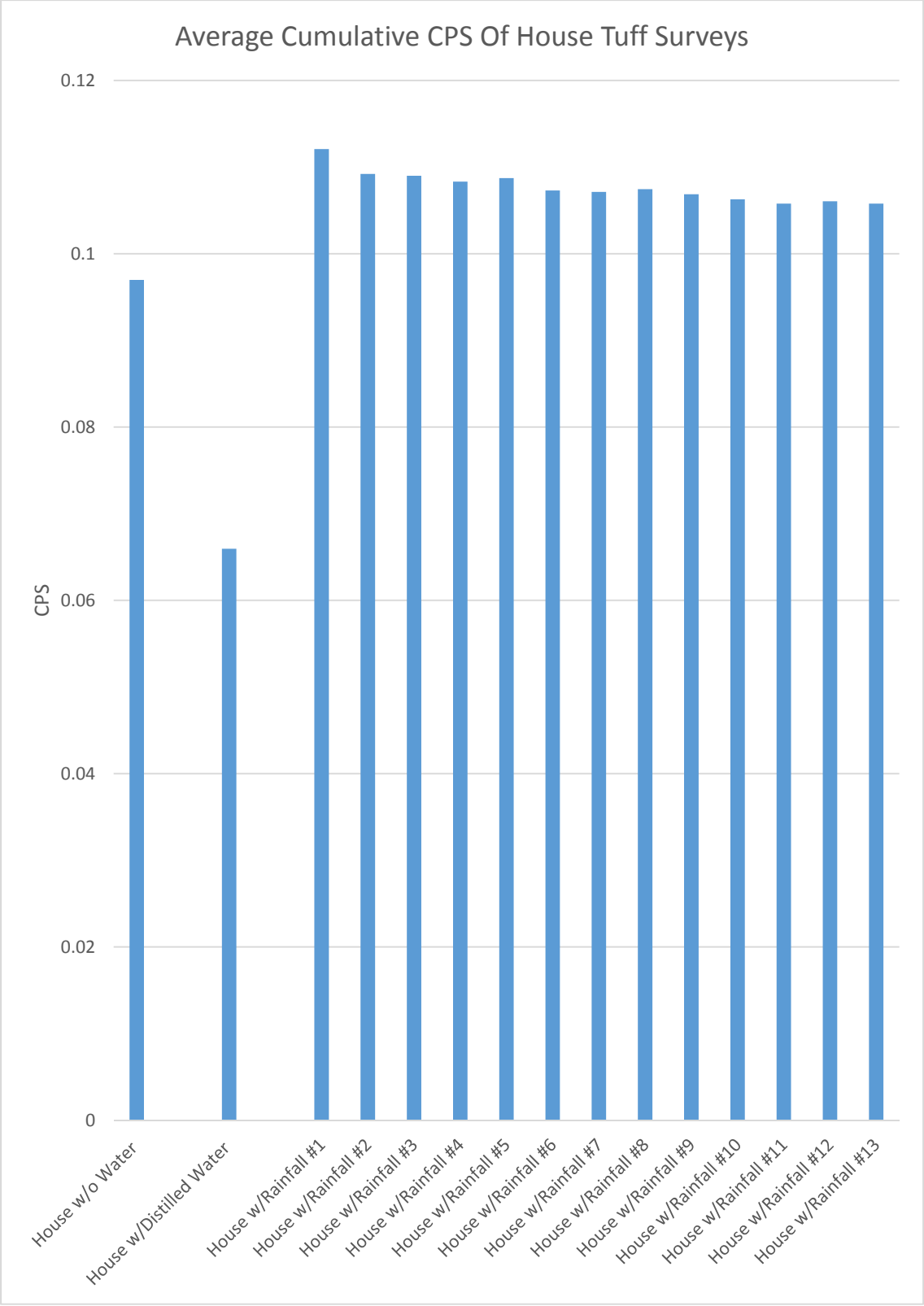


Figure 4-22: Housel Tuff Surveyed Cumulative Average CPS

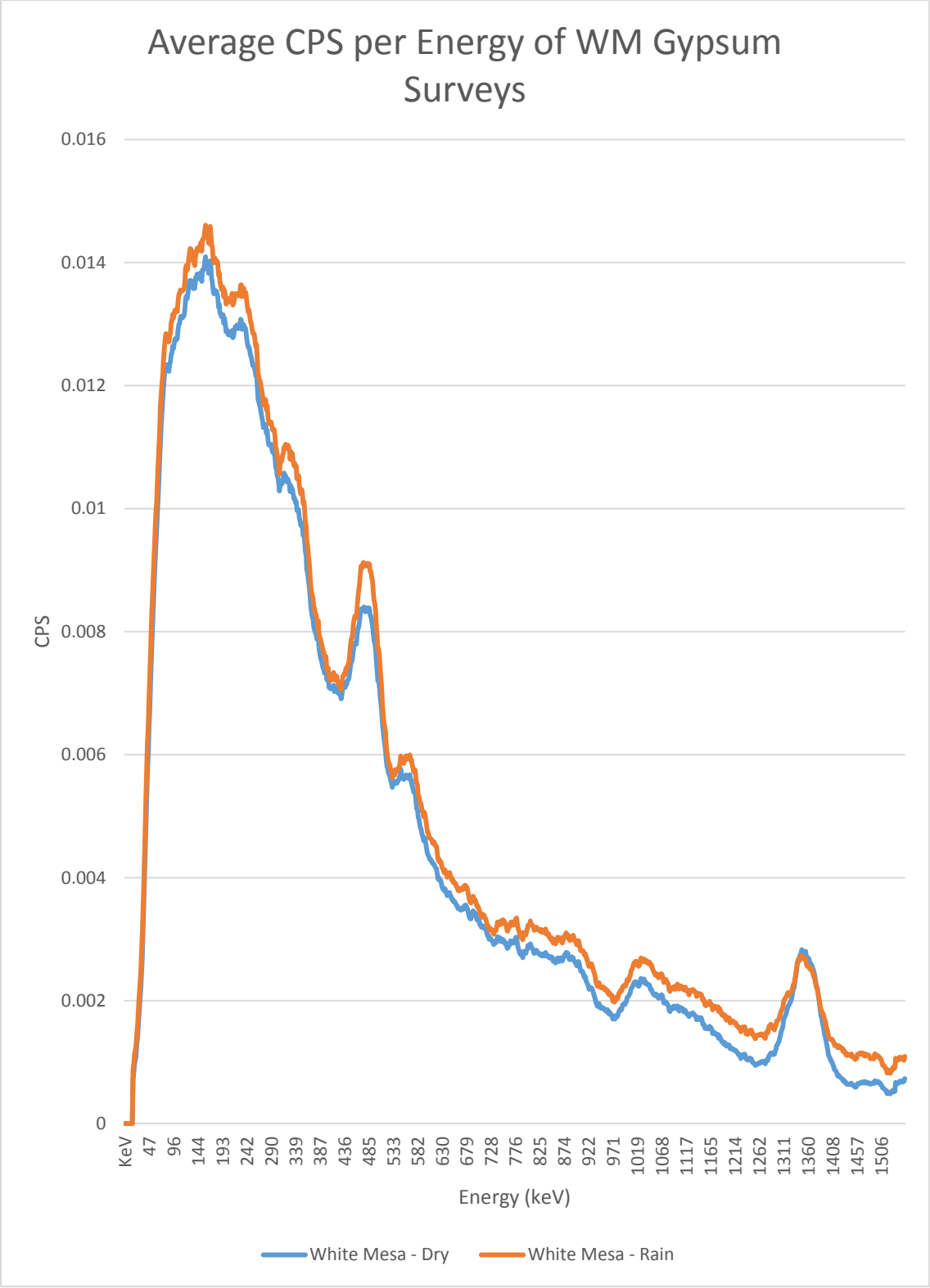


Figure 4-23a: White Mesa Gypsum In-Situ Surveyed Average Counts per Second per Gamma Energy – Rain vs Dry

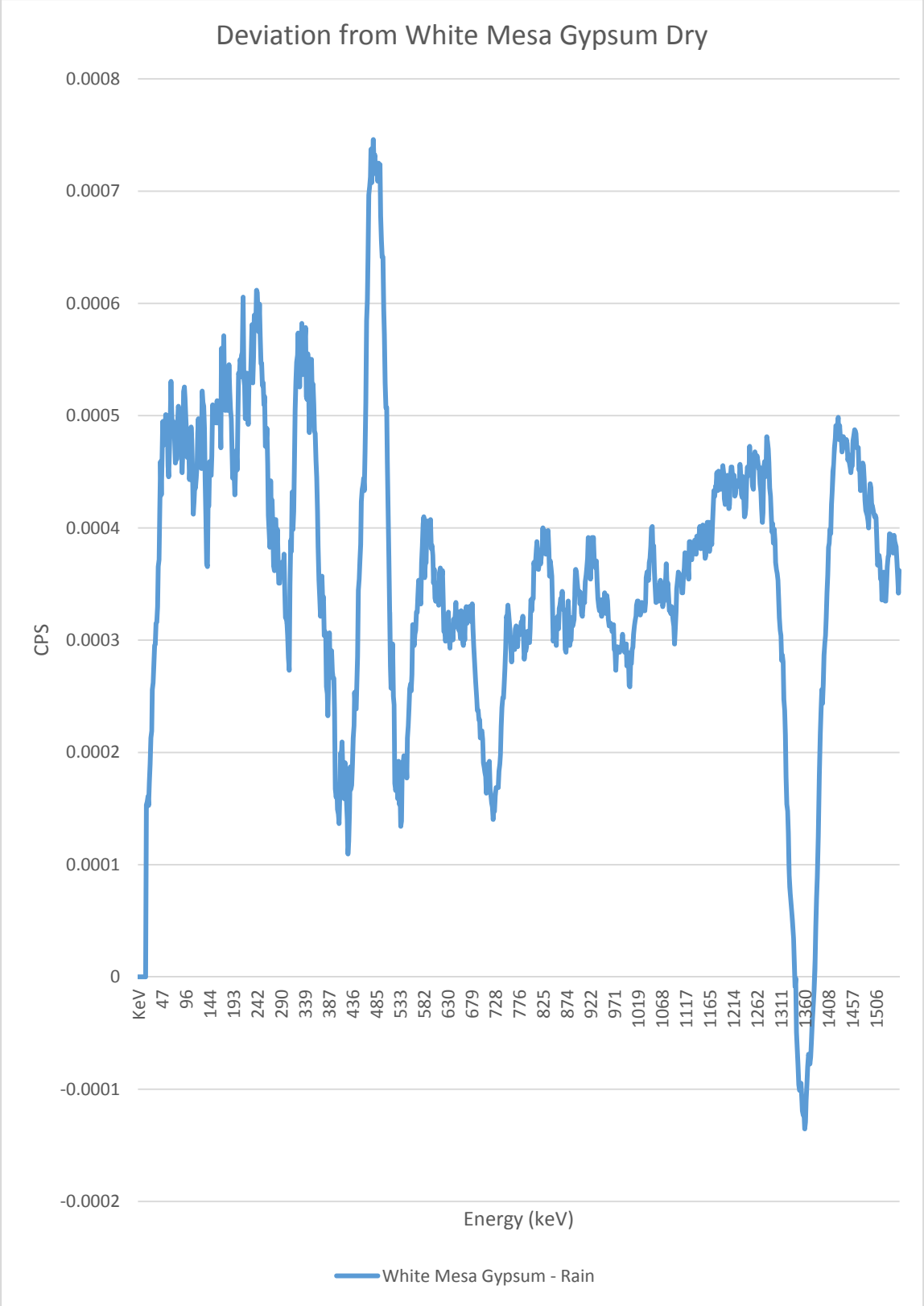


Figure 4-23b: Deviation in CPS from Dry Site to Site w/Rainfall

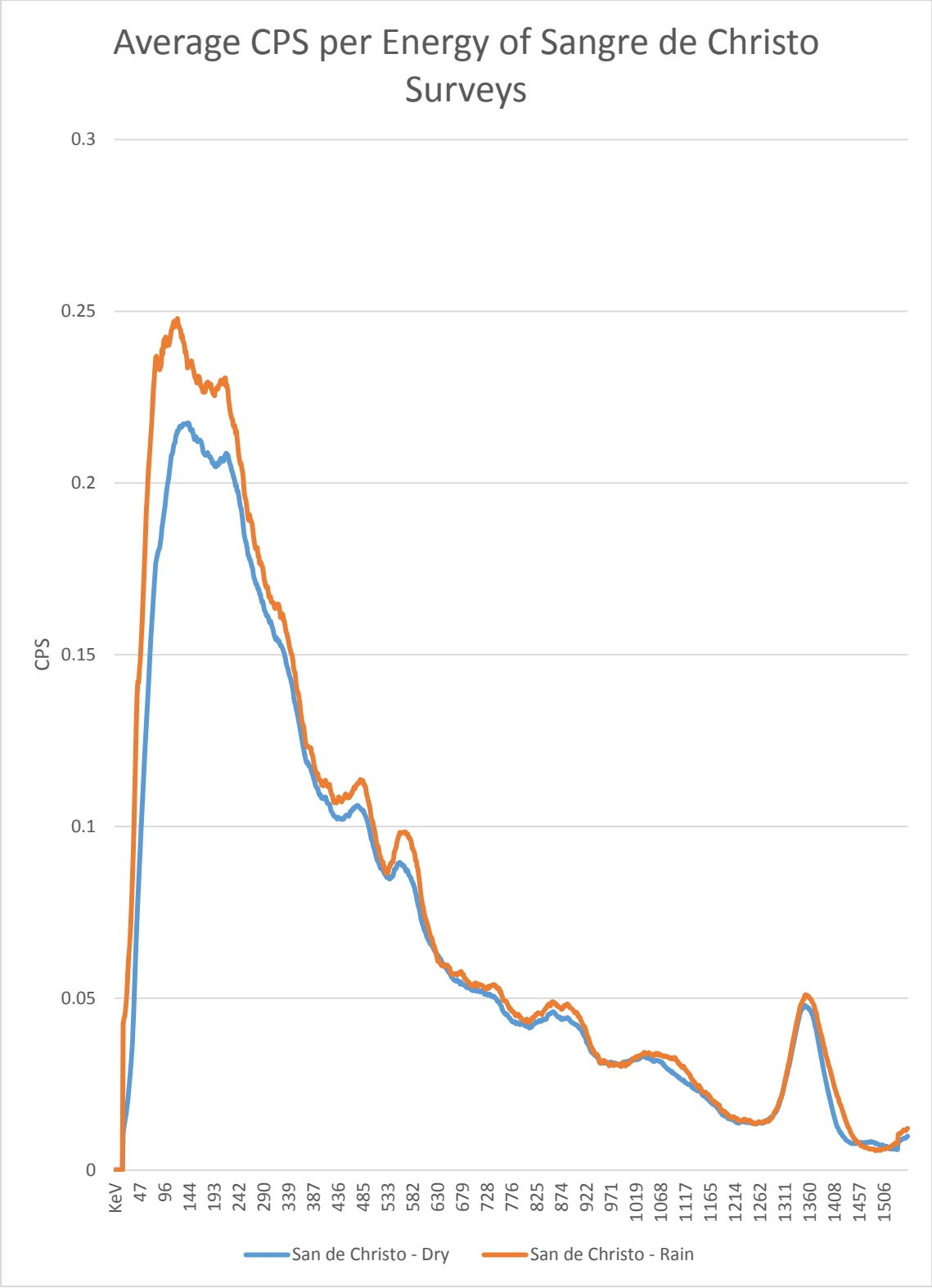


Figure 4-24a: SdC In-Situ Surveyed Average Counts per Second per Gamma Energy Compared – Rain vs Dry

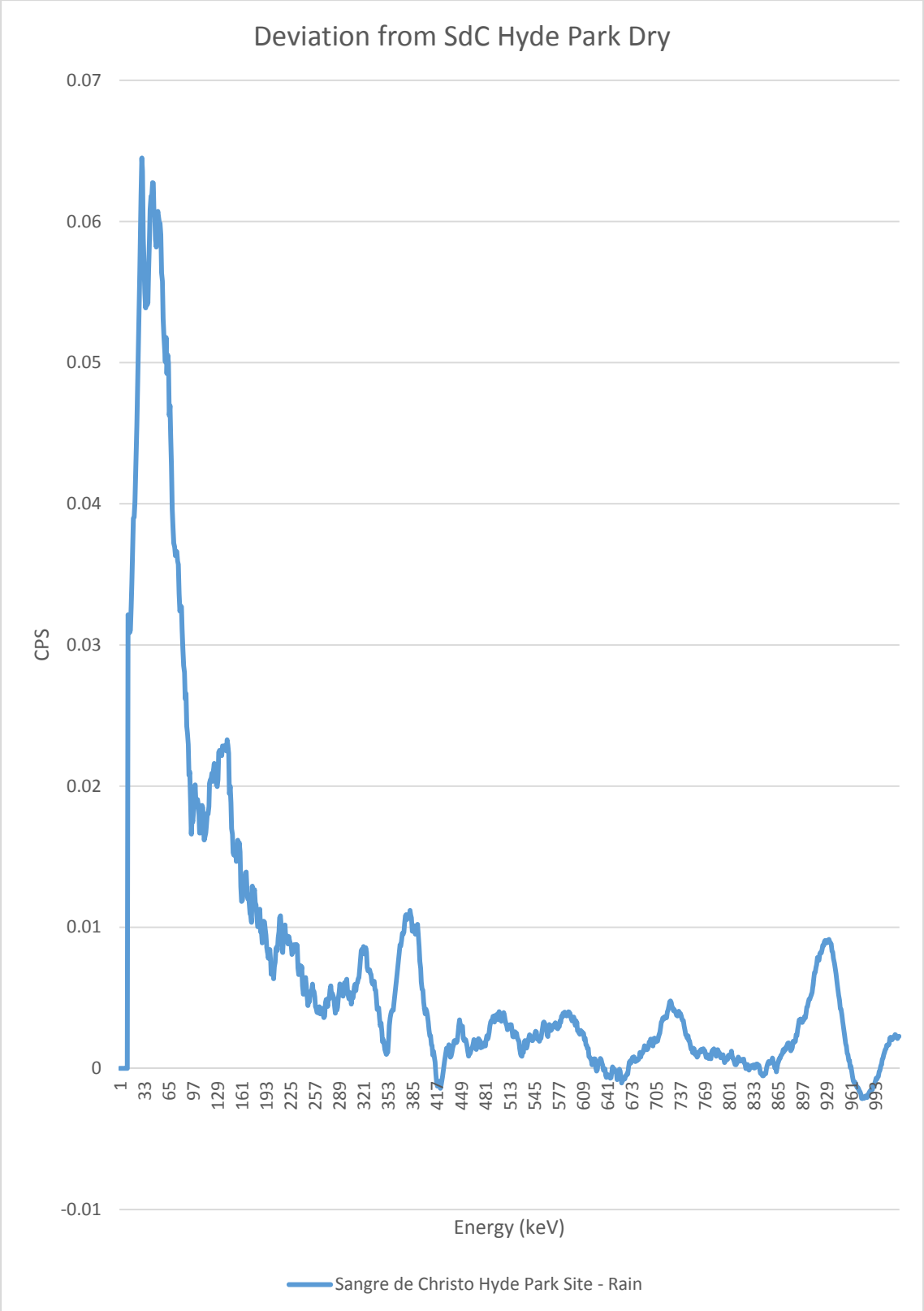


Figure 4-24b: Deviation in CPS from Dry Site to Site w/Rainfall

Chapter 5: Interpretation

The results of the surveys presented in Part 4 are based on grouping similar sampling methods for each sample. The graphs present the average counts per second per gamma energy for each experimental condition and the same CPS of the enclosure environment. An experimental condition would include the dry sample, or the sample with 800cm^3 of water, etc. Everything is presented relative to the enclosure environment and to the bare sample, because we are attempting to determine differences from the background combined with the bare sample. This is to determine if a water has an impact on the emissions spectrum, the average cumulative CPS, the average CPS per gamma energy, or a combination of the three. This section begins with examining how well the enclosure and the surrounding water were able to attenuate outside radiation. Following that, I will examine results from the surveys of the samples themselves, the samples combined with the different waters, and finally the samples combined with rainwater. Finally, I will analyze and interpret the data gathered from *in-situ* surveys. This includes the first rainwater data from Hawaii, followed by the rainwater data from surveys taken in New Mexico.

Enclosure Apparatus & Controlled Environment Survey Results

Interpretations

The background emissions that are present in the environment of the enclosure are relatively high. This is not ideal for determining small differences from one experimental condition versus another. Because of this, the attenuation due to the enclosure and the

shielding water are of the utmost importance. With the introduction of the enclosure apparatus, as well as the insertion of the enclosure into the water-filled container, there was a significant decrease in surveyed gamma emissions. The enclosure itself was effective in reducing the emissions by 26%. Placing the enclosure in the container, and then filling the container with distilled water results in a further 26% reduction in background emissions. Therefore, the final average cumulative CPS for the enclosure environment is equal to about 65 CPS. It is worth noting that when adding the enclosure and the water, there are no additional peaks in the spectrum, only a decrease in the peaks that were present from the background. The combined attenuation of the enclosure and the water results in a 49% change in background emissions as compared to the open environment. The 49% reduction equates an average cumulative CPS of 65. This is the background that all enclosure samples were measured against, and any change will be compared to this.

When the waters are introduced to the enclosure, there is very little change in CPS with the introduction of the water. From ***Figure 4-3*** it is possible to see a small reduction in averaged total gamma CPS. The average cumulative CPS changes from 0.5% to 1%. However, the change is always a decrease in average cumulative CPS, as well as average CPS per gamma energy (***Figure 4-2***), relative to the enclosure background. While the change is small and variable, there is always a reduction in CPS, including in later surveys of the samples combined with the same waters. Also, of note, there is very little discernible difference between each emission spectrum, except for a slight change to some of peaks' energy.

The first sample site that was surveyed was gypsum from the White Mesa area. Three samples were gathered from the overlook near a USGS survey marker, at the head of the White Mesa valley. The samples were crushed and sieved, resulting in a very fine powder and some small grains. The combined emissions spectrum of those surveys is shown in ***Figure 4-4***. The sample itself raised the total CPS by less than an average of 1%. With the introduction of the waters, the CPS dropped to within 0.1% of the experimental background. The small decrease is in line with the earlier data (***Figure 4-2, Figure 4-3***) showing a small decrease with the addition of the water. There were also not any apparent reactions that resulted in additional gamma energy being released. These reactions would be apparent by increased average cumulative CPS or additional gamma rays releasing at a specific energy, which would be visible through an additional peak in the spectrum. Also, there were no reactions visible to the naked eye, either, suggesting that the waters do not react with the samples. This is true for all samples surveyed. However, with the introduction of the first rainwater, there is a nearly 10% increase in average cumulative CPS relative to the experimental apparatus background, and a general increase in average CPS per gamma energy. ***Figure 5-2*** shows the increase with the introduction of the rainwater relative to the other samples also with rainwater added, and to the baseline enclosure background.

The second sample site that was surveyed was sand from the White Mesa Area, with results presented in ***Figure 4-5***. The surveys resulted in an only slightly more than a one percent change relative to background, with a decrease in CPS once the waters, excepting rainwaters, were introduced. An *in-situ* survey (Milazzo, 2016) of the same

site resulted in an average cumulative CPS 4 times that of gypsum, but the surveys in the enclosure resulted in a similar cumulative CPS and average CPS per gamma ray energy as the gypsum (***Figure 5-1)***, both with the waters and without. With the introduction of the first rainwater sample, which was composed of the same waters from the gypsum surveys, there was an 8% increase in CPS (***Figure 5-2, Figure 5-3)***. When the second rainwater is introduced, there is a slight increase (3.5%) in the average cumulative CPS relative to the same rainwater in the gypsum (***Figure 5-2, Figure 5-3)***. As with the gypsum, there was no indication of any other reactions occurring with the introduction of the waters that lead to gamma emissions, or any reactions occurring at all that could be visually identified.

The Albuquerque Volcanoes' samples were next to be surveyed. These are a basaltic lava and formed about 150,000 years ago (NPS 2018). The results are very similar to the White Mesa sand, with a slight increase in the CPS relative to experimental background of about 1% (***Figure 4-6***). This is more consistent with the *in-situ* survey results, which shows a similar average cumulative CPS for the White Mesa Sand and the Volcanoes but is not representative of the difference between the basalt and the gypsum (***Figure 5-1***). These differences, while small, indicate that the detector is registering additional gamma rays being emitted from the sample. While 1% is a small difference, there is a constant increase, averaging 1%, with some surveys registering slightly more, and some slightly less. Following the introduction of the waters, there is a slight decrease in the average cumulative CPS and the average CPS per gamma ray energy. This decrease percentage is like what is seen with the prior samples, and continues to be similar all

samples tested, resulting in a decrease of 0.10% to 1.0% in the average cumulative CPS and a varying decrease in average CPS per gamma energy. With the introduction of the first rainwater, there is a 5% increase relative to background, which is roughly three percent lower than the previous surveys of the White Mesa sand. With the introduction of the second rainwater, there is about a 3% increase relative to background but a decrease relative to the sand and the gypsum rainwater surveys (***Figure 5-2, Figure 5-3***).

The Madera limestone was also surveyed near Sandia Peak. The average CPS (***Figure 4-7)*** relative to background was about the same as the Albuquerque West Mesa volcanoes (***Figure 5-1***), including the samples with the waters added. The increase in average cumulative CPS was present, as in other samples when rainwater was introduced, but the change is smaller relative to the change with the earlier samples (***Figure 5-2***, ***Figure 5-3***).

Next to be surveyed were the samples from Lamy Park, and from the Galisteo basin. All the samples were from areas listed on the New Mexico geologic map as Holocene alluvial deposits (NMGMR 2003). These deposits increased the CPS (***Figure 4-8)*** relative to the background by about 2%. This 2% increase is double the increase of the gypsum, basalt, and limestone (***Figure 5-1***). This is consistent with the *in-situ* surveys (Milazzo, 2016), with these locations having double the average cumulative CPS compared to the basalt and sand. The increases are likely caused by an increase in concentration of naturally occurring radionuclides present in the lithologies. This is true for all the lithologies, and is being recognized in the surveys, with greater concentrations emitting more gamma rays, and lower concentrations emitting less. With the addition of the

waters, there was a decrease in CPS between half a percent and roughly one percent. As with the other samples, there weren't any indications of reactions releasing gamma radiation. These samples were exposed to rainwater collected on August 9th. This was the first significant rainfall after two weeks of little to no rainfall and breezy conditions. The Lamy samples' surveys resulted in an average of more than 6% over background, while the Galisteo samples averaged near 5.5% (***Figure 5-2***). The decrease in the increase of average CPS from one group of surveys to the next continued (***Figure 5-4***).

The Sandia Foothills samples were surveyed following the Lamy and Galisteo samples. The average CPS was about 4% over background, with the metamorphic rock coined "Country Rock" registering a bit lower than the granite (***Figure 5-1***). This decrease likely reflects a decrease in concentration of the naturally occurring radionuclides uranium, thorium, and potassium. Once the waters were introduced, the CPS decreased by the expected half percent to around one percent. The first rainwater sample was newly collected on August 15, 2018 following a slightly drier period with little significant rainfall. This rainwater resulted in a nine percent increase over background, or a five percent increase over the "Country Rock", and an average cumulative CPS three percent greater than the bare granite. The "Country Rock" was also sampled with the same rainwater collected on the 9th as the Lamy and Galisteo samples and had a CPS of slightly more than five percent over background emissions (***Figure 5-2***). The granite was not exposed to a second rainwater (***Figure 5-4***).

The penultimate surveys were of samples collected in the Sangre de Christo Mountains to the north of Santa Fe. Their average CPS was almost 6% over the background CPS,

again reflecting an increase radionuclide concentration **(*Figure 5-1*)**. While there is a 6% increase in average cumulative CPS, this is not relative to the difference seen in *in-situ* surveys. The difference in CPS was 20 times that of the White Mesa Gypsum (Milazzo, 2016). This large difference between survey and *in-situ* measurements may be explained by the small volume of sample material. An *in-situ* survey is receiving a signal from at least ten times the volume of material that is in the enclosure. With a much greater volume, a greater number of gamma radiation is emitted. With the various waters added, a reduction in CPS of not more than one percent was recorded, as has been the case with all surveyed samples. The rainwater used in the analysis was collected on September 2nd, with no rain the week prior. The surveys resulted in a CPS of around ten percent higher than background. The surveys were around four percent higher than the sample alone (***Figure 5-2, Figure 5-5***).

The final sample site and surveys were from the Abiquiu Tuff. This tuff emitted the highest level of gamma emissions of any of the samples but is still not representative to the *in-situ* surveys, again likely due to the small volume of sample compared to the volume of sample in an *in-situ* survey (***Figure 5-1***) (Milazzo, 2016). Its CPS was more than seven percent higher than the experimental background emissions, again reflecting an increase in the concentration of naturally occurring radionuclides. With the introduction of water to the samples, a reduction in CPS of not more than one percent occurred, which is consistent with every other samples' surveys. With the introduction of the rainwater samples to the tuff, a two to three percent increase in CPS occurred

(***Figure 5-2***). This is the same water collected and used with the Sangre de Christo surveys, and is slightly lower than the results from those surveys (***Figure 5-5***).

Final Interpretations of Enclosure Surveys

For every survey in the enclosure of the samples collected from each site, there is a discernible difference between the enclosure without the sample, and the enclosure with the sample. This difference varies from less than one percent for the White Mesa gypsum, to more than seven percent for the Abiquiu Tuff. This is also the case when the waters are introduced. The average CPS always decreases, although the decrease may be less than half a percent. There is variability from survey to survey of the same experimental condition, but it is always a decrease. When the rainwater is introduced, the difference becomes more pronounced. It is often contributing more to the overall gamma radiation being emitted than the sample itself, and only once a relatively strongly emitting material such as granite or tuff is surveyed, does the difference in emissions become more even. There are also clearly recognizable peaks present through nearly every emission spectrum. These peaks are typically at about 490, 600, and 1460 KeV, with additional peaks in some of the surveys, and an occasional shift in gamma energy of these peaks. There are also less defined peaks throughout. As mentioned earlier, these peaks correspond to known gamma rays emitted from naturally occurring radionuclides and will be more fully explained in the Chapter 6.

In-Situ Survey Interpretations

The enclosure surveys were undertaken to minimize variables and to isolate the sampled lithologies and experimental conditions from outside influence. This was done to more accurately gauge the impact that each experimental condition had on the lithology, to more accurately attribute the results to the condition itself. While ideal, the experimental environment is not where this research will prove most practical. Therefore, *in-situ* surveys were also performed. These surveys began with a trip to the Kona Island of Hawaii. Following that, surveys were performed during thunderstorms in the White Mesa Gypsum, the Sangre de Christo mountains, and multiple areas in Los Alamos County.

The Hawaiian rainwater surveys were compared to the dry background of the same area, and every rain survey produced a measurable increase in average cumulative CPS (***Figure 4-18***) and average CPS per gamma energy (***Figure 4-17***). The most notable result is the 27% increase in cumulative CPS relative to the averaged dry surveys. The survey location was downwind of the 2018 volcanic eruptions on the northeastern corner of Kona. The smallest increase was 3%. Comparing the rain surveys to dry surveys of the same location, the increase in average CPS was as much as 12%, and as little as 1%.

Upon commencement of rainfall in New Mexico in July of 2018, surveys were taken in Los Alamos County, the Sangre de Christo mountains, and White Mesa (***Figures 4-17*** through ***Figure 4-24***). These surveys differed from enclosure surveys in that many of the surveys had peaks that were more distinct than those seen at the same energies in the survey enclosures of the same lithologies. This is likely caused by the much greater quantity of material being surveyed, and a much greater quantity, not concentration, of

radionuclides present. In the surveys of the White Mesa gypsum (***Figure 4-24***), there is a small trough in the data, which is not present in any other survey. However, like the enclosure surveys, the addition of water to the surveyed area decreased the average cumulative CPS (1.5%) and the average CPS per gamma energy. Like all surveys, the decrease was more significant at gamma energies below 650 KeV, while at higher energies the decrease become difficult to discern from the graph.

Like the enclosure surveys, with the addition of rainfall to the soil/sediment, there is an increase in average cumulative CPS, and average CPS per gamma energy. The change in CPS was as little as a 5% increase, and as much as a 15% increase. During a rain event, a long duration survey period of 75 minutes, broken into 5-minute intervals, resulted in a 15% increase compared to background, that decreased to a 9% increase in background at the end of the survey period (***Figure 4-21***). This was notable for the large increase over a relatively high background CPS, as well as a decrease in the increase during the rain event. Examining the graph, there is also a leftward shift in the peaks as the survey period continues.

The survey of the Bandelier tuff, labeled "Grill Tuff" is also notable (***Figure 4-19***). Unlike all other *in-situ* surveys, there was no prior collection of the rainfall, nor was there rainfall in the bucket. The survey was begun shortly after the rain event began, and continued for a 25-minute period, with survey interval of 5 minutes. The result of the survey was a 5% increase in average cumulative CPS and an increase in average CPS per gamma energy relative to the same area surveyed dry. Also, the results presented do

not reflect the larger increases at the beginning of surveying, and the continual decrease in the CPS's as the surveys continued.

Final Interpretations of the *In-Situ* Surveys

The results from the *in-situ* surveys also show similar trends to those of the enclosure surveys. There is a decrease in the average cumulative CPS and generally, the average CPS per gamma energy with the addition of water. There is also an increase in the CPS during a rain event. The *in-situ* surveys differ in that some locations exhibit much larger increases in those same numbers than the enclosure surveys. The "House Tuff" is a prime example. They also differ in that there are often 5 or more discernable peaks in the *in-situ* surveys, rather than the 3 to 4 typically seen in enclosure surveys. This can likely be attributed to the greater quantity of material being surveyed and therefore a greater quantity of radionuclides present. With the greater quantity present, the number of gamma rays emitted increases, and that results in larger, more distinct peaks. Several of the surveys that continued for extended periods also exhibit a shift in the location of those peaks, which is not the case with the enclosure surveys.

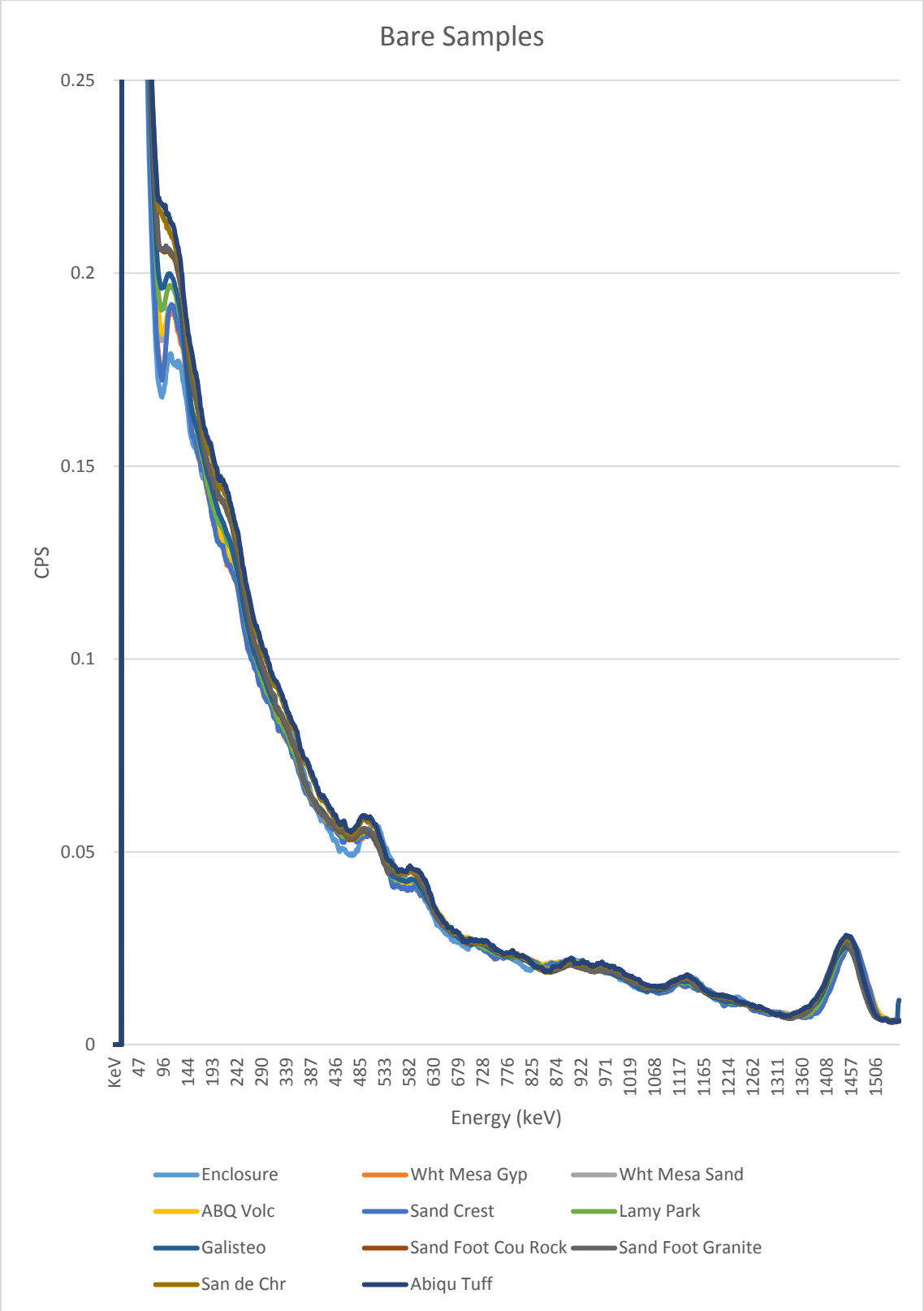


Figure 5-1: Bare Samples Average CPS per Gamma Energy Compared to Enclosure CPS

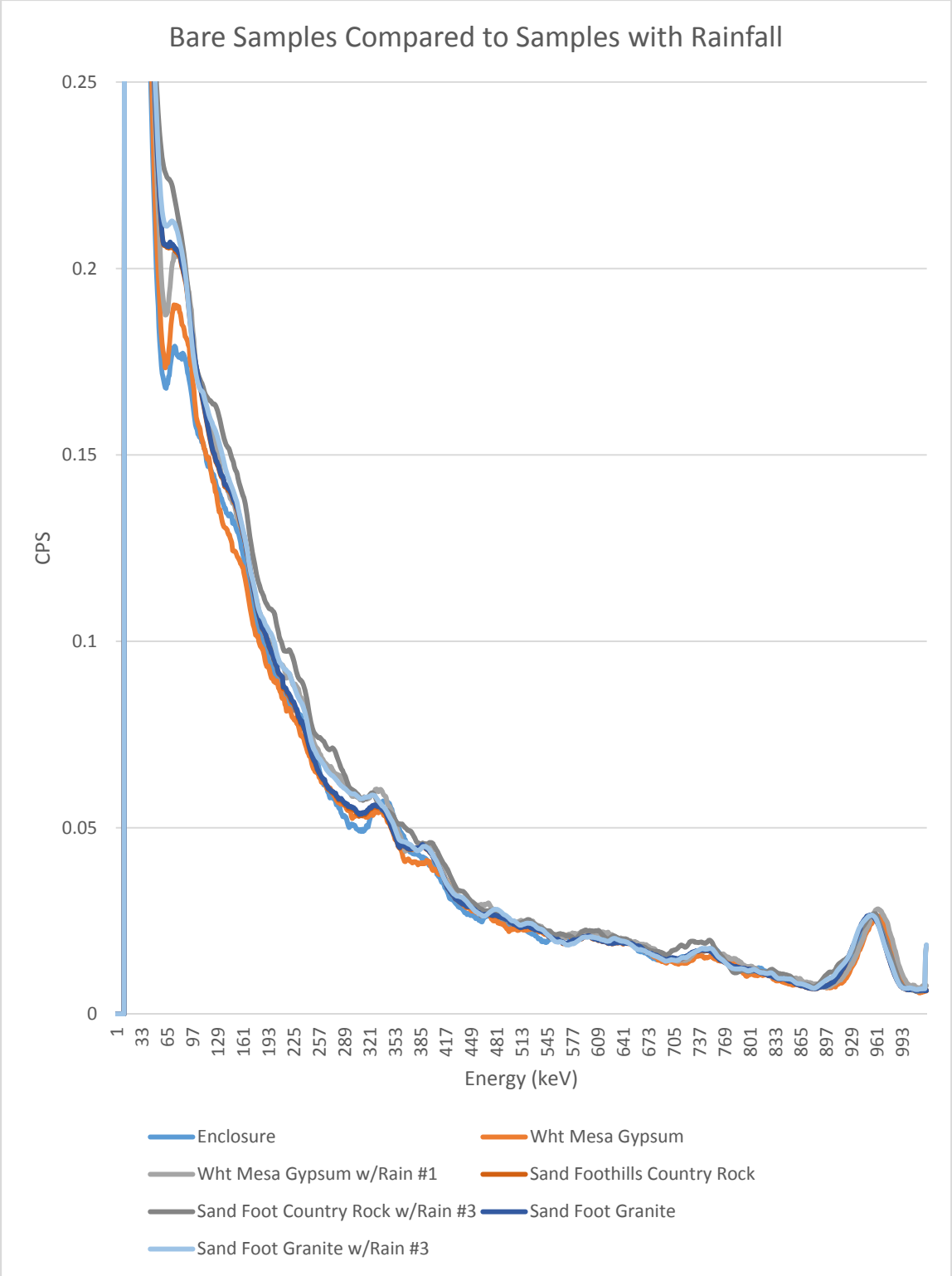


Figure 5-2: Bare Samples Average CPS per Gamma Energy Compared to Samples w/Rainwater

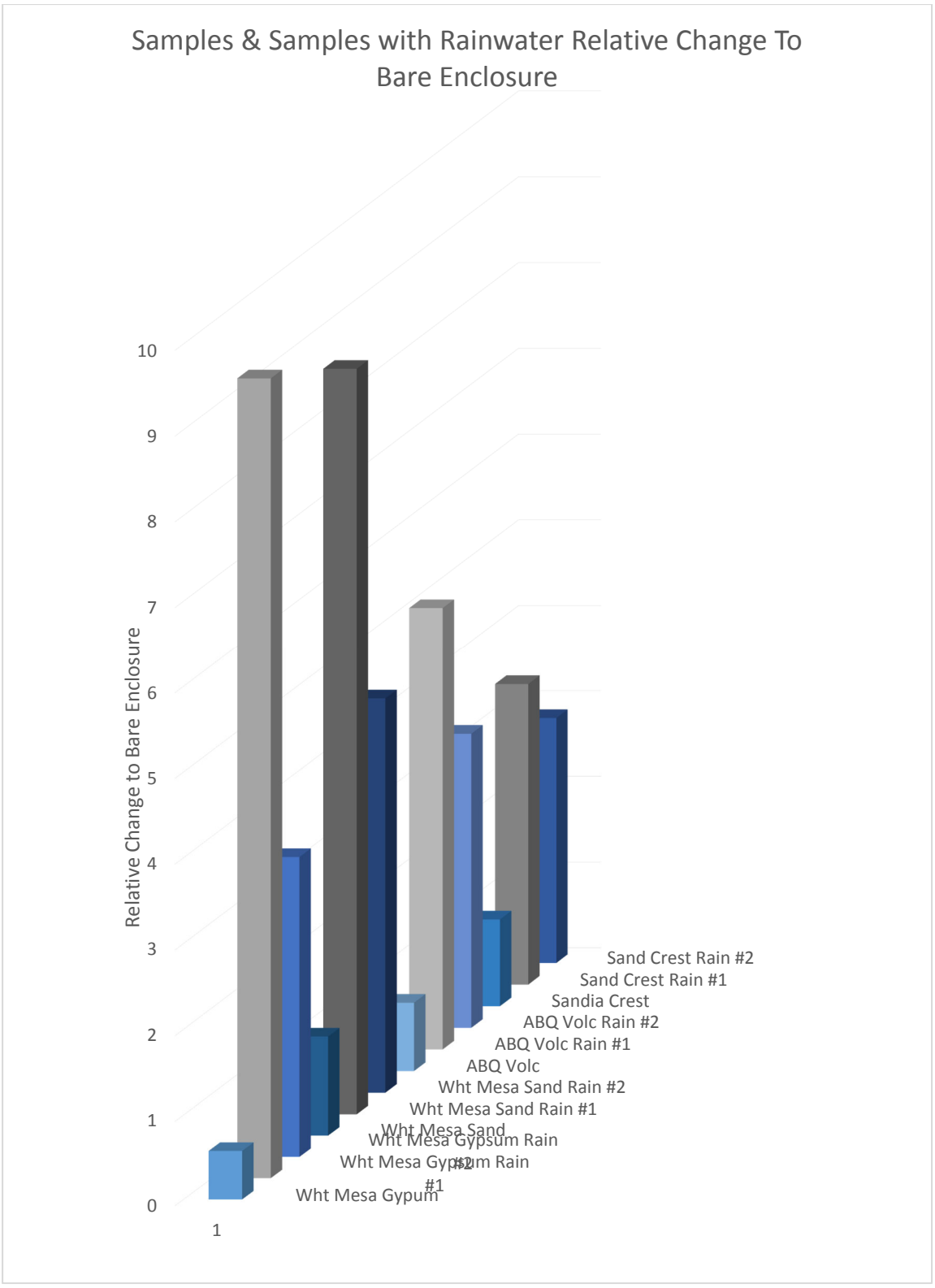


Figure 5-3: Bare Samples and Samples with 1st Rainwaters Relative Change to Enclosure and Subsequent Samples

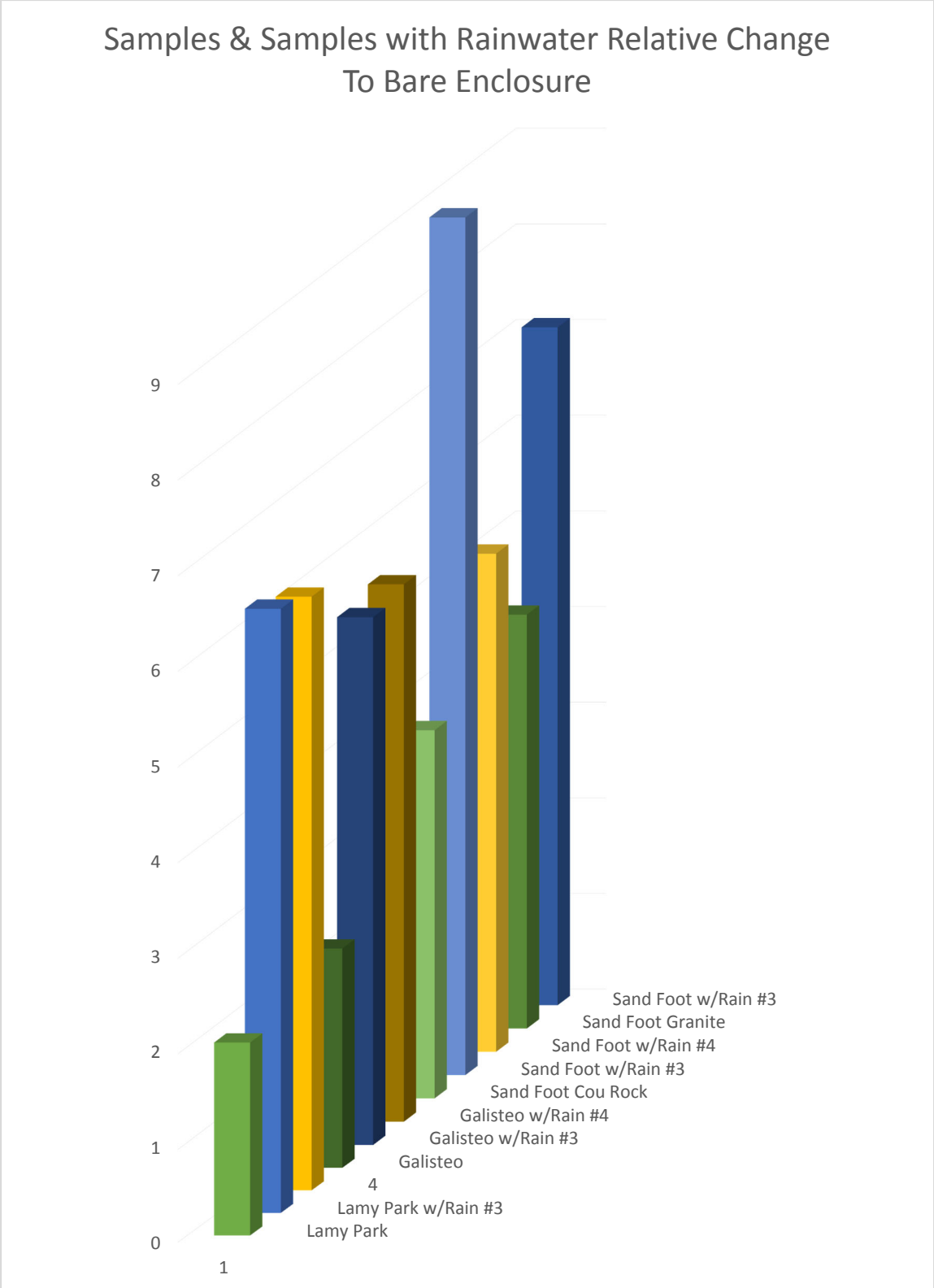


Figure 5-4: Bare Samples and Samples with 3rd/4th Rainwater Relative Change to Enclosure and Subsequent Samples

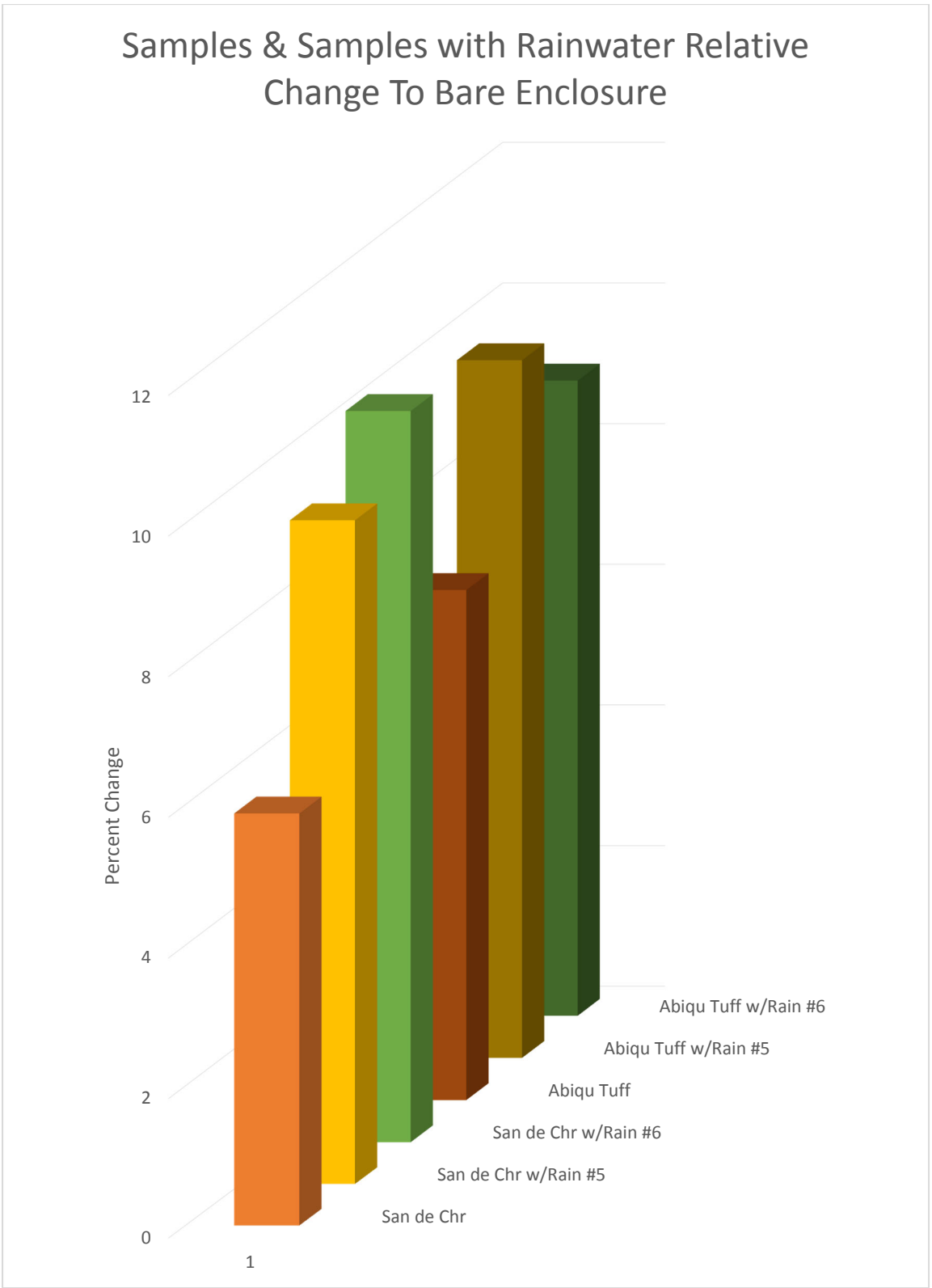


Figure 5-5: Bare Samples and Samples with 5th/6th Rainwater Relative Change to Enclosure and Subsequent Samples

Chapter 6: Conclusions

Examining the results of the surveys, a number of conclusions may be made, beginning with the enclosure apparatus. The enclosure was built with the intention of using lead shielding to prevent the intrusion of gamma energy. Lead is an extremely effective attenuator of gamma energy, especially for its cost (McAlister, 2018). Rather than utilize lead, the enclosure was instead placed in a large container (50 gallons) full of distilled water. This water helped to attenuate a further quarter of the background radiation. However, the same effect was attained with a relatively thin shielding of steel plates, which were responsible for the first quarter of attenuation. This would suggest that water is a poor attenuator of gamma radiation, which is supported by McAlister (2018), amongst others (Nelson, 1980; Ugdani, 2013; Zeb, 2016). This was also supported by the minimal reduction of gamma radiation evident from the results of the surveys that occurred when the water was introduced to the sample holder in the enclosure, and when it was incorporated into the samples. In fact, to attain a similar degree of attenuation from water as from steel, the water needs to be nearly 9 times as thick, depending on the energy of the gamma ray (McAlister, 2018). However, at high energies of 650 KeV or greater, the difference in the degree of attenuation decreases due to the nature of high energy gamma radiation and the way it interacts with matter.

With the survey of the enclosure, the placement of the enclosure into the container, and the introduction of the waters to the samples, there was the noted small decrease in attenuation. What was not there was any apparent increase in average CPS, nor average CPS per gamma energy. This suggests that neither the enclosure nor the waters

contain any gamma emitting radionuclides. Perhaps more accurately, this suggests that while Gainon (2007) and Ajayi (2015) found that waters may contain radionuclides, the concentrations present in the tested waters are low enough to no contribute enough gamma radiation to be measured, nor to outweigh the degree of attenuation that the waters cause. In fact, there was very little variation from one water to another, rainfall excepting. There were also no apparent chemical or energetic reactions that resulted in measureable gamma emissions either, suggesting any reactions that did occur did not produce gamma radiation. These surveys are not alone in finding this negative result, as work performed by Drndarski (1988), Stralberg (2003), Avwiri (2007), etc., never mentioned any interactions of waters and soils producing measurable gamma radiation when mixed.

The samples' average cumulative CPS and average CPS per gamma energy, relative to other samples in the enclosure, should be proportionally representative to their average cumulative CPS and average CPS per gamma energy of their *in-situ* surveyed numbers (LANL 2016). If gypsum has an *in-situ* surveyed average cumulative CPS of 7 CPS, and granite had a measured CPS of 70, the samples in the enclosure would be separated by a factor of 10. It may also be expected that if the measured *in-situ* CPS of gypsum is 7 CPS, that it would contribute an additional 7 CPS to the surveyed reading of the sample and enclosure. This did not prove to be the case, and while there were increases dependent on the sample, they were not proportional to their *in-situ* measurement. This may be caused by a number of factors, such as detector efficiency and the statistically random travel paths of those gamma rays but is primarily caused by the

small volume of sample as compared to the large volume of the enclosure environment. The *in-situ* surveys record a volume of material 20 times the volume of material placed in the enclosure, and in the enclosure setting, the sample makes up only tenth at best, of the total sample volume.

In the case of the gypsum, there is also very little expectation of it contributing radionuclides, as gypsum is composed of calcium, sulfur, hydrogen, and oxygen. There are few naturally radioactive isotopes of these elements, with the sole radioactive element of calcium being calcium-47, with an abundance of only 0.003 percent (IAEA 1963). The small increase in average CPS seen during the surveys taken in the enclosure may also be influenced by the concentration of calcium in the gypsum, which is known to have a strong effect on attenuation of gamma energies below 165 KeV (Al-Masri, 2013). This attenuation is not obvious, and may be due to the small sample volume.

Other samples were not characterized in terms of their chemical composition due to the focus being on differences between rainwater and other waters, not differences between lithologies. However, generalizations may be made based on what they are and where they were gathered from. An eolian sand is likely to be composed of quartz, while a New Mexico granite is likely to be composed of feldspar. Quartz is not radioactive, while the potassium-40 in a feldspar is, albeit it's present in only small concentrations. A volcanic basalt is also not likely to have many radionuclides, while a tuff may contain a significant number. These generalizations were supported by the results, and an increase or decrease from one sample to the next can be seen. The results are also consistent with work by Pertsov (1964) and Shahbazi-Gahrouei (2013)

which examined the natural radioactivity that surrounds us. That natural radioactivity, as explained in those works, varies depending on the quantity of uranium, thorium, and potassium that is present in the lithologies.

With these things noted, this research's focus was not to characterize the natural radioactivity of lithologies, nor to determine what a small volume of the material will contribute to the background radiation of an environment, but to determine if rainfall contributes a measurable quantity of gamma radiation to a site's background. The first opportunity to do so came on the trip to Kona Island. At all survey locations where rainfall was present, there was an increase in average cumulative CPS when surveys were performed during the rainfall. This increase varied significantly, from 3% to more than 12%. These increases continued when rainfall came to New Mexico, both when rain samples were placed in the enclosure with a locale's sample, and when *in-situ* surveys were taken.

While all the surveys taken with rainfall showed an increase in CPS, the first rainfall sample introduced into the enclosure merits additional discussion. The first rainwater sample was collected on July 8th, which was the first rainfall since 2017. The first rainwater sample's addition to the average cumulative CPS is anomalously high (9% increase). The high CPS relative to other rainfall samples may be explained by the inclusion of dust, particulates, and gases, notably radon and its daughters, in the rain. It is unlikely to be explained by any chemical interactions with the sample. The increase in the background radiation caused by precipitation-laden radon and its daughters is well documented, notably by Livesay (2014), as well as Lundberg (2005). The inclusion of

dust and particulate is also well documented. The dust and particulate can be radioactive material from a nuclear disaster transported across the planet, as documented by Puhakka (1990), or may be the dust and particulate from eroded mountains that becomes airborne, as suggested by Shuk-ming (2014). Therefore, the large increase in CPS is likely due to the complete lack of rainfall, as well as high winds and the change in pressure associated with thunderstorms, which allowed the first rainwater sample to incorporate more of the radioactive emitters than any subsequent rainfall. By comparison, the second rainwater sample that was collected on July 23rd, showed a significant decrease in additional CPS relative to the first rainwater sample (3.5% increase).

The likelihood of the increase in CPS being caused by radionuclides is further supported by the decrease in the increase during subsequent measurements. The surveys of the White Mesa sand, using the same rainwater, resulted in an increase of 8%, relative to the enclosure, rather than the 9% increase. The third sample the rainwater was incorporated into saw only a 5% increase, and the fourth sample a 3.5% increase. This decrease from the first gypsum survey to the sand and so on is likely due to the decay of the radioactive isotopes present in the rainwater. In the case of radon, its daughters lead 214 and bismuth 214 both emit gamma radiation, and both have a half-life of about twenty minutes (Chabot 2008). The decrease may also be due to the decay of uranium 238, the most abundant uranium isotope, into its daughter products, which are also known to emit gamma radiation (Bath 2015).

This pattern is repeated across all surveys from the enclosure. A fresh rainwater sample results in a greater increase in CPS compared to the subsequent survey. The change is relatively consistent, with a decrease of 2% to 4%. The variability may be due to variations in sample preparation times, and the thoroughness in cleaning the sample chamber from one sample to the next.

While the results from the enclosure seem to confirm the veracity of my hypothesis, the results from the *in-situ* surveys go even further. The rain surveys from Kona all showed an increase in CPS relative to the dry surveys. In the location with the highest expected concentrations of particulate and dust, the increase was over 4 times greater (12% increase) than other survey locations on the island. The survey method was also shown to consistently reproduce similar values to the control area and the averaged values of all surveys, suggesting that the technique was consistent in its results.

The *in-situ* surveys taken in New Mexico only continued to confirm the results from Hawaii and from the enclosure. The surveys performed in White Mesa saw a 7% increase in average cumulative CPS, while the results from the Sangre de Christo site saw an increase of 10%. Both of these sites were surveyed during a highly energetic thunderstorm, with significant lightning and thunder. These conditions are known to produce an increase in gamma energy (Yamazawa, 2008; Wilkening, 2015). The sites were thoroughly surveyed prior to the monsoon start, guaranteeing a minimum of soil moisture. The “House Tuff” surveys saw a 15% increase in average cumulative CPS, which decreased as the rain event continued to a 9% increase, before surveying was stopped. This increase was in addition to a relatively high background count. Finally, the

"Grill Tuff" surveys resulted in a 5% increase. This survey was the most similar to how actual *in-situ* surveys would be performed. Water was not collected, and the detector was just placed over a newly wet lithology. There was also a decrease during the collection period, of about 2% over a 20-minute period.

With the consistent decrease in average cumulative CPS and average CPS per gamma energy, it is unlikely that the introduction and mixing of waters to a lithology results in the release of gamma energy. While the samples in the enclosure were small in volume, there were measurable differences between each sample, and each sample and water. At no point was there any indication that additional gamma energy was released, and in fact, when the non-rainwater waters were introduced to the sample, there was always a decrease in CPS. This continued to be the case with larger volumes of material as well. When 50 gallons of distilled water was used to saturate a 2.5m^2 area of tuff in Los Alamos, there was no measurable increase in CPS or additional gamma peaks, just a general decrease. This was true of all waters, except rainfall. With the introduction of rainfall, there were measurable increases in average cumulative CPS and generally in average CPS per gamma energy with every survey. There was variability in the increase, but it was always an increase, which assuredly confirms the hypothesis.

One other area of the results to examine are the peaks present in the emission spectrums. There is a large peak that is present from the origin of the graph, to roughly 350 keV. In this peak, dependent on survey and sample, is a distinguishable peak at 100 keV. This peak, along with peaks at roughly 500, 600, 900, 1000, and 1100 keV, can be attributed to uranium-238 and its decay products (Peakeasy, 2018). The large peak on

the left of the graph also represents the decay products from radon. This includes peaks at 240, 290, and 350 keV. These peaks may not be clearly defined, as they are very close in energy and due to the averaging methods employed in the processing of the data. There is an additional peak near the right extreme of the graph at ~1460 keV that can be attributed to potassium-40 (Peakeasy, 2018). These peaks average CPS may change depending on the concentration in the sample and in the sampled waters, as well as the effects of the waters' attenuation at a given energy. This would be reflected by a large or smaller peak being present on the graph. All of these peaks represent the decay products of naturally occurring radionuclides, and at no point was the presence of anthropogenic radionuclides detected. In the future, including an anthropogenic source would be ideal, as it would enable the practical application of this research to be utilized in determining the boundaries for detection of those types of sources. During the long-duration survey, there is a shift to the left in the location of the peaks recorded during the survey. There are also other instances of peak-shift in other surveys, but it is most noticeable in that group of surveys. This is due to the temperature change of the crystal, which experienced a change in temperature greater than 30 degrees and is a well-documented phenomenon (Hibbard 1975).

Unusual Results, and Further Refinement and Research

During the surveys, there were a few unusual results that should be mentioned. The first is the unusually high CPS of the first rainfall, which may be adequately explained by the first-rain-in-a-while theory. Another rainfall related peculiarity is the increase in average cumulative CPS from the gypsum to the sand with the second rainfall. There is a small

increase of 1.5% which I cannot explain. It may be a detector-related peculiarity, but it was not apparent with any other survey.

It was noted earlier that when non-rainwater waters are introduced into the experimental apparatus with a sample, the average cumulative CPS decreases. However, there are instances of water being introduced and the resulting surveys show an increase at a particular gamma energy. This would suggest that the water had radionuclides present, which is reflected in the graph. The presence of the radionuclides is supported by the work of Gainon (2007), IAEA (2016), etc. While this may suggest the presence of radionuclides, it may also be attributed to variability due to the spectrometer and the way it functions, as the change in average CPS per gamma energy from the bare sample to a sample with water was typically less than 0.25%. This is unlikely however, as the surveys represent a huge data population, which suggests that any change in huge population that is apparent is not due to measurement variability, but is instead caused by an actual change.

The use of an enclosure to measure the change in values is of debatable worth in these circumstances. The initial proposal called for determining the attenuation of water and of the rocks and sediments I was going to gather. This proved unnecessary, as the two subjects are well researched, and in experimental conditions with far more control than I could produce. That control, meaning a lack of background emissions, was not attainable given the tools I had. It also seems likely that the enclosure was not an adequate size to reproduce results from an *in-situ* survey. Also, while it was too small to function in that manner, with no longer being able to use lead, it was too large and

unwieldy to be mobile, defeating one of the initial purposes of it. This particular shortcoming was worked around, and in a more satisfactory manner than it being mobile. Where the enclosure did prove valuable was in determining that the waters did not react with the samples, and that they did not contain radionuclides in any measureable concentration.

In future research, the most important failure to remedy will be the lack of rainfall. This may necessitate a change in research venue. Given adequate rainfall, the surveys could all be performed *in-situ*. This would introduce uncontrollable variables, but it would also be representative of the actual practical application of this research. One such test, the "Grill Tuff" was already completed using this method, and the results were successful in supporting the hypothesis. This suggests the other methods may not be crucial, or will simply provide additional support to the work, rather than being the foundation of the work. Also, the collection of rainfall into one concentrated location is not ideal, as it leads to a concentration of the radionuclides, which would not be expected while using these methods during practical applications. The evidence for this concentration is exhibited with the long duration survey called "House Tuff". The surveys used rainfall collected from a roof, and there had not been a rain event for more than two weeks prior. It is likely the roof had a great deal of dust and particulate that contributed to the high CPS over background. This dust and particulate was concentrated into one location, rather than being spread over a much larger area as it would had the roof not been present. This may not be representative of actual concentrations of the dust/particulate in the water, and may skew the results.

As well as continuing this research in an area with greater rainfall, placing the enclosure in an environment with lower background would be beneficial as well. A high background reduces the weight that a small change has on a survey. Given a background of 5 cumulative CPS, if a sample added 1 cumulative CPS to the survey that would represent a 20% change. With the enclosure environment utilized during this research, that 1 cumulative CPS is only a 1.5% change. With all surveys having slight variability, and the results being averages of averages, that 1.5% change in cumulative CPS may be lost. This would also assist in determining if the variations seen between surveys are statistically significant. The greatest issue in making conclusions about the non-rainwater waters were the small changes from the sample and the sample combined with a water. Often the change was less than 1%, which is difficult to attribute to being statistically significant, especially with variability between dry survey samples also being close to 1%. Therefore, to increase the chances of being successful at determining differences between the samples and experimental conditions, lowering the background emissions in the environment is of paramount importance.

One additional tool to add assuming *in-situ* surveying, would be a moisture meter. While two *in-situ* locations had not seen precipitation in 4 months or more, the ability to rule out the effect of existing moisture would be ideal.

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