Analytical Inverse Model for Post-Nuclear Event Attribution of Plutonium

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ABSTRACT

An integral part of deterring nuclear terrorism is the swift attribution of any event to a particular state or organization. By quickly being able to identify the responsible party after a nuclear event, we can ensure that the appropriate people are held accountable for their actions. Currently, there is a system in place to determine the origin of nuclear devices and materials from post-event data; however, the system requires time to produce an answer within acceptable error margins. Described here is a deterministic approach derived from first principles to solve the inverse problem. We start with the basic change rate equation, and derive relationships for important nuclear concentrations and device yield. This yields a very computationally efficient method for producing an estimate of the material attributes. This estimate can then be used as a starting point for other more detailed methods, thus reducing the overall computation time of the post-event forensics. This work focused on a specific type of nuclear event: a plutonium IND explosion. From post-event isotopic ratios, this method determines the device's pre-event isotopic concentrations of special nuclear material. From the original isotopic concentrations, we can narrow the field of the possible origins for the nuclear material. In this method, knowing where the nuclear material did not originate is as important as knowing where it did. We have seen results in an ideal case (consisting of 95% Pu-239 and 5% Pu-240) within a two percent margin of error.

INTRODUCTION

With the proliferation of weapons technology to rouge nations like North Korea and Iran coupled with the increasing prevalence of terrorism around the globe, there is growing concern that a nuclear device could be detonated in the United States or abroad as part of a terrorist incident. As a result, there has been a renewed interest in nuclear forensics to determine the origin and perpetrators of such a heinous act.[1] Nuclear forensic attribution is focused not only on the origin of the material but the illicit trafficking pathways which the material followed to either it seizure by officials or use.[2] The attribution effort as a whole is a compilation of several different fields including chemistry, physics, intelligence, and social science.

Since much of the science that aids in the origin determination of nuclear material is inherently coupled to weapons development, much of the current work is classified. There are a few good sources available; however, almost all of these deal exclusively with seized materials that have not been used. We are specifically concerned with identifying material that has already been used in a nuclear explosion. In the 2005 book *Nuclear Forensic Analysis*, Moody et al. discuss the possibility of post-event forensics, but no specific capabilities or assessments are disclosed as is expected.[3] Media sources have reported that during training exercises at Sandia National Laboratory in 2004, several teams of experts were given data from nuclear tests in the 1960's and some of the teams were able to identify the materials.[4] Wallenius et al. have developed a method to determine the type of reactor in which a specific sample of Plutonium was made.[5] Each of these shows that there is a concerted effort going into this problem, but the overall capability can not be clearly seen at this point.

METHODOLOGY

Since we are focusing in on unsophisticated Plutonium based weapons, the work assumes that the explosion in question would be from an improvised nuclear device (IND) using only Plutonium as the fissile material. This work does not consider boosted or two-stage weapons. Since we are looking at

IND's, the yield of the device will be relatively low. We can focus our work on yields well below 100 kT. We also have assumed a uniform distribution of fission products and debris around the immediate area of the explosion. It is also worthy of note, that this methodology is designed to produce immediate results in order to provide an estimate for more sophisticated methods.

In approaching this problem, we start at first principles to derive a system of equations to lead us to pre-detonation isotopics. In characterizing the pre-detonation isotopics, we will find ratios of a given isotope to the main fissile material of ^{239}Pu. The main physical quantity related to the production of all isotopes in this system is the scalar flux of neutrons (neutrons/second). Both fission products and higher actinide production is directly related to the total flux of neutrons incident within the device. The total flux is related to the overall yield or burn up of the material. The yield per unit volume is the integral of the macroscopic fission cross-section of ^{239}Pu and the neutron flux with respect to time. By averaging this integral over time we can replace the integral with physical constants of burn-up (Ψ), the energy recoverable from fission (E_r), and the density of ^{239}Pu ($\rho^{Pu\text{-}239}$):

$$\int_0^T \Sigma_f^{Pu-239}(t)\phi(t)dt = BU\frac{1}{E_r}\rho^{Pu-239} \tag{1}$$

where $\Sigma_f^{Pu\text{-}239}$ is the macroscopic fission cross-section of ^{239}Pu.

This approximation of the yield will tie all of the equations for different isotopic ratios together. We will use a burn-up factor (Ψ_X), which is the burn up multiplied by a physical constant () given by,

$$\Psi_X = \Psi\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)}\frac{N_{Pu}(0)}{N_{Pu-239}(0)} \tag{2}$$

where $N_X(t)$ denotes atom density of isotope X at time t.

The next task is to develop equations related to determining the yield, which is proportional to the flux, from fission product data. We must carefully choose fission products which are relatively long lived in order to ensure that samples can be taken and analyzed before a significant portion of the sample decays. The three fission products we chose are Strontium-89 (half-life of 50.52 days), Strontium-90 (half-life of 28.78 years), and Zirconium-95 (half-life of 64.02 days).[6] We also specifically chose these fission products because they have small neutron absorption cross-sections in the fission averaged spectrum.[7] This means that the fission products produced in the event will not significantly "burn" out of the sample during the event.

Due to the relatively long half-lives of the chosen isotopes in comparison to the detonation time, we assume that fission product decay is negligible. Similarly, since absorption the cross-section for each fission product is small, we can ignore the burn out of the fission products after their initial production. Thus we find one equation for each fission product measurement to relate to the burn-up of the material. An average of the three results, will give us the best approximation for our burn-up factor. The equation for Sr-90 is

$$\Psi_X = \frac{N_{Sr-90}(T)}{N_{Pu-239}(T)}\frac{E_r N_a}{Y_{Sr-90}m_{Pu}} \tag{3}$$

where Y is the yield, N_a is Avogadro's number, and m_{Pu} is the mass of Pu. ^{95}Zr and ^{89}Sr have similar equations with the corresponding fission product yields.

We also correct our measured fission product data for the decay time between detonation and analysis. We get the exact time since detonation by using measured samples of ^{89}Y/^{239}Pu (stable[8]) and ^{89}Sr/^{239}Pu. The fission yield of ^{89}Y is very small, and we can set the initial value of ^{89}Y after detonation to zero, thus any ^{89}Y will have grown into the samples by radioactive decay of ^{89}Sr. This decay time (T_{decay}) is given by

$$T_{decay} = \frac{1}{\lambda_{Sr-89}} \ln\left[\frac{N_{Y-89}(T)}{N_{Sr-89}(T)} + 1\right] \tag{4}$$

where $\lambda_{Sr\text{-}89}$ is the decay constant for ^{89}Sr.

Undoubtedly, the exact time of detonation will be recorded by other means allowing an opportunity to compare the calculated detonation time with the observed detonation time. This gives a check to see if the swab samples taken have produced credible data.

Ultimately we need to determine the isotopic ratios of all the major Plutonium isotopes that could be in the system. This is the range of isotopes between ^{238}Pu and ^{242}Pu, including ^{241}Am. The equations for this are all solved simultaneously using the burn-up factor found from the fission products. We identified the dominant physical process for production or loss for each isotope in question. We then simplified the change rate of each equation to take into account only the dominant process.

The ratio of ^{239}Pu atoms before detonation to ^{239}Pu atoms after detonation is given by

$$\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = 1 + \frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{5}$$

where $\sigma_a^{Pu\text{-}239}$ is the microscopic absorption cross-section of ^{239}Pu, $\sigma_f^{Pu\text{-}239}$ is the microscopic fission cross-section of ^{239}Pu,.

Each other isotope of interest has an equation to determine its pre-detonation isotopic ratio with the post-detonation atom density of ^{239}Pu. These values are given by:

$$\frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-240}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{6}$$

$$\frac{N_{Pu-242}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-242}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{7}$$

$$\frac{N_{Am-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Am-241}(T)}{N_{Pu-239}(T)} \exp\left[\frac{\sigma_{absorb}^{Am-241}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X\right] \tag{8}$$

where $\sigma_\gamma^{Pu\text{-}239}$ is the microscopic (n,γ) absorption cross-section for ^{239}Pu, $\sigma_\gamma^{Pu\text{-}241}$ is the microscopic (n,γ) absorption cross-section for ^{241}Pu, and $\sigma_{absorb}^{Am\text{-}241}$ is the microscopic (n,γ) absorption cross-section for ^{241}Am .

After determining the ratios of all the Plutonium isotopes, we can now calculate the yield of the device. This is done by summing the plutonium isotope ratios and dividing this into the burn-up factor.

$$\Psi = \Psi_X \frac{N_{Pu-239}(T)}{N_{Pu}(0)} \tag{9}$$

We also find the initial ^{240}Pu/^{239}Pu ratio using:

$$\frac{N_{Pu-240}(0)}{N_{Pu-239}(0)} = \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \Big/ \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} \tag{10}$$

Similarly for all isotopes of interest we have:

$$\frac{N_{Isotope}(0)}{N_{Pu-239}(0)} = \frac{N_{Isotope}(0)}{N_{Pu-239}(T)} \Big/ \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} \tag{11}$$

These are the full attributes of the pre-detonation Plutonium.

TESTING

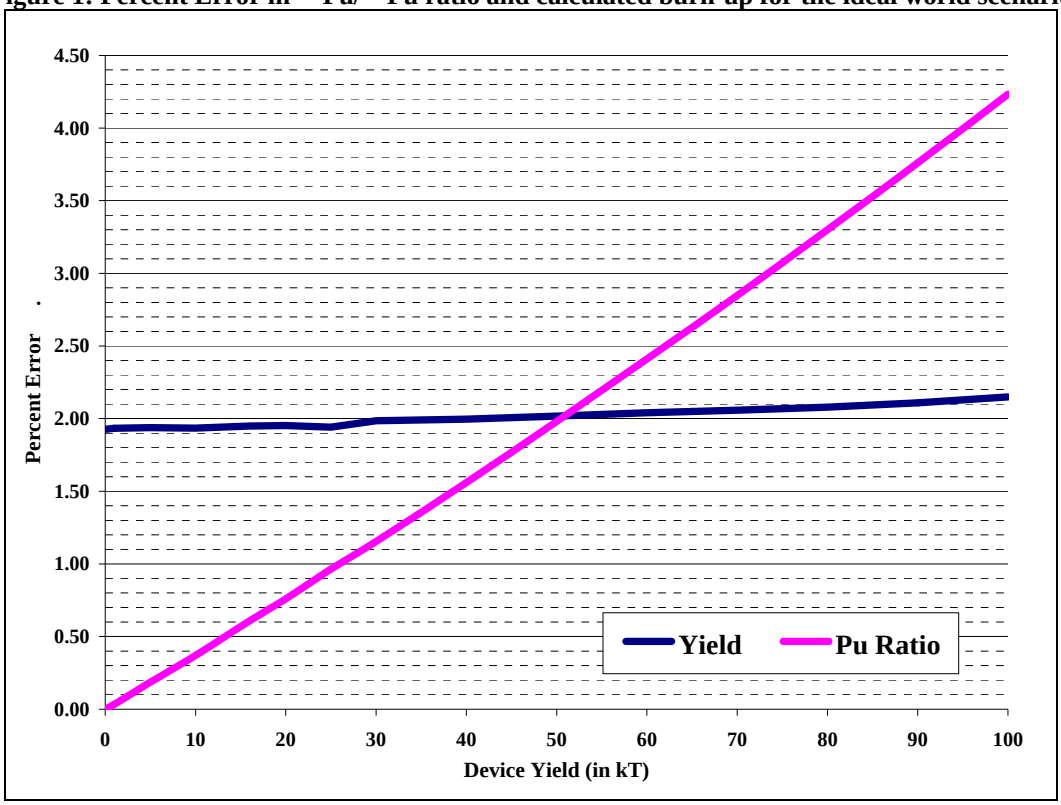
In order to test this method, the ORIGEN2[9] computer code was used to simulate the burn-up of appropriate nuclear weapons materials. ORIGEN2 has a broad set of libraries used to simulate different types and designs of nuclear reactors[10]. The fast flux library for Plutonium/Uranium fuel (FFTF[11]) was used to simulate the neutron flux and cross-sections that would be typical in a nuclear explosion. ORIGEN2 calculates the amount of material for all nuclides after irradiation and includes an option for post-irradiation decay. Since ORIGEN2 is a zero-dimensional model, the results are averaged over the entire system[12].

Ideal world cases and real world cases were tested using ORIGEN2. In the ideal world cases, there are only two nuclides present in the bomb material: ^{239}Pu (95 weight percent) and ^{240}Pu (5 weight percent). In the real world cases, the other isotopes of Plutonium and ^{241}Am were included. Weapons grade Plutonium and Reactor grade Plutonium are considered in the real world cases. The weight percent of each isotope used in the cases are listed in Table 1.

Table 1: Plutonium Samples Used with ORIGEN2

Nuclide	Ideal World weight percent	Weapons Grade weight percent	Reactor Grade weight percent
^{234}U	0.0	0.00046	0.0
^{237}Np	0.0	0.00088	0.0
^{238}Pu	0.0	0.00498	2.87
^{239}Pu	95.0	94.98	54.34
^{240}Pu	5.0	4.8	25.72
^{241}Pu	0.0	0.123	7.05
^{242}Pu	0.0	0.0052	6.72
^{241}Am	0.0	0.089	3.3

Figure 1: Percent Error in ^{240}Pu/^{239}Pu ratio and calculated burn-up for the ideal world scenario



The percent error of both the initial Plutonium isotopic ratio and the calculated yield are shown as a function of device yield for the ideal world case in Figure 1. The error in the calculated burn-up is approximately 2 percent for all yields between 0.1 kT and 100 kT. The error in the initial Plutonium isotopic ratio increases linearly with yield of the device. The Plutonium ratio reaches 2 percent error at a yield of 50 kT. The error in the Plutonium isotopic ratio stays well below 2 percent between 0.1 kT and 30 kT (the approximate yield range of an IND explosion). Thus we see that the methodology will yield accurate results within the stated yield bounds of the problem.

In testing the methodology on the real world case at a yield of 10 kT, the error in each of the Plutonium ratios was 1 percent or lower. The results of this test can be seen in Table 2. It is also of note that the change from ideal world to real world did not significantly change the amount of error in the $^{240}Pu/^{239}Pu$ ratio and calculated yield. Thus the system of equations performs as expected for a realistic Plutonium sample based on a theoretical irradiation using ORIGEN2.

Similarly, the Reactor grade Plutonium case was tested using a yield of 10 kT. The results of this test can be seen in Table 3. All the errors remain below 2 percent; however, each of the isotopic ratios have larger percent errors than in the Weapons grade Plutonium case. This is due to the assumptions used in deriving the methodology. Since each of the minor (non ^{239}Pu) isotopes have higher atom densities, neglecting secondary effects on these isotopes introduces additional error than.

Table 2: Weapons Grade Plutonium Benchmarking

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.265E-05	5.229E-05	0.70
$^{240}Pu/^{239}Pu$	5.033E-02	5.053E-02	0.41
$^{241}Pu/^{239}Pu$	1.284E-03	1.271E-03	1.03
$^{242}Pu/^{239}Pu$	5.365E-05	5.398E-05	0.62
$^{241}Am/^{239}Pu$	9.292E-04	9.295E-04	0.02
Burn-up (MWd/MT)	6.053E+03	5.938E+03	1.90

Table 3: Reactor Grade Plutonium Benchmarking

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.305E-02	5.242E-02	1.17
$^{240}Pu/^{239}Pu$	4.714E-01	4.736E-01	0.46
$^{241}Pu/^{239}Pu$	1.287E-01	1.263E-01	1.82
$^{242}Pu/^{239}Pu$	1.222E-01	1.213E-01	0.69
$^{241}Am/^{239}Pu$	6.023E-02	6.071E-02	0.79
Burn-up (MWd/MT)	6.053E+03	5.953E+03	1.65

SENSITIVITY ANALYSIS

There are 12 physical constants that are used in the system of equations. These constants consist of fission yields, cross-sections, masses, and the energy recoverable from fission. Errors and uncertainties in these constants could have a significant impact on the final results. Typically, the cross-section data for Plutonium is known very well due to years of weapons testing and design. The uncertainty in this particular quantity will come from choosing which spectrum averaged cross-sections to use. The cross-sections built in to the ORIGEN2 libraries were used here. Fission spectrum average cross-section would be used in an actual event. Likewise, the fission yields, energy recoverable, and initial plutonium atomic mass could contribute to errors in the final results.

To test these, each of the constants was independently peturbated by a change of 5 percent. The final results for each desired ratio, as well as the calculated yield, were noted. As expected, each constant only affected specific ratios. Figure 2 shows the largest change seen by perturbing each variable.

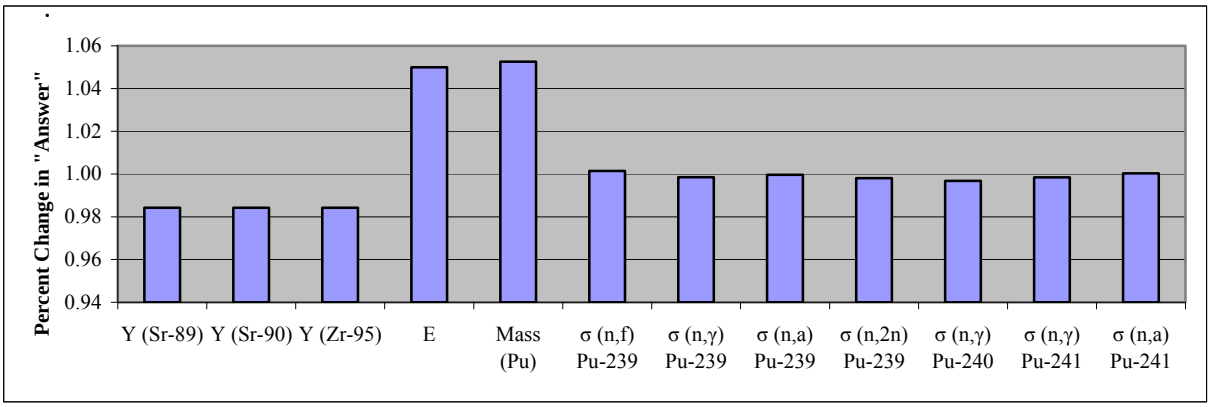


Figure 2: Errors caused by 5 percent pertubations in physical constants

The fission yield data increases affected every answer, but again they were fairly small changes as well. The largest change was in the calculated yeild. The initial Pu mass and recoverable energy had a significant impact on the yield calculation, but did not affect the ratios at all. This effect is expected since these quantities essentially are only introduced to find the burn-up.

CONCLUSIONS

We have shown that it is possible to approximate the pre-detonation isotopic ratios of Plutonium based improvised nuclear devices using measured isotopic ratios taken from debris. We have focused on the speed of computations in order to provide a starting point estimate for more sophisticated methods to utilize in iterative computing. This will hopefully decrease the overall attribution time. Even though we were unable to recreate the exact numbers given by our ORIGEN2 output, we have been able to recreate that data within acceptable limits of error. From this, we can begin to couple our work with that of others to give a more complete picture of the source and route taken to detonation as an IND.

The next steps in this work are to continue the error sensitivity analysis. We will specifically focus on correlated errors in the physical constants. Once we have determined the most error sensitive variables, we can then modify the methodology into an iterative process to reduce errors as much as possible.

Further work is to combine this method with a similar analysis done for Uranium devices of similar yield. Emphasis will be put on the scenario where Uranium and Plutonium are blended to create a device as well as defeating possible spoofing techniques.

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