# Understanding Uncertainties in Nuclear Archaeology

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**ABSTRACT.** There is a lack of techniques to verify the completeness of baseline declarations of fissile material holdings. Such a capability is important whenever states declare existing fissile material inventories, e.g. as part of joining the Non-Proliferation Treaty and disarmament treaties such as Treaty on the Prohibition of Nuclear Weapons. One of the most promising approaches is nuclear archaeology, i.e data collection and analysis to reconstruct the past fissile material production history. Some of these data are collected from past operating records and declarations while other data are generated using different measurement approaches, e.g. measuring the activation of structural and moderator materials in reactors. For each of these sources, the data are affected by uncertainties. These uncertainties result in an overall uncertainty of the final assessment of material stockpiles.

It is thus imperative for nuclear archaeology to be able to identify and understand these uncertainties, and then - to the extent possible - reduce them. This paper provides an overview of the possible sources of uncertainty in nuclear archaeology for plutonium production. Some uncertainties may be directly contained in records (e.g. from past measurements), others will be the result of incomplete information in the records. Uncertainties of measurements taken during inspections must be taken into account. In addition, to deduce past activities from such measurements, calculations and simulations are required that again bear uncertainties, for example from the use of compiled nuclear data sets (e.g. cross section libraries), or computational representations of physical realities. For the paper, a comprehensive list of uncertainties sources will be compiled. For each of these uncertainties, we will qualitatively analyze their impact on the final results and discuss to which extent possibilities to reduce them may exist.

# Introduction

While there is extensive experience in verifying both the correctness and completeness of nuclear material declarations issued by non-weapon states that are members of the Non-Proliferation Treaty (NPT), there is a lack of methods to verify nuclear material "baseline" declarations, i.e. the first verified declaration a state makes upon entering an agreement. This would be relevant for states that formerly possessed nuclear weapons wishing to join the NPT, and former or current weapon states joining the Treaty on the Prohibition of Nuclear Weapons (TPNW), which requires verifying the absence of undeclared nuclear materials. Additionally, baseline declarations will be important for achieving disarmament: A solid understanding of fissile-material holdings is needed to achieve a meaningful degree of predictability and irreversibility of future arms-control initiatives. Speculations about unaccounted fissile-material stockpiles, possibly equivalent to hundreds of nuclear weapons, could make progress in this area very difficult.

As was attempted after South Africa had joined the NPT in 1991[1], the most promising approach to verify the completeness and correctness of baseline declarations is the reconstruction of the state's fissile material production history.[2] This is called nuclear archaeology, a concept introduced in 1990.[3] Since then, however, there has only been limited research; there remains a research gap on applicable tools and methods, which must be developed for a systematic and effective nuclear archaeology approach.

This paper discusses the issue of reconstructing past plutonium production, although the past production of highly enriched uranium also requires significantly more research. For plutonium, one approach is to take samples from structural materials inside the cores of shut-down reactors, and deducing the neutron fluence from isotopic ratios of trace elements obtained from forensic analysis. Some examples include boron, lithium, chlorine, calcium, titanium, chromium, iron, nickel, zirconium, and lead.[4] From the fluence, the plutonium production can be deduced. An example of this approach is the Graphite Isotope Ratio Method, which is applicable for graphite-moderated reactors.[5] While this method has been tested, similar methods for heavy water reactors are in an early stage of development. Another concept has been proposed which can be used in reactors that use aluminum tubes, such as the Indian CIRUS reactor.[6]

Deducing plutonium production of a reactor requires three steps, outlined by Gasner and Glaser.[7] First, the neutron fluence at a specific location in the reactor must be expressed as a function of the measured isotopic ratio of samples at that location. According to the depletion equation, this depends on the production and depletion paths of the considered isotopes. It also depends on the corresponding one-group reaction cross-sections, which depend on the neutron energy spectrum at that location. This can be obtained from neutron transport simulations. Second, the plutonium production at a specific location must be expressed as a function of the local fluence estimate. For this, fuel burnup calculations are required, which require input parameters such as reactor power or discharge burnup. Third, the global plutonium production of a reactor must be inferred from local plutonium production. This requires full-core reactor simulations to obtain spatial information.

With all of these three steps, various kinds of uncertainties and errors are introduced, which all must be propagated through the calculation process to obtain the final uncertainty on the plutonium production estimate of a reactor. This is important, especially given that the uncertainties can be expected to be much larger than a single significant quantity of plutonium.

# Uncertainties

Uncertainties[8] in nuclear archaeology result both from measurements and simulations. Uncertainties of physical measurements are the result of random and systematic errors made while measuring. Random errors are inevitable, especially when the number of measurements for a single value is limited. Systematic errors are introduced by all parts of a measurement system - humans, technology or protocol. Although they are called systematic, it is not always easy to eliminate them by improving systems. Many measurements for nuclear archaeology particularly suffer from statistical uncertainties. These uncertainties are easy to reduce if the measurement time can be extended. However, a particular problem for nuclear archaeology is the limited repeatability for measurements. At some point, facilities might be destroyed or dismantled, so the object of measurements gets removed. Hence no more measurements are possible.

Simulation - modelling of the physical world and physical effects of the past - is also error-prone. Of course, modelling uncertainties are influenced by measurement uncertainties. They also add additional errors due to omission of some effects - often not all physical effects known are modelled for performance reasons. And lastly, they can suffer from errors made by those who carry out the simulations.

## Nuclear Data

Any simulation of a nuclear reactor core requires the use of a variety of nuclear data libraries. These include data on mass, decay constants and cross sections for a variety of reactions based on incident neutrons and other incident particles. For nuclear fission reactors, the details of the fission process including fission product yield, fission energy and the number and energy of neutrons produced by fissions are important, too. These data are usually obtained through empirical measurements and are commonly published with uncertainties. A realistic assessment of these data uncertainties is necessary for evaluating the reliability of simulation results.

An effective way to assess the influence of such uncertainties on output parameters is to conduct Monte Carlo simulations of perturbations of the uncertain input parameters according to their probability distributions, and assessing the corresponding distribution of output parameters. These parameters can be neutron flux or one-group cross-sections, but also the isotopic evolution during burnup. Pertubabtion calculations require a large number of neutron transport or fuel burnup calculations. Sampling-based methods are incorporated into several code packages, for example SAMPLER as part of the SCALE package.[9] In principle, uncertain parameters can be perturbed at the same time, so that a sensitivity analysis can be carried out based on the Monte Carlo results and by calculating the correlation coefficients between an output variable and the specific input variables. An implementation for depletion calculations is for example described by Zwermann et al.[10]

Nuclear data uncertainties impact the nuclear archaeology reactor assessment in the three analysis stages mentioned above. A neutron transport simulation has to be carried out to obtain the one-group cross-sections so that the evolution of the trace isotopes in structural materials can be linked to the neutron fluence. All nuclear data uncertainties mentioned above come into play here, as they impact the neutron energy spectrum. They must also be considered for the second step, as nuclear data uncertainties impact the evolution of plutonium production as a function of burnup or time for a fuel batch. Also the spatial information (third step) depends on it, as these data are used for the full-core simulations.

For example, one study examines the uncertainties of isotopic concentrations based on nuclear data uncertainties for pressurized and boiling water reactors at 40 GWd/tHM. The actinide concentrations

are most sensitive to neutron cross-sections. The uncertainties of the Pu-239 and Pu-240 concentrations are around 2%.[11] Note, however, that burnup for reactors aimed at plutonium production will be much lower. Still, a careful study would be required to evaluate the full impact of nuclear data uncertainties on all steps of this nuclear archaeology technique and the final plutonium estimate. It appears, however, unlikely that this will be the major contributor to the overall uncertainty of the plutonium estimate. Such outcome would be fortunate, given that improving nuclear data is a costly experimental undertaking.

## Design Parameters

Design parameters can also contain uncertainties. In this paper, design parameters include all parameters that are defined at the design stage of a nuclear reactor and that can not be promptly modified on a regular basis. This encompasses design geometry such as reactor size, assembly geometry, assembly-to-assembly pitch, assembly channel layout throughout the reactor core, control instruments arrangement and initial isotopic composition of structural material and of fuel. Uncertainties will have effects on the reactor simulation, but also on measurements of isotopic ratio in structural material.

The way design geometry affects the final assessment of fissile material production is simple to grasp but can be complex to model quantitatively. Uncertainties on the overall size of the reactor or the assembly channels pattern of the reactor cause the geometry of the simulated system to deviate from reality which will inevitably lead to substantial errors in the results. Studying how these uncertainties play out on the final assessment of fissile material will depend a lot on the geometry parameters in question. An uncertainty on the overall size of the reactor could play the role of a factoring element in the final assessment of fissile material production in the first order of approximation. Uncertainties and errors that stem from lack of information on the assembly pattern of the reactor will be difficult to propagate analytically to the final assessment. In this situation, studying the output of multiple simulations with various assembly layouts is the only option in order to have an idea of how this plays out on the final assessment. Errors and uncertainties in the initial composition of the fuel material and certain structural material will have effects on the neutronics simulation. This is not necessarily the case for uncertainties in the initial abundance of trace isotopes that are going to be used for nuclear archaeology. However, since knowing how specific trace isotopes ratios have evolved from their initial values to the time of the measurement is critical in nuclear archaeology, these uncertainties and errors will have strong effect on the final result. These uncertainties directly interfere in the relation that can be obtained between local fluence and the measured isotopic ratio. For example, suppose we are measuring isotopic ratio $R = N_b/N_a$, where $N_a$ and $N_b$ stand for the density of isotopes a and b respectively. The initial isotopic ratio will be written as $R_0 = N_b(t = 0)/N_a(t = 0)$. Let us say a produces b via the neutron induced reaction $\phi\sigma$. Densities for a and b will be subject to the following system of depletion equations:

$$\frac{dN_a}{dt} = -\phi\sigma N_a \quad \frac{dN_b}{dt} = +\phi\sigma N_a \tag{1}$$

If we solve this system and take fluence as $\Phi = \phi \cdot t$, we can find the following relation between R and $\Phi$:

$$\Phi = [\log(R + 1) - \log(R_0 + 1)] \cdot \frac{1}{\sigma} \tag{2}$$

If we assume that $R_0$ is known with uncertainties $\delta R_0$ (which can be negative of positive), then applying Taylor expansion will yield:

$$\delta\Phi = \frac{1}{\sigma \cdot (R_0 + 1)} \delta R_0 \tag{3}$$

Since we can always find a closed form relation between the production of fissile material and the fluence $\Phi$, it is then straightforward to find a relation between $\delta R_0$ and the former. Reducing the uncertainties for both sources will involve similar efforts. In the ideal case, inspectors would be allowed to access design documentation as well as being granted direct observation of the reactor to confirm the design parameters. If the host is not cooperative, other investigations could still reduce these uncertainties. For example, even if no documentation is available on the reactor investigated, there might be some other reactors in other countries or regions whose designs are similar and for which documentation is available. Operational Records (Parameters) Generating plutonium production estimates based on nuclear archaeology simulations involves simulating a reactor in the way it was operated while the plutonium was produced. To do so, a simulation needs knowledge of the total power the reactor was operated with, the distribution of that power in different areas, the irradiation time of the fuel, the refueling schedule, the original fuel composition, the temperatures prevalent in the reactor over time, position of the control rods over time and the status of other possible neutron absorbers like boron in cooling water or other neutron poisons. For a variety of reasons, getting an estimate for these values can be difficult. In the worst case, values have not been measured at all. This might be true for certain operating parameters that are hard to obtain (e.g. temperature gradients in reactor core), or not important for operation safety. Also, values might have been measured at the time of operation, but were not documented (e.g. some trace isotopes in fresh fuel were not recorded) or the documentation was lost or discarded. Even if values are documented, errors can be introduced. Most obvious are direct recording errors ("typos") - which are very hard to recover. It can also be due to unnecessary rounding of values, removing precision more than required by actual measurements. An example for a document including officially rounded numbers are the declarations that plutonium separating countries make as part of the IAEA Information Circular 549.[12] Plutonium figures in this case are rounded to full 100 kg of plutonium, although country's domestic accounting systems should have more accurate values. While the IAEA declarations here are not used for nuclear archaeology, it is an example for artificially introduced uncertainties. Lastly, uncertainties in operational parameters are introduced from actual measurement errors when operational parameters are produced. For example, power level measurements of LWR are typically done by combining flow rate of water and temperature. Depending on the technology used, this can lead to uncertainties of more than 1%.[13] If missing operational values are only gaps in the documentation, it might be possible to interpolate values. If they are completely missing, there might still be a way to reconstruct them. Using simulations with a variation of initial assumptions, and comparing results to measurements of structural reactor material samples, it might be possible to calculate some of these missing values. A study shows for example that it may be possible to determine the reactor operation mode (whether it produced plutonium or tritium, or both at different times) by such a calculation.[14] But even if values are recorded for many operational parameters, it could be impossible to reduce their uncertainties, as they have been recorded in the past. Only if the host cooperates closely and has some additional data that she or he could add to the process, such reductions might be possible.

## Model Approximations

Model approximations include all the approximations that are created when defining the physical model as well as the numerical model of a system to be simulated.

Simplifying and reducing the network consisting of the nuclides connected by neutron-induced reactions and decay events will make the model deviate from reality, thus inducing errors in the results. To illustrate this fact, we can consider the creation of Pu-239 in a typical nuclear reactor. Pu-240 produces Pu-239 via a (n, 2n) reaction. If the physical model does not take into account (n, 2n) reactions, then this branch in the nuclide network will be absent and the Pu-239 inventory calculated with the model might be slightly smaller than in real life. A similar situation would be produced if the model, for some reasons, does not take into account Pu-240 in the nuclides network. The absence of a type of reaction in the model can have more complex effects on a result, too. For example, not considering (n, 2n) reactions might also increase the inventory of certain nuclides that participate in the creation of Pu-239, leading to higher production value estimates.

Time discretization, a commonly used practice in neutronics codes, produces errors on the final result. For time discretization, the whole burnup interval is divided into smaller intervals within which the total value of the flux as well as its energy distribution are held constant. Since one-group cross sections only vary when the flux spectrum changes, holding the spectrum constant is equivalent to holding one-group cross-sections constant. This enables the decoupling of the depletion equation[15] from the neutron transport equation and allows to solve the former in a rather simple manner.[16] However, this simplification will make the numerical simulation deviate from reality since the total value of the flux and the one-group cross-sections are continuously changing with time.

Two other types of discretization approximations are predominant in neutronics: space and spectral discretization. Space discretization is the fact that neutronics codes will separate the whole system in several regions or cells. Within a cell, material is considered as an homogeneous mix of all the elements present in the cell, and neutronics codes will calculate only one neutron spectrum, one neutron flux total value and one set of nuclide densities for this cell. One of the major consequences of spatial discretization is to negate what is commonly known as spatial self-shielding. Spatial self-shielding is the fact that a highly absorbant material for neutron will see a lower neutron flux in their regions. When a highly absorbant material, such as a fuel pin, is grouped in the same cell with a less absorbant material, such as cladding, the resulting cell is going to be a homogeneous mix of both cladding material and fuel pin material. The single neutron flux value calculated for this cell is going to be bigger than the real one the fuel pin is exposed to. This will result in more fission events than in reality which will have important consequences on the neutronics and isotopic inventory. Spectral discretization stands for the fact that within each cell, the neutron spectrum is divided into several energy groups. For each of these groups, a neutron flux is computed (the sum of these group fluxes is the cell total flux) and cross-sections are derived. Since the neutron energy spectrum is continuous in real life, spectral discretization brings about approximations which make the model deviate from reality and thus induce errors.

Finally, a simulation commonly uses other simplifications that will all make the model deviate from reality. These approximations comprise geometrical simplifications when it comes to setting the reactor geometry within the simulation, cross-sections temperature limitations as neutronics codes do not provide cross-sections for all temperature or partial core simulation as it is common practice to simulate only a part of the reactor core to save computing resources.

Obviously, the more detailed the model is, the closer to the reality it is. Hence, adding more nuclides and reactions to the nuclide network, using finer time steps, spatial meshes as well as a finer energy group structure will tend to reduce the errors produced by the model. Other techniques also exists to attenuate these errors. For example, burnup codes can use an algorithm called Predictor-Corrector that reduces the error produced when keeping the flux constant during burnup steps. However, each of these improvements will also increase the running time of the simulation. For example, increasing the number of cells to describe the system also increases the time of the Monte Carlo simulations. Since

Monte Carlo simulations tend to be time-consuming, the user has to think carefully of an appropriate balance between accuracy and running-time.

# Computational Errors & Uncertainties

Besides reactor simulations, storing data on and processing data with computers is necessary for nearly every step of nuclear archaeology. This can lead to another uncertainty in results. The commonly used approach to store real numbers in computers involves the use of floating point arithmetic, allowing to store any number in a fixed number of bits. As such, only a finite number of real numbers can actually be stored. For example storing the half-life of U-238 (4.468e9 years) in a 32 bit floating point value actually stores 4.467999744e9. Clearly, that difference is very small, and in most calculations does not lead to an actual uncertainty. However, there are special cases for which it becomes important.

A real world example of grave consequence is a software error in the Patriot Missile Defense System. In February 1991, such a system failed to intercept an Iraqi scud missile, leading to the death of 28 Americans. It was later discovered that the failure was due to an improper use of floating point numbers - over time, an error accumulated, eventually the incoming missile could not be tracked accurately.[17] For nuclear archaeology applications, the most relevant area for accumulating floating point errors is the calculation of matrix exponentials. This uncertainty can be avoided by taking careful measures to select algorithms and sizes of floating point numbers appropriate for the respective problem. There exist also routines allowing for digital representation of numbers with arbitrary precisions, which could be used if necessary.

Whenever the Monte Carlo method is used for calculations, a statistical uncertainty is added to the result. Any value produced by random sampling suffers from the limited number of sampling events that are included in the result. This uncertainty is easy to quantify; most Monte Carlo code packages readily provide these values. It is also easy to overcome, given that computing resources are available. To reduce the uncertainty, simulation times have to be increased. A reactor simulation has to calculate transport of more source neutrons. A second way to reduce uncertainty is to optimize simulation geometries for specific problems and as such reduce computational load for areas with low influence on final values.

## Measurement Uncertainties

One of the most crucial components in nuclear archaeology is the "on-the-field" work of inspectors where they collect samples from a nuclear reactor and analyse them. For the convenience of this paper, both steps have been lumped together under the term of "measurement".

When inspectors collect a sample from structural or moderator material, the ideal situation would be that they directly monitor or witness the sample collection process. Not knowing the original location of the sample collected will greatly complicate the task of nuclear archaeology and will produce important uncertainties on the final result: The value of an isotopic ratio might be different depending on the location (even a few centimeters apart), hence there might be uncertainties on the isotopic ratios from the simulation with which the measured isotopic ratio has to be compared.

In order to measure isotopic ratios from a sample, inspectors would usually use mass spectrometry. Measurements with mass spectrometry technologies always come with uncertainties. Another potential issue are interferences caused by isobars. Uncertainties on the measured isotopic ratios will then be propagated to the final assessment of fissile material production.

It is reckoned that for both sources of uncertainty, analytical error prograpagtion could be used in order to have a sense of their importance on the final assessment of fissile material production. In fact, both effects play out during the process of comparing the measured ratio with the simulated ratio to find the local fluence. If we take the same system of equations as presented in the section on design parameters and if we assume this time that $R$ is measured with uncertainties R(which can be negative of positive), then applying Taylor expansion will yield:

$$\delta\Phi = \frac{1}{\sigma \cdot (R+1)}\delta R \tag{4}$$

Since the production of fissile material is directly proportional to the fluence $\Phi$ it is then straightforward to find a relation between $\delta R$ and the former.

When it comes to reducing these uncertainties, both sources have to be handled differently. There exist many different technologies for mass spectrometry and some are more suitable for certain type of sample material or yield higher precision and accuracy for certain isotopic ratios. Therefore, the choice of technology needs to take into account the nature of the sample analysed as well as the isotopic ratios to be measured. Another way to reduce the uncertainties when measuring isotopic ratios with a mass spectrometry instrument is to make sure the abundance of the isotopes involved in the ratio is high enough. Spectrometry technique uncertainties tend to be higher when the concentration of isotopes are low. Regarding uncertainties on the original location of the sample, there are mainly two different ways to reduce them. Ideally, the inspectors themselves take the samples or observe the host while the sample is taken so that the knowledge of the location is completely reliable.

# Conclusion

As laid out in this paper, nuclear archaeology has to cover a large range of uncertainties. We focused here only on plutonium production in nuclear reactors, future work also will have to address nuclear archaeology of other fissile material production processes, e.g. uranium enrichment with gas centrifuges. The quantification of uncertainty is an important step, especially as small uncertainties early on propagate through more complex calculations. And even small uncertainties can turn out to be of high significance for larger stockpiles (e.g. 1% uncertainty for a stockpile of one metric ton of plutonium could be enough material for 2-3 additional/less nuclear warheads). A next step from this collection of sources is trying to quantify their impact using different methods of error propagation and sensitivity analysis. This can show areas where reduction of uncertainties are most valuable for the final result.

## Endnotes

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