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ARTICLE

Verifying North Korea's Plutonium Production with Nuclear Archaeology

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ABSTRACT

North Korea produced weapon-grade plutonium in its graphite-moderated 5-MWe reactor. Estimating the total production of fissile materials provides an important baseline for denuclearization efforts. Nuclear archaeology can improve such production estimates by measuring isotope ratios in the graphite moderator of the reactor. The accumulation of certain trace isotopes in the graphite enables to accurately estimate life-time reactor fluence which can then be related to plutonium production. This article uses the open-source reactor physics software ONIX to simulate the operation of the 5-MWe reactor. It discusses consolidated estimates of plutonium production from 1986 to 2020 based on publicly available operation history data. An updated mathematical framework to relate isotope ratio uncertainties to fluence uncertainties and its implementation in a special ONIX module for nuclear archaeology are also presented. The module is used to identify which isotope ratios should be measured in the 5-MWe reactor to minimize uncertainties on plutonium estimation.

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Background

Between 2006 and 2021, the Democratic People's Republic of Korea (DPRK) has detonated six nuclear explosive devices in nuclear weapon tests. For at least four of these tests, the DPRK used plutonium-based nuclear weapons.[1] Plutonium is one of the two fissile materials, besides highly-enriched uranium, commonly used in nuclear weapons. The element plutonium does not exist in nature. It is, however, produced during nuclear reactor operations if a reactor is fueled with uranium.

North Korea has been operating a small plutonium production reactor, the 5-MWe reactor at the Yongbyon nuclear complex, sporadically for the last 35 years. Production was interrupted both for technical as well as for

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political reasons. In addition, North Korea operates the Radiochemical Laboratory (RCL), a plutonium reprocessing facility where plutonium is extracted from the 5-MWe reactor irradiated fuel. With few exceptions, the DPRK has not allowed international inspectors to monitor these facilities. As a consequence, the international community has been unable to verify plutonium production in the country.[2]

Nuclear archaeology methods can determine past production in a nuclear reactor, even after the reactor has shut down.[3] Such an analysis requires access to the reactor prior to full dismantlement. Knowledge of past production would allow inspectors to verify the completeness of fissile material declarations of a country. In recent years, several rounds of negotiations were initiated between former U.S. President Donald Trump and the North Korean leader Kim Jong-Un to advance denuclearization efforts on the Korean peninsula, but these initiatives ultimately failed. Resuming negotiations could lead to future disarmament or arms control agreements. Such agreements should include verification of past plutonium production in the 5-MWe reactor with nuclear archaeology.

This article has two objectives. First, it estimates plutonium production in the 5-MWe reactor using recent public information on the operation history of the reactor. Estimates are based on simulations with ONIX, a recently developed open-source reactor physics code.[4] As open-source software, the tool-chain is freely available. In principle, anyone, including DPRK scientists, could replicate the calculations presented here. Second, the article introduces an updated approach to fluence estimation in reactors. A rigorous mathematical framework allows for the selection of suitable isotope ratios to reduce uncertainties on fluence estimates. The framework has been implemented as a module within ONIX. Applying the framework to the 5-MWe reactor, the article presents the most suitable isotope ratios to estimate plutonium production in the North Korean reactor, both for past production as well as for potential future monitoring.

The 5-MWe reactor in Yongbyon

Construction of the 5-MWe reactor began in 1979 and was completed in 1986.[5] The reactor is capable of producing a power output of 5 megawatt electric (MWe), hence it is typically called the "5-MWe reactor." The design is similar to the British Calder Hall reactor, which the United Kingdom used to produce weapon-grade plutonium. The reactor uses graphite to moderate neutrons, and is cooled using CO_2. Stacked graphite blocks form the core and channels through the blocks hold the fuel rods and allow for cooling gas flow.[6]

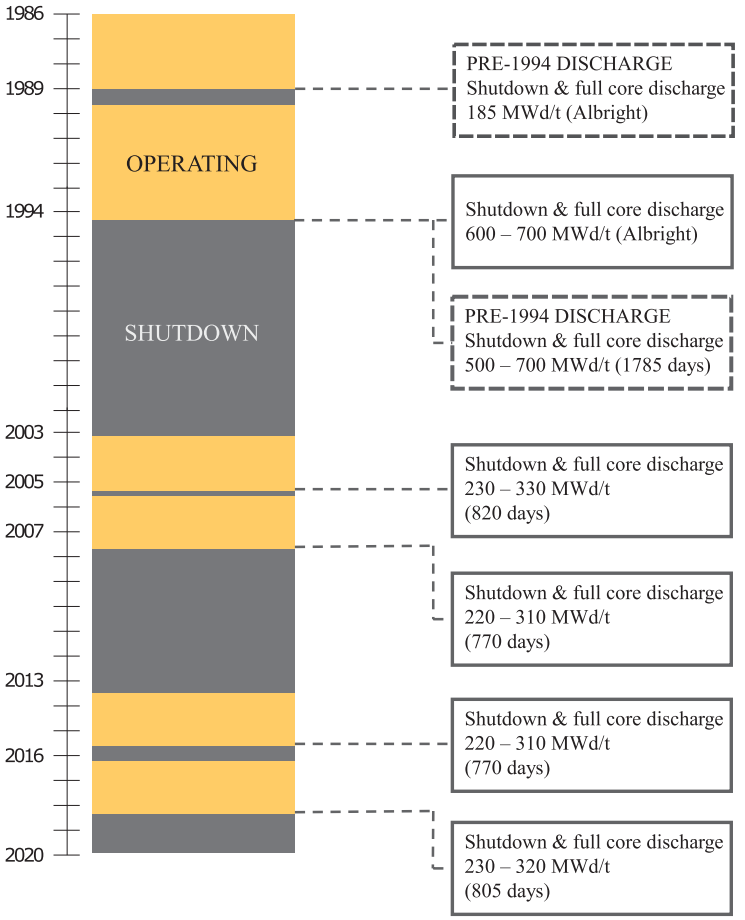

Figure 1. Potential 5-MWe reactor operation history timeline.

The 5-MWe reactor operated intermittently since 1986. Figure 1 illustrates a possible timeline of reactor operations. The first 70–100 days shutdown took place in 1989. There are suspicions that the DPRK discharged either a partial or the full core at that time.[7] In 1994, the reactor was shut down and defueled. Later that year, North Korea and the United States signed the Agreed Framework. North Korea froze the production of fissile material for nine years. In 2003, the country restarted the 5-MWe reactor and reprocessed the irradiated fuel that was unloaded in 1994. The reactor operated for two production campaigns: from February 2003 to April 2005 and from June 2005 to July 2007. The irradiated fuel from both campaigns was reprocessed. In June 2008, in the context of the "six-party talks" on the denuclearization of the Korean peninsula, North Korea destroyed the cooling tower of the 5-MWe reactor as a symbol of good faith.[8] Nevertheless, the reactor was started again and operated from August 2013 to October 2015. The irradiated fuel was reprocessed the following year.[9] Finally,

operations resumed in early 2016 and the reactor was running until the spring of 2018. The irradiated fuel was reprocessed shortly thereafter.[10] There are no indications that the reactor operated in 2019 and 2020.[11]

The main production scenario considered here assumes that North Korea had not discharged and reprocessed fuel elements prior to 1994. The average burnup level of the fuel in 1994 (with no prior discharge) is estimated by Albright and O'Neill in *Solving the North Korean Nuclear Puzzle* to be between 500 and 700 MWd/t.[12] For other operation periods, a lower and an upper bound for the burnup are estimated as follows: the lower bound is found by assuming that the reactor was operating at 20 MWth with a capacity factor of 70%. The upper bound is derived by assuming 25 MWth and 80% capacity factor. An alternative scenario where North Korea is assumed to have discharged and reprocessed a full core twice (in 1989 and in 1994) is also discussed briefly. For this scenario, referred to as "pre-1994 discharge," the average burnup in 1989 is taken at 185 MWd/t (Albright and O'Neill).[13] The burnup level for the production campaign between 1989 and 1994 is calculated in the same way as in the main scenario.

Estimating plutonium production

ONIX simulations were used to model the change of the material composition in the core of the 5-MWe reactor. ONIX couples a depletion module with the open-source neutron transport code OpenMC.[14] For this work, the geometry of the 5-MWe reactor was simplified to a single unit cell. The modeled cell contains a fuel channel in the graphite moderator and is represented in Figure 2. Six reflective surfaces around the unit cell define an infinite lattice that approximates the core.

The full core comprises 812 channels in which a maximum of 8,120 fuel elements can be inserted. The initial load of natural uranium metal is 50 tonnes. The uranium metal is an alloy with 0.5 at% aluminum.[15] Appendix A contains a table that summarizes some of the technical properties of the reactor core. The power density was set to 0.5 kW/kg of Initial Heavy Metal (IHM) which corresponds to an operating power of 25 MWth. Simulations showed that power density had a negligible influence on the neutronics. The simulations are divided in multiple burnup macro-steps.[16] For each of the macro-steps, OpenMC computed one-group reaction rates and one-group neutron flux. Each step included the simulation of 1 million neutron histories. Three micro-steps divide each macro-step in the depletion module. Nuclear data for decay, fission yields, and cross sections were taken from the ENDF/B-VIII.0 library. The input files and output data of the simulation used for this work can be accessed online.[17]

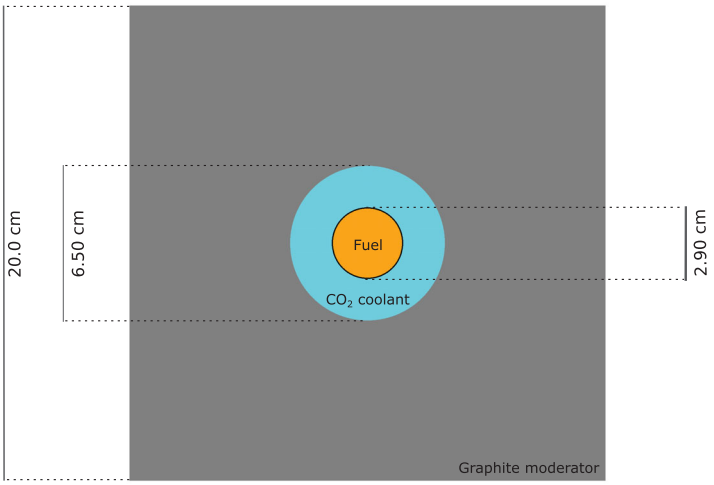

Figure 2. Geometry of the unit cell of the 5-MWe reactor. The natural uranium fuel is in orange, the CO_2 coolant in cyan and the graphite moderator in dark gray. The cladding is displayed as a thin line around the fuel.

Table 1. Estimates of plutonium produced in the 5-MWe reactor and current stockpile.

Production campaign	Plutonium [kg]
1986–1994	27.6 ± 1.8
January 2003–April 2005	12.8 ± 2.2
June 2005–July 2007	12.1 ± 2.0
August 2013–October 2015	12.1 ± 2.0
January 2016–March 2018	12.8 ± 2.2
Total Produced (2020)	77.0 ± 9.9
Reprocessing loss (10%)	-(7.7 ± 1)
Metal fabrication loss (10%)	-(6.9 ± 0.9)
Weapon use	-22
Current stockpile (2020)	40.4 ± 8.0

The ranges are given based on two sets of calculations: One using a power level of 20 MWth at 70% capacity factor and a second with a power level of 25 MWth and 80% capacity factor.

Table 1 presents the production of plutonium for each campaign with ranges that correspond to lower and higher burnup bounds as well as estimates of current stockpiles of plutonium in North Korea. These results are directly taken from ONIX simulations' output data. The cumulative total plutonium produced in 2020 is 77 ± 9.9 kg. While the plutonium produced during different production campaigns does not have the exact same content of plutonium-239, simulations show that all of them produced plutonium with more than 93 wt% plutonium-239.

Theoretically, at maximum capacity (at a power of 25 MWth and at 100% capacity factor) the reactor could produce on average 8.1 kg of plutonium per year. Assuming a more realistic reactor performance (22.5 MWth and 75% capacity factor), the reactor could produce on average 5.6 kg of plutonium per year.

To estimate the current stockpile losses during plutonium reprocessing and metal fabrication are each taken to amount to 10% of the quantity processed. This reduces the total to 62.4 ± 8 kg. Then, plutonium lost in nuclear tests was subtracted. The first two tests (in 2006 and 2009) are assumed to have consumed 6 kg of plutonium each. The fourth test (in 2016) and the sixth test (in 2017) are assumed to have used 5 kg of plutonium each. It is assumed that the two remaining tests used highly enriched uranium (HEU) only.[18] The resulting final estimate for North Korea's current stockpile of plutonium in metallic form is 40.4 ± 8 kg. This is slightly higher than the most recent estimate by Siegfried Hecker, who estimates 36.5 ± 11.5 kg.[19] If two discharges are considered before 1994 ("pre-1994 discharge" scenario), the estimate for the current stockpile of plutonium in metallic form is 45.7 ± 9.5 kg.

Nuclear archaeology to verify plutonium production

Nuclear archaeology is a set of scientific methods aimed at deducing past operation of fissile material production facilities.[20] As such, nuclear archaeology methods can be used to verify both past and ongoing production of plutonium in nuclear reactors.

Plutonium is mainly produced when nuclei of uranium-238 in the fuel absorb neutrons:

$$\text{U-238} + n \overset{(n,\gamma)}{\rightarrow} \text{U-239} \underset{24m}{\overset{\beta^-}{\rightarrow}} \text{Np-239} \underset{2.4d}{\overset{\beta^-}{\rightarrow}} \text{Pu-239} \tag{1}$$

The other isotopes of plutonium are primarily produced through subsequent (n,γ) reactions, transmuting plutonium-239. Plutonium is also depleted when fissioned by neutrons. Hence, the quantity of plutonium in the core is a function of the time integral of the neutron flux in the reactor, i.e., the total neutron fluence. As the neutron flux, and thus the fluence, is not spatially uniform, knowledge of the local fluence is necessary. By measuring the local fluence at various positions in a reactor, one could estimate the total quantity of plutonium produced.

The concentrations of certain trace isotopes in structural materials change with fluence. Measuring them allows one to deduce local fluence as structural materials remain in the reactor over long periods of operation, ideally the full lifetime. Since the initial concentration of trace elements is typically unknown, using ratios of two isotopes from the same element is necessary. The initial values for isotope ratios are assumed to be the ratio

of natural isotopic abundances.[21] Therefore, it is possible to estimate past plutonium production by measuring suitable isotope ratios in structural materials from a reactor. To estimate ongoing production, the concept of monitor tags was proposed.[22] Monitor tags, still at the conceptual level, would be small elements added to the reactor core. Their design and composition should be such that they have a negligible influence on the neutron flux in the reactor. As of now, there are no defined designs for monitor tags. Because they are custom-made, they can contain isotopic compositions that are ideal for fluence estimation. Such tags would be placed at various locations in the reactor for plutonium production monitoring, ideally where inspectors could easily extract them. Implementing this approach is only possible in cooperation with the reactor operator.

The initial ideas for nuclear archaeology have been proposed as theoretical models. There are models for graphite moderated reactors, called "Graphite Isotope Ratio Method" (GIRM)[23] and for heavy-water moderated reactors.[24] In the GIRM, isotope ratios are measured in samples from the graphite moderator. Various methods have been proposed to deduce total plutonium production from a set of local fluence measurements.[25] The GIRM was successfully demonstrated experimentally using graphite samples from the British reactor in Trawfynydd[26] and the feasibility of the method for the 5-MWe reactor was studied for the case of boron isotope ratio measurement.[27]

Nuclear archaeology in a broader sense can also be used to analyze other questions and other facilities in the nuclear fuel cycle. For example, it can be used to determine whether reactor operation was optimized to produce tritium rather than plutonium,[28] to estimate production of enriched uranium,[29] or to reconstruct reactor histories from reprocessing waste.[30]

Criteria to identify suitable isotope ratios

Not all isotopes can be used for fluence estimation based on isotope ratio measurements. Suitable isotopes are only the small subset of isotopes that satisfy the following criteria:

A. Stable or long half-life (both isotopes).
B. Non-negligible neutron cross section (at least one).
C. Both isotopes from the same element.
D. No significant production pathways from other elements.
E. Not a noble gas.
F. Traces present in sampled material.

The criteria ensure that the change in ratio only depends on the neutron fluence. Without criterion A, the change in ratio would also depend on time.

Without criterion B, the ratio would not change with fluence. Concentrations of different elements can vary from sample to sample even for the same type of material. With criterion C the initial value of the ratio is set by the natural abundance of isotopes. Criterion D ensures that the isotope ratio from one element is not perturbed by isotopes from other elements via production from decay or neutron-induced reactions. Criterion E discards isotopes that might leak, probably only partially, from the sample. Finally, criterion F ensures that the isotopes that make the ratio are present in the material used for measurements. This criterion does not need to be met when designing monitor tags.

Once suitable isotope ratios are identified based on these criteria, the next step is to derive the relationship between a ratio and fluence. The most useful ratios are those which can be translated to local fluence with minimum uncertainties. Multiple factors affect fluence uncertainties: uncertainties on the reactor design and operation, uncertainties on the nuclear data and the neutronics model and the uncertainties on the measurement of the isotope ratio.[31] Previous work studied the propagation of some of these uncertainties on fluence estimates.[32] The next section provides a rigorous mathematical framework to select isotope ratios that minimize uncertainty propagation from ratio measurements to fluence estimates.

Relative errors on measurement and fluence

To first order, any error ΔR on the ratio R translates into an error on the estimated fluence $\Delta\Phi$ as described in Equation (2).

$$\Delta\Phi = \left|\frac{\delta\Phi}{\delta R}\right|\Delta R \tag{2}$$

Error for a measured ratio is typically given in terms of relative error $\frac{\Delta R}{R}$. It is also more useful to compute the relative error on fluence rather than the absolute error. Rewriting Equation (2) gives

$$\frac{\Delta\Phi}{\Phi} = \left|\frac{\delta\Phi}{\delta R}\right|\frac{R}{\Phi}\frac{\Delta R}{R} \tag{3}$$

It is practical to define the *sensitivity factor* as

$$\mathrm{SF}_R(\Phi) = \left|\frac{\delta R}{\delta\Phi}\right|\frac{\Phi}{R} \tag{4}$$

with which Equation (3) can be changed to

$$\frac{\Delta\Phi}{\Phi} = \frac{1}{\mathrm{SF}_R(\Phi)}\frac{\Delta R}{R} \tag{5}$$

The magnitude of the relative error on fluence not only depends on the magnitude of the relative error on the ratio but also on the sensitivity

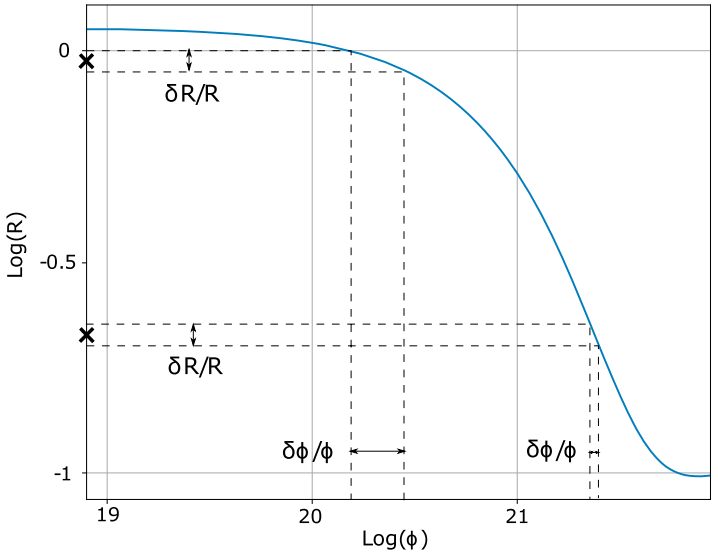

Figure 3. Illustrative logarithm of a ratio over the logarithm of the fluence. Relative errors are represented by width on the axes. Two ratio measurements are shown, indicated by "×" markers on the ordinate axis. They lead to different relative errors on fluence.

factor.[33] For a fixed relative error on the ratio, it is best to increase the sensitivity factor as much as possible. The factor depends on the neutronics properties of the isotopes in the ratio (one-group cross sections) and is also a function of fluence. When selecting the isotope ratios with the highest sensitivity factor, it is important to specify the fluence range over which their sensitivity factor is considered sufficiently high.

The physical meaning behind the sensitivity factor can be easily grasped by considering that it is the derivative of the logarithm of the ratio against the logarithm of the fluence as can be seen in the following equation:

$$\mathrm{SF}_R(\Phi) = \left|\frac{\delta R}{\delta \Phi}\right| \frac{\Phi}{R} = \left|\frac{\delta R}{\delta \Phi}\frac{\Phi}{R}\right| = \left|\frac{\delta R}{R}\frac{\Phi}{\delta \Phi}\right| = \left|\frac{\delta \log R}{\delta \log \Phi}\right| \tag{6}$$

In other word, the sensitivity factor is the slope of the evolution of the logarithm of the ratio against the logarithm of the fluence. Figure 3 illustrates how the sensitivity factor amplifies or dampens the propagation of relative errors from ratio measurement ($\frac{\delta R}{R}$) to local fluence ($\frac{\delta \Phi}{\Phi}$).[34]

In this plot, a relative error on the ratio, $\frac{\delta R}{R}$ is equal to a difference in the logarithm of the ratio $\delta \log(R)$. The same applies to the relative error on fluence. Therefore, relative errors can be represented by widths on the x-axis or the y-axis. The ratio is measured at two different times in the operation life of the reactor. It is also assumed that these two measurements are made with the same relative error; the width representing the relative error on these two measurements is therefore equal in the graph. It can be observed that when the slope of the logarithmic ratio evolution is steeper,

the propagated relative error on fluence is smaller. On the other hand, the relative error on fluence is magnified when the slope gets flatter. Since the slope of the logarithmic curve and the sensitivity factor are the same, this observation coincides with Equation (3).

The use of the sensitivity factor to assess the quality of an isotope ratio for fluence estimation has a clear advantage: it does not change when the ratio is inverted. It is physically evident that the sensitivity of the ratio $\frac{{}^{A}_{Z}X}{{}^{A+1}_{Z}X}$ should be the same as the sensitivity of the ratio $\frac{{}^{A+1}_{Z}X}{{}^{A}_{Z}X}$. The linear slope of a ratio evolution $(\frac{\delta R}{\delta \Phi})$, however, changes when the ratio is inverted. Therefore, the linear slope of a ratio evolution should not be used to assess the sensitivity of the ratio to fluence changes.

On the other hand, using the mathematical expression of the sensitivity factor, it can be shown that it remains unchanged when the ratio is inverted:

$$\begin{aligned} \mathrm{SF}_{R^{-1}}(\Phi) &= \left|\frac{\delta R^{-1}}{\delta \Phi}\frac{\Phi}{R^{-1}}\right| = \left|\frac{\delta \log\left(R^{-1}\right)}{\delta \log\left(\Phi\right)}\right| = \left|\frac{-\delta \log\left(R\right)}{\delta \log\left(\Phi\right)}\right| = \left|\frac{\delta \log\left(R\right)}{\delta \log\left(\Phi\right)}\right| \\ &= \mathrm{SF}_{R}(\Phi) \end{aligned} \tag{7}$$

Best fluence indicators for the 5-MWe reactor

This section derives fluence indicators for two different verification cases. First, the historical case where international inspectors are allowed to go to North Korea to verify past plutonium production declarations based on samples taken from the reactor's graphite. Second, a case of future monitoring, which could be used in the context of denuclearization or arms control efforts. Here, monitor tags would be installed and collected in subsequent inspections.

The nuclear archaeology module of ONIX ("NAX module") was used to identify and choose the best fluence indicators for both cases, graphite sampling and monitor tags. For both the graphite sampling and for the monitor tag approach, a small cylinder with a diameter of 2 mm was defined in the geometry of the simulation on the surface of the graphite block. This is a likely location for tags as well as sampling of irradiated graphite. For the case of graphite sampling, this cylinder was made of graphite with corresponding impurities. In the case of the monitor tag, the tag was made of the same material as the cladding (MAGNOX alloy) and was enriched with minute quantities of all suitable isotopes for fluence estimation. Both the small size and the limited quantities of isotopes ensured that such a tag did not cause significant disturbances in the neutronics of the reactor, nor relevant self-shielding effects within the tag.

The NAX module has three main functions:

1. Identify isotope chains from the same element that have at least two stable or long-lived ($T^{1/2} >$ 10,000 years) members, of which at least one must have a non-negligible neutron cross section (Criteria A, B, and C);
2. Deplete these chains according to an operation history defined by the user, with initial ratios set according to natural abundances; and
3. Compute the evolution of isotope ratios from these chains and their associated sensitive factors against fluence.

To increase computational efficiency when depleting these chains for operation histories that span multiple refueling cycles, a reference simulation to a burnup level of 700 MWd/t provides neutronics parameters such as one-group neutron flux and cross sections. These parameters are then fed into a module that solves a set of Bateman equations to calculate the densities of each isotopes of the chains over the full operation history.[35] The full history can consist of multiple refueling cycles, each with different burnup.[36]

The same infinite lattice simulations as described in "Estimating Plutonium Production" provided neutronics parameters for both cases. It can thus be assumed that these results represent the average behavior of the full core. Given an uncertainty of 0.1% on the ratio measurement (for all ratios tested), isotope ratios were considered good fluence indicators if the resulting relative error on fluence was below 1%.

It is reasonable to assume that the relationships that are presented between isotope ratios, fluence and plutonium production remain valid despite the assumptions and approximations made in this work on the reactor operation and its modeling.

Past plutonium production

To verify past plutonium production in the 5-MWe reactor, isotope ratios would be measured from trace impurities in the graphite moderator. Table 2 lists impurities that were found in two reactor graphite samples.[37] Here, it is assumed that the graphite components of the 5-MWe reactor have not been removed since the beginning of the operation of the reactor in 1986 and can thus be used as a fingerprint of the operation history of the reactor.[38]

The operation history used was the same as described in "The 5-MWe Reactor in Yongbyon" and Figure 1 (main scenario). For each campaign, the average between lower and higher burnup bounds was taken. Figure 4 presents results for selected isotope ratios. As can be seen in the upper graph, several ratios could be used to measure fluence with a low relative error. The ratio of boron has the lowest relative error on fluence at the end

Table 2. Impurities found in two samples of reactor graphite.

	C reactor ppm	G-2 reactor ppm
Barium	0.007	Not measured
Boron	0.10	0.20
Calcium	0.22	30.0
Chlorine	7.8	3.5
Chromium	0.003	Not measured
Dysprosium	<0.001	0.010
Europium	Not measured	0.0007
Iron	0.19	2.0
Lithium	0.003	Not measured
Samarium	<0.01	0.017
Strontium	0.002	Not measured
Titatium	0.001	5.0
Vanadium	0.015	32.0

The C reactor graphite has been irradiated in the U.S. Hanford C reactor while the G-2 reactor graphite has been irradiated in the French G-2 reactor.

of the operation history. Alternatively, the ratios of dysprosium-163 over dysprosium-164 or samarium-147 over samarium 148 could be used. All other potential isotope ratios have relative errors on fluence greater than 1% at a fluence of the order of 2×10^{21}cm/cm^3 (track length over unit volume).

The ratio of samarium-148 over samarium-149 is shown to illustrate a special case. This ratio increases rapidly until it reaches a near-equilibrium at a fluence level of 0.4×10^{21} cm/cm^3 (not shown on the graph). The density of samarium-148 changes slowly with fluence. The density of samarium-149 increases rapidly until its destruction rate from (n, γ) reactions equates the production rate from (n, γ) reaction at which point samarium-149 reaches equilibrium. It can be observed that its associated relative error on fluence is low over the same fluence range but presents a sharp increase when approaching 0.4×10^{21}cm/cm^3. When a ratio reaches equilibrium, its value remains the same despite increasing fluence. The ratio is not sensitive to fluence change anymore and the sensitivity factor goes to infinity (and so does the relative error on fluence). The ratio of samarium-148 over samarium-149 is not suitable for current measurements but would be a good fluence indicator, however, for scenarios where North Korea replaced the irradiated graphite with fresh graphite, for example just before the last production campaign of 2016–2018.

Different mass spectrometry technologies are suitable to measure the discussed isotope ratios. In past nuclear archaeology demonstrations, researchers employed thermal ionization mass spectrometry (TIMS)[39] and secondary ion mass spectrometry (SIMS).[40] Recently, the use of inductively coupled plasma mass spectrometry (ICP-MS) has been discussed for nuclear archaeology.[41] The required sample size to reliably detect the individual isotopes and thus the ratio depends on a number of parameters.

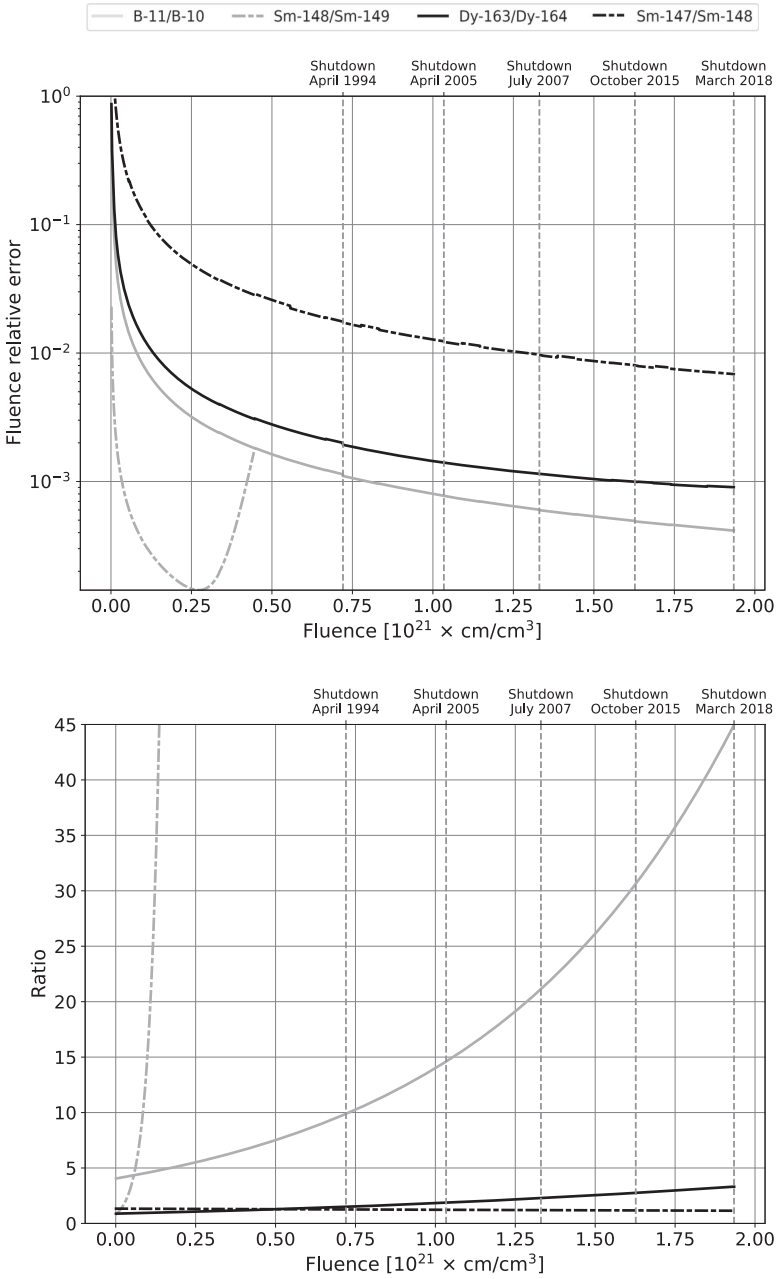

Figure 4. Fluence relative error associated with selected isotope ratios found as impurities in graphite (upper graph) and the evolution of these ratios (lower graph) over fluence. A ratio measurement uncertainty of 0.1% is used to compute the relative error on fluence.

These include potential losses during sample preparation, ion yield of specific isotopes and overall experimental efficiency.

Table 3 presents absolute isotopic concentrations of boron, samarium, and dysprosium in graphite at the end of the operation history. Concentrations

Table 3. Isotopic concentration in irradiated graphite for selected fluence indicators.

	C reactor		G-2 reactor	
	Initial content [ppm]	Final content [atoms/gram]	Initial content [ppm]	Final content [atoms/gram]
B-10	0.10	8.9×10^{13}	0.20	1.8×10^{14}
B-11		4.0×10^{15}		8.0×10^{15}
Sm-147	<0.01	$<7.0 \times 10^{13}$	0.017	1.2×10^{14}
Sm-148		$<6.1 \times 10^{13}$		1.0×10^{14}
Dy-163	<0.001	$<1.4 \times 10^{13}$	0.01	1.4×10^{14}
Dy-164		$<4.1 \times 10^{12}$		4.1×10^{13}

Two initial concentrations are assumed for each element: the one found in the Hanford C reactor graphite and the other one found in the G-2 reactor as described in Table 2.

were computed by ONIX following the previously described irradiation history, up to a fluence level of 1.95×10^{21} cm/cm^3. Final isotopic densities computed by ONIX range from 10^{12} to 10^{15} atoms per gram of graphite. One gram is the size of samples used in past nuclear archaeology experiments.[42] The concentrations are significantly higher then the detection limits of TIMS and ICP-MS described in a study on age determination of uranium.[43] While the study considered actinides, sample sizes of the order of nanograms have also been discussed for example for boron isotope ratios.[44] All isotopes listed in the table have a concentration of more than one nanogram per gram of graphite sample.

Assuming that the simulated unit cell is representative for the average fuel behavior, it is possible to relate the evolution of these ratios to total plutonium production in the reactor. As discussed in the previous section, ONIX burnup simulations yielded a plutonium production of 77 kg over the whole operation history of the reactor. Figure 5 presents the evolution of the ratios of boron-11 over boron-10, samarium-147 over samarium-148 and dysprosium-163 over dysprosium-164 on a logarithmic scale against total plutonium production from these calculations. If in the future inspectors are able to measure isotope ratios in the reactor, they could use this figure as a first order approximation to deduce total plutonium production before engaging in more detailed nuclear archaeology analysis.

To demonstrate the practical use of these ratios, simple functions have been fitted to the curves for the dependence of plutonium production on these ratios. These expressions can be effectively used to relate a measured ratio to the plutonium production in the reactor:

$$m_{\mathrm{Pu}}(R_{\mathrm{B}}) = 31.960 \log(R_{\mathrm{B}}) - 44.971 \tag{8}$$

$$m_{\mathrm{Pu}}(R_{Sm}) = 363.48 R_{\mathrm{Sm}}^2 - 1311.1 R_{\mathrm{Sm}} - 1102.2 \tag{9}$$

$$m_{\mathrm{Pu}}(R_{\mathrm{Dy}}) = 2.0347 R_{\mathrm{Dy}}^3 - 18.438 R_{\mathrm{Dy}}^2 + 79.108 R_{\mathrm{Dy}} - 56.202 \tag{10}$$

In these equations, R_{B}, R_{Sm}, R_{Dy} stand for the respective isotope ratios as discussed above and m_{pu} is the total mass of plutonium in kg.

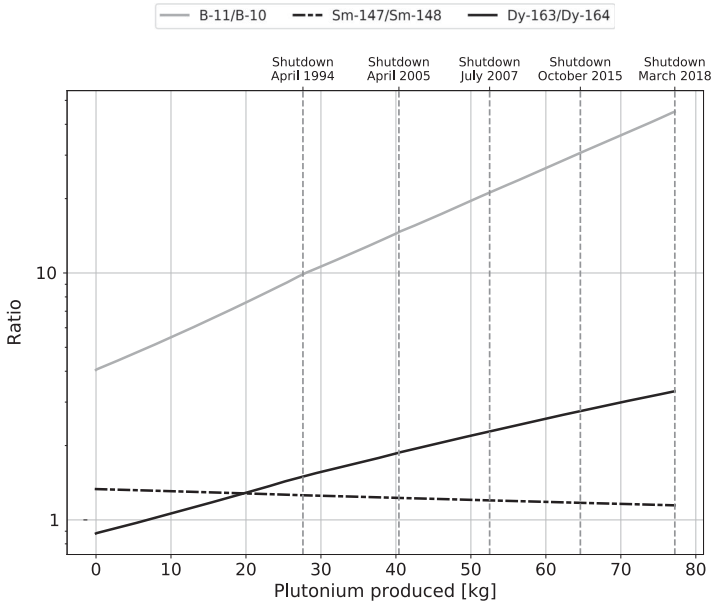

Figure 5. Evolution of the ratios of boron-11 over boron-10, samarium-147 over samarium-148 and dysprosium-163 over dysprosium-164 against total plutonium production. In order to clearly compare the rates of evolution of the ratios with no distortions due to their magnitudes, a logarithmic scale was used for the y-axis.

Table 4. Absolute error on total plutonium production associated with different isotope ratios and various measurement errors.

Relative error on ratio	0.05%	0.1%	0.5%	1%	2%	Ratio value
B-11/B-10	0.031 kg	0.062 kg	0.31 kg	0.6 kg	0.91 kg	44.9
Sm-147/Sm-148	0.51 kg	0.80 kg	2.9 kg	5.6 kg	11 kg	1.15
Dy-163/Dy-164	0.067 kg	0.13 kg	0.63 kg	0.97 kg	1.6 kg	3.32

These absolute errors are to be compared to an estimated total plutonium production of 77 kg.

Table 4 presents the absolute errors on total plutonium production estimates caused by different measurement errors on the three ratios. These results were obtained by using the main scenario for the operation history of the 5-MWe reactor previously described, using a final burnup level corresponding to the average between the lower and upper burnup bound. It is important to note that other uncertainties (e.g., on reactor operation or reactor modeling) influence results for nuclear archaeology.[45] Such uncertainties are not considered here. When reading this table, it is useful to consider that North Korea has been assumed to manufacture nuclear weapons with 5 kg of plutonium or less. Uncertainties obtained with the ratio of boron remain low even for measurement uncertainties as high as 2%. For samarium, plutonium uncertainties become greater than 5 kg for measurement uncertainties between 0.5% and 1%. It can be concluded that the ratio of samarium can only be used to verify plutonium if low measurement uncertainties are achievable.

Monitor tags

To verify on-going production of plutonium, monitor tags could be placed in a reactor and used to measure isotope ratios on a regular basis (e.g., annually) to verify recent plutonium production. Monitor tags can be enriched with chosen isotopes or elements as long as they do not affect reactor performances.

In the case of the 5-MWe reactor, the DPRK could request continued operation for other purposes than weapon-grade fissile material production, and in exchange allow inspectors to conduct inspections to install and analyze monitor tags. However, future agreements with the DPRK might seek to terminate operation, similar to prior shutdown periods. The results presented here show the technical feasibility of the monitor tag concept for the 5-MWe reactor, and ONIX capabilities to identify adequate isotope ratios for monitor tags. The results can also be useful for an assessment of monitor tags in other graphite-moderated reactors.

The potential monitor tags are assumed to be placed on the surface of graphite blocks, within fuel channels. Isotopic chains from monitor tags were depleted over three years of operation with a capacity factor of 75% and a power of 22.5 MWth. This could, for example, reflect inspection intervals of 1 to 3 years.

Figure 6 shows results for isotopes that present a strong potential for fluence estimation with monitor tags. Two hundred and fifteen ratios were reviewed with the NAX module of ONIX. Of these, the three isotope ratios shown in Figure 6 were selected because they assume the lowest relative error on fluence at one, two, and three years of operation, respectively. The ratio of gadolinium-158 over gadolinium-157 can be efficiently used to estimate plutonium production after one year of operation. The ratio of samarium-148 over samarium-149 can be used for two years and the ratio of cadmium-114 over cadmium-113 can be used for three years of operation. For comparison, the ratio of boron is also plotted.

Table 5 presents absolute errors on total plutonium obtained by measuring the ratio of gadolinium after one year of operation, the ratio of samarium after two years of operation and the ratio of cadmium after three years of operation with various measurement uncertainties. As for Table 4, other uncertainties are not considered here. All three ratios allow to estimate plutonium with high accuracy even for measurement uncertainties as high as 2%. The maximum relative error obtained on plutonium is 0.7% when measurement is done after one year with gadolinium and with 2% uncertainty. These results prove that efficient monitor tags can be designed with these isotopes for routine verification of plutonium production in the 5-MWe reactor.

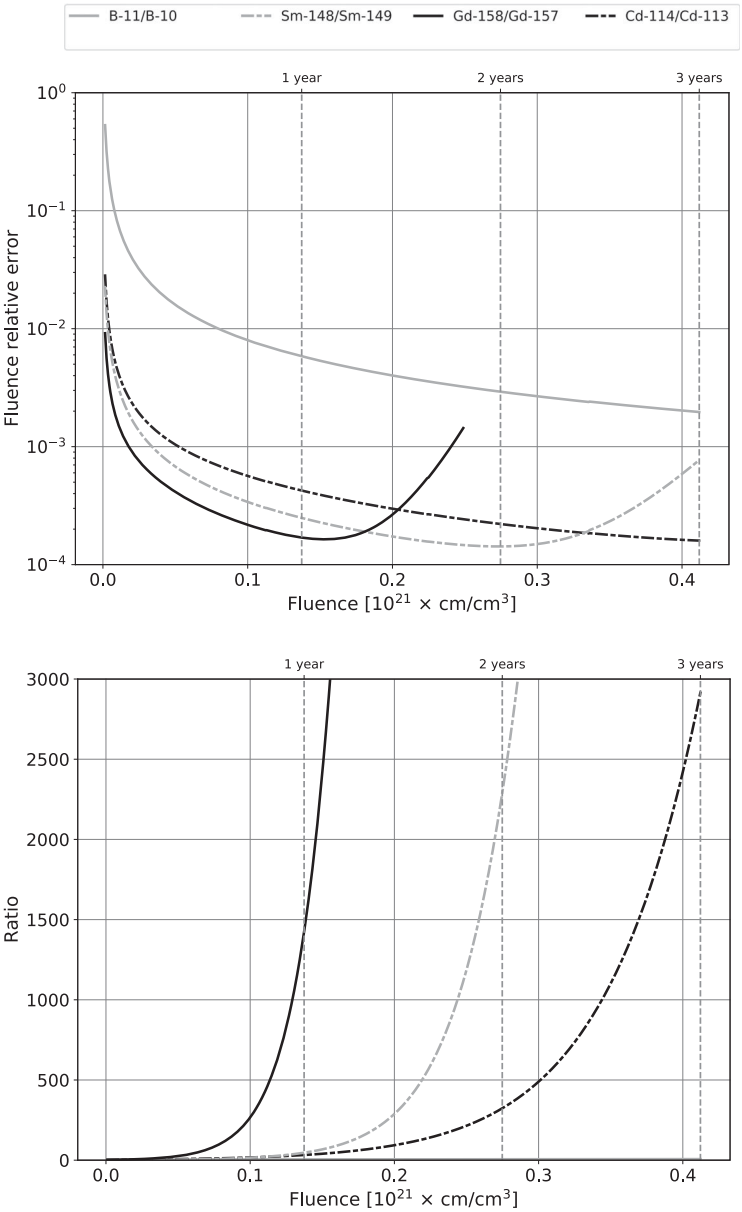

Figure 6. Fluence relative error associated with some isotope ratios (upper graph) and the evolution of these ratios (lower graph). The monitor tag was exposed to an operation history of three years with a capacity factor of 75% and a power of 22.5 MWth for the reactor. A ratio measurement uncertainty of 0.1% is used to compute the relative error on fluence.

Table 5. Absolute error on total plutonium production associated with different isotope ratios for one, two and three years of operation.

Relative error on ratio	0.05%	0.1%	0.5%	1%	2%	Plutonium produced	Ratio value
1 year							
Gd-158/Gd-157	0.97 g	1.9 g	9.7 g	19 g	39 g	5.8 kg	1,424
2 years							
Sm-148/Sm-149	1.5 g	3.1 g	15 g	31 g	62 g	11 kg	2,290
3 years							
Cd-114/Cd-113	2.5 g	4.9 g	24 g	49 g	98 g	17 kg	2,916

Conclusion

New estimates of historic plutonium production in North Korea are presented in this work. These estimates have been obtained with ONIX, an advanced and open-source reactor simulation tool, and agree well with estimates obtained by other experts. As open-source software, anyone, including inspectors and authorities from countries involved in arms control processes on the Korean peninsula (even North Korean experts) can replicate and further this analysis.

A new mathematical framework for the selection of best isotope ratios for nuclear archaeology in reactors is presented and implemented in the NAX module of ONIX. This new selection process allows for the identification of isotope ratios that minimize the propagation of uncertainties from ratio measurements to plutonium production estimates. While a real implementation of plutonium estimation at the 5-MWe reactor with nuclear archaeology would also be influenced by other forms of uncertainties, this study shows that uncertainties from ratio measurements and relative errors on fluence can be efficiently controlled by selecting appropriate fluence estimators with the NAX module of ONIX.

This work is primarily a demonstration of the usefulness of new methods presented. Further implementation of these concepts would require more detailed, full-core reactor analysis that takes into account spatial variations in the reactor. To further improve the results found for nuclear archaeology, sensitivity analyses should be carried out looking at various modeling parameters, including, for example, temperature or geometry. Future work should also investigate the potential of the selection process for fluence indicators presented in minimizing the propagation of other sources of uncertainties on plutonium production such as uncertainties from simulation models and past operation.

Nuclear archaeology for past plutonium production can help verify baseline declarations and support routine verification of North Korea's nuclear reactors. As such, the methods and tools presented in this work could be used in future disarmament or arms control efforts on the Korean peninsula.

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We are grateful for the continuous support by Alexander Glaser throughout the development of ONIX, and the help from Siegfried Hecker with regards to the details on the North Korean program. Calculations were carried out using Sherlock, the Stanford University Cluster. Julien de Troullioud de Lanversin received generous funding from the Stanton Foundation as well as the John D. and Catherine T. MacArthur Foundation.

Disclosure statement

The authors have no conflicts of interest to declare.

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Appendix A. Reactor specification

Table A1. Assumed characteristics of the 5-MWe reactor.

Thermal power	20–25 MWth	Effective core diameter	643 cm
Electric power	5 MWe	Effective core height	592 cm
Specific power	0.40–0.50 MWth/tHM	Upper reflector	77.50 cm
Uranium loaded	50 t	Bottom reflector	66.50 cm
Graphite-moderator	300 t	Fuel composition	U (0.5 at% Al)
Graphite-reflector	300 t	Diameter of fuel meat	2.90 cm
Number of channels	812–877	Length of fuel meat	52 cm
Number of fuel channels	801	Length of fuel rod	60 cm
Number of control rod channels	44	Uranium per fuel rod	6.24 kg
Number of fuel rods per channel	10	Cladding composition	Mg (1 at% Al)
Distance between channels	20 cm	Cladding thickness	0.05 cm
Radius of channel	6.50 cm		

Notes: Data taken from Science & Global Security 19 (2011): 121–129.
The parameters and information compiled in this table are those of a Magnox reactor design which is similar to the design of the 5-MWe reactor.