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The Future of Nuclear Archaeology: Reducing Legacy Risks of Weapons Fissile Material

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This report describes the value proposition for a "nuclear archaeological" technical capability and applications program, targeted at resolving uncertainties regarding weapons fissile materials production and use. Central to this proposition is the notion that one can never be sure that all fissile material is adequately secure without a clear idea of what "all" means, and that uncertainty in this matter carries risk. We argue that this proposition is as valid today, under emerging state and possible non-state nuclear threats, as it was in an immediate post-Cold-War context, and describe how nuclear archaeological methods can be used to verify fissile materials declarations, or estimate and characterize historical fissile materials production independent of declarations. Methods for accurately estimating plutonium production from graphite reactors have been demonstrated and could be extended to other reactor types. Proposed techniques for estimating HEU production have shown promise and are under development.

SUMMARY

In 1993, Steve Fetter called for a program of "nuclear archaeology" to clarify the historical record of Cold War plutonium and highly enriched uranium (HEU) production.[1] At that time, it was widely believed that the United States and Russia would offer reciprocal declarations of weapons materials production, and that these declarations would lead to opportunities for technical verification.

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A study of this concept, funded by the Department of Energy Office of Research and Development (DOE/NN-20) and conducted by Pacific Northwest National Laboratory (PNNL) with support from the Oak Ridge Site, examined the technical feasibility of verification measurements and analysis for both plutonium and HEU production. This study led to analytical and experimental programs that demonstrated a highly accurate isotope-ratio technique to determine plutonium production in graphite reactors.

The result is an operational capability for sampling, sample preparation, analytical chemistry, and reactor physics that yields lifetime plutonium production estimates with standard errors of less than 2% for well-characterized graphite reactors.[2] An estimate for the total plutonium production of several graphite reactors would have even greater accuracy due to aggregation of independent error terms.[3] This technique has been generalized to other reactor designs and made practical by refining sample preparation techniques and isotope ratio measurements.

Although substantial progress has been made in the last 20 years on verifying plutonium production, little has been accomplished or even attempted on estimating and verifying HEU production. The initial PNNL feasibility study found no readily exploitable signatures from which uranium production histories could be directly inferred, and subsequent active research on the HEU verification challenge was suspended as resources were devoted to developing methods for plutonium verification. Today, PNNL and others are re-evaluating the technical feasibility for verification, or independent estimation, of HEU production.[4] There are compelling reasons to re-examine this question.

Uncertainty about historical HEU production in the former Soviet Union is much greater than for plutonium. The best estimates of Russian HEU production have an uncertainty of 120 MT, representing thousands of International Atomic Energy Agency (IAEA) significant quantities. In contrast, Anatoli Diakov estimated uncertainty in Russian plutonium production to be 8 MT.[5] Any hope of assurance that "the entire stockpile" of HEU has been disposed of or entered into a safeguards regime will require more definitive accounting than is currently available.

The Fissile Material Cutoff Treaty (FMCT) as recently envisioned within the Conference on Disarmament does not address stocks of fissile materials from past production. At this time, the FMCT is regarded as feasible only by the five permanent members of the United Nations Security Council (P-5), none of which face any constraint on nuclear weapons doctrine or efficacy from a cut-off of fissile material production. A robust capability (including verification methods for HEU production) would enable a much more complete fissile materials regime, with respect to both the scope of the treaty, and possibilities for technical verification. Such a treaty among the P-5 states might ultimately lead to a broader fissile materials transparency regime.[6]

The world in which nuclear archaeology[7] might be applied to the problem of fissile materials transparency is a far different one than envisioned when the term was coined shortly after the break-up of the Soviet Union. In particular, the level of trust between the United States and Russia, while still relevant, may not be the key driver in requiring fissile material stockpile verification. Rather, the joint U.S.–Russian responsibility to set a high standard and example for the rest of the world, and a desire for a firm technical footing for state-level fissile materials accounting on a global basis, may be the key motivating factors in methods development and early applications.

ORGANIZATION OF THE PAPER

This paper is organized into the following sections.

- **Nuclear Archaeology—A Summary History:** provides a brief summary history of nuclear archaeological methods developed at PNNL and other laboratories over the last twenty years, focused primarily on verification of plutonium production, and concludes with a summary status of archaeological methods status by technology.
- **Current State of Knowledge of Fissile Materials Production:** summarizes the current knowledge of worldwide fissile material production at the country level, including quantitative estimates of uncertainty for various production and inventory quantities.[8] This represents the current baseline from which a program of fissile materials transparency would begin.
- **Generalization of the Nuclear Archaeology Concept:** describes the utility of the concept beyond verification of production to include accounting for both utilization and disposition of fissile material. This reflects the fact that both arms controls and disposition of surplus fissile material will require long time frames.
- **Prospective Accuracy in Nuclear Archaeology:** examines how uncertainty in current estimates might be reduced over time—to be minimized at the all-important "end-game" stage of nuclear disarmament.
- **Summary and Findings:** provides specific recommendations for near term actions.

NUCLEAR ARCHAEOLOGY—A SUMMARY HISTORY

The pursuit of nuclear archaeology was motivated by the principles that: 1) uncertainty regarding fissile materials management leads inevitably to increased risk and 2) the greatest prospective risk reduction should guide investments in this area. Given that the consequences of nuclear materials diversion (either

within a state or to a group seeking nuclear capability) are unquestionably as large as they ever were, reduction of uncertainty regarding fissile materials production, stockpiles, and assured disposition *is the agenda* for nuclear archaeological methods. Application of these principles requires we begin from the best possible accounting available today.

The original feasibility study performed at PNNL and the Oak Ridge Gaseous Diffusion Plant concluded that isotopic ratios in activation products of neutron irradiation of impurities in reactor graphite offered a promising signature that might be exploited to estimate neutron fluence, and thus cumulative plutonium production. Over the almost 20 years since this study, a series of increasingly challenging and sophisticated experiments have been conducted to extend, verify, and generalize the sampling and analytical basis for plutonium production estimates. The technique has now been demonstrated in a range of isotopes, collectively offering sensitivity over a wide range of neutron fluence. In addition to strictly experimental advances, the program has built an operational capability for field sampling and analysis. Table 1 summarizes some of the key milestones in this program. Most notably, this program demonstrated, at reactor scale, an isotope ratio technique[9,10] with very high accuracy for estimating plutonium production in graphite reactors.[11]

This technique has been generalized to other reactor designs using metallic samples from core structural components, and made much more practical by refinements of the analytical chemistry techniques for sample preparation and measurements of isotope ratios.

The isotope ratio techniques for quantifying reactor production of plutonium have several key features from an applications perspective. First, they have been developed and published largely on an unclassified basis. Thus, the underlying phenomenology is accessible to any interested parties, and the credibility of the method is well founded and general. The nuclear cross-sections and natural isotopic abundances that govern the accuracy of the technique are known with high confidence and precision, and published in standard nuclear libraries. Second, the technique can be based on the ratios of isotopes in several elements (including boron, calcium, hafnium, thallium, plutonium, and uranium) , making it broadly applicable to both graphite reactors, where impurity composition varies widely but typically includes multiple useful indicator elements, and other reactors, which typically contain useful indicators in metallic alloys in core structural components. Third, the use of multiple indicator elements in a given reactor can be used to greatly reduce error and adds redundancy to the method. Finally, the isotope ratios are between stable or very-long-lived isotopes, making the "signal" a permanent one, rather than one that decays over time.

While research and development remains to be done on plutonium production verification, the work of the last 20 years has established it as a relatively mature capability, with prospective accuracy that is thought to be better

Table 1: Nuclear archaeology timeline.

Date	Milestone
1992	Steve Fetter publishes first paper on "nuclear archaeology"
1993	PNNL feasibility project established, isotope ratio method for graphite reactors first considered
1993	Simulation studies at PNNL show promising prospective accuracy
1994	First elemental characterization studies on PNNL graphite archives
1994	Proof of principle for graphite isotope ratio method (GIRM) demonstrated with Hanford C-reactor and French G2 irradiated reactor graphites
1994	Thermal ionization mass spectrometry (TIMS) methods developed and employed for titanium and calcium as indicator elements
1995–96	Full scale demonstration of Grim at U.K. Trawsfyndd-II commercial Magnox reactor
1997	Efforts to extend GIRM application to low fluence reactors
1998	Secondary ionization mass spectrometry (SIMS) methods developed to use boron as an indicator element for low fluence applications
1999	TIMS methods developed to use uranium and plutonium as indicator elements primarily for low fluence applications
2000	Proof of principle demonstration at U.K. VEPO reactor for low fluence applications of GRIM
2001	Development of GIRM specific graphite sample acquisition equipment
2002	Establishment of U.K. based graphite sampling team and equipment
2002	Method development begins for extension of isotope ratio methods to other reactors, such as research reactors
2003	Development of SIMS capability to assess indicator elements chlorine, titanium, and boron as well as TIMS for uranium and plutonium in activated metals
2004	Proof of principle demonstration for IRM application to Michigan's Ford research reactor
2005–06	Proof of principle demonstration for IRM application to Russian designed research reactor (Tbilisi, Georgia)
2007	Qualified GIRM laboratories established throughout U.S. and U.K. complexes
2010	Demonstration of IRM sample acquisition equipment for research reactors
2011	Establishment of U.S. based graphite sampling team and equipment
2012	Proof of principle application of IRM to heavy water reactors

than the official U.S. estimate of plutonium production, and significantly better than unofficial estimates of Russian weapons plutonium production. From a research and development and methods development perspective, the problem of estimating historical HEU production is both more technically demanding and far less experimentally mature.[12]

Table 2: Extent of fissile material production technology deployment and use.

	Number of countries	Number of plants	Production
	Plutonium production		
Graphite-moderated reactors	7	>25	>250 tons
Heavy-water reactors	10	>18	>100 tons
Other reactors	4	4	<100 Kgs
Accelerator-driven systems	2	2	a few Kgs
	Reprocessing		
Plutonium separation facilities	11	>19	>350 tons
	HEU production		
Centrifuge	7	11	>1000 tons
Gaseous diffusion	5	8	>1000 tons
Other enrichment technologies	3	3	>100 kg

Tables 2 and 3 summarize the extent of fissile material production technologies throughout the world and the status of archaeological methods for each. The figures in Table 2 are approximate and intended only to give a sense of the importance of the technology. As indicated in Table 3, there is *some* work complete or underway on almost all technologies of importance, but substantial research and development work remains to be done to make archaeological methods generally available for use as production verification tools, particularly for uranium enrichment. The current frontier of experimental work on this problem is described in the section on prospective accuracy in nuclear archaeology.

CURRENT STATE OF KNOWLEDGE OF FISSILE MATERIALS PRODUCTION

Quantifying the amount of fissile material (defined as HEU and weapons-grade separated plutonium) produced and currently stockpiled is a necessary first step toward any nuclear security, nonproliferation, or disarmament efforts between the declared nuclear weapons states. Without accurate and verifiable disclosures or estimates (within acceptable limits of uncertainly), accounting for fissile materials poses a serious challenge. This section describes current knowledge of fissile materials at the country level, with an emphasis on the degree of certainty with which production and inventories are known.

As of January 2012, the reported national stockpiles of fissile materials far exceeded weapons needs in the nuclear weapons states. Table 4 provides a

Table 3: Status of archaeological methods development by production technology.

Plutonium production	Status of archaeological methods
Graphite-moderated reactors	GIRM - well developed, verification at reactor-scale
Heavy-water Reactors	IRM of process tubes, demonstrated on research reactors
Other reactors	IRM of structural components, demonstrated on research reactors
Accelerator-driven systems	Conceptual approach
	Reprocessing
Plutonium separation facilities	Isotopic analysis of holdup materials, reprocessing waste volumes, gas sampling to determine plutonium inventories
	HEU production
Centrifuge	Experimental work with surrogates and plant components underway
Gaseous diffusion	Proposed scheme based upon UO_2F_2 deposition
Other enrichment technologies	Conceptual approach

summary of the current fissile material stocks obtained from the International Panel on Fissile Materials.[13] The United States and the Russian Federation currently hold over 95% of the current stockpiles of fissile materials. However, official sources do not account for any production uncertainties and, in the case of two countries, provide only inventory discrepancies. These two measures of

Table 4: Fissile material stockpiles, estimates by country (International Panel on Weapons Fissile Materials).

	HEU (MT)		WG-Pu (MT)	
	Stockpile	Uncertainty	Stockpile	Uncertainty
Russia	737	120	128	8
France	30.6	6	6	1.8
China	16	4	1.8	0.5
Pakistan	2.75		0.14	
India	2		.05	
North Korea			0.03	
Others	20.0		—	
	HEU (MT)		W G-Pu (MT)	
	Stockpile	Inventory difference	Stockpile	Inventory difference
United States	740.7	3.2	95.4	2.4
United Kingdom	21.2	.022	7.6	0.29

uncertainty, which often are used in the literature on fissile materials production and stocks, are defined below.

- *Production Uncertainty* is the accuracy with which we know the cumulative production of a given material, and is typically given in metric tons (MT) or percentage of production. This measure is most often derived as the standard error (standard deviation) of an estimate, and is influenced by many factors.
- *Inventory discrepancy* is cumulative production minus known uses and current inventory. Inventory discrepancy is a function of both production uncertainty and uncertainty in cumulative disposition, and of any correlation between them. In general, inventory discrepancy is only meaningful for accurate (low uncertainty) production estimates.

The state-of-knowledge for each of the following countries has been sourced from information available from the International Panel on Fissile Materials,[14] except where noted.

Russia shut down its last plutonium production reactor in 2010 and ceased producing HEU for weapons in 1989. Though no official historic accounts of fissile material production and disposition in Russia have been released, two separate analyses[15] by non-governmental agencies have been recently published.[16] As of 2011, Diakov reported Russian stockpiles of 128 $\pm$ 8 MT of weapons-grade plutonium which represents an accuracy of approximately 6% of total production. The uncertainty in these estimates stems primarily from the uncertainties of the power levels of the individual production reactors and the assumed durations of their operation at those power levels. Pavel Podvig published the most definitive estimates of Russian HEU production estimating a stockpile of 737 $\pm$ 120 MT. The reported uncertainty in HEU production is larger than all production outside of Russia and the United States.

China has not officially declared an end to its production of HEU and plutonium for weapons or provided reports of its fissile material production and disposition. However, it is believed that HEU and plutonium production were stopped in 1987 and 1990, respectively. According to a report by Zhang, it is estimated that China produced 2 $\pm$ 0.5 MT of weapons-grade plutonium (WG-Pu) and 20 $\pm$ 4 MT of HEU.[17] These new estimates are significantly lower than most previous estimates, which ranged from 2.1 to 6.6 MT of plutonium and 17 to 26 MT of HEU. As of 2011, it is estimated that China's current stockpile comprises of 1.8 $\pm$ 0.5 MT of WG-Pu and 16 $\pm$ 4 MT of HEU. These production figures indicate the smallest military stockpile of plutonium and HEU available for weapons among the nuclear weapons states, but show significant uncertainties—28% and 25%—for WG-Pu and HEU, respectively.

France has not publicly disclosed its military fissile material stockpiles or production history, and no informal analyses have been published either.

Though France stopped production of WG-Pu in 1992, it was in 1996 that they announced a definite end to the production of fissile materials and stopped HEU production. Based on available information, France's fissile material stockpiles include 6.0 ± 1.8 MT of WG-Pu and 31 ± 6 MT of HEU. According to France's IAEA Information Circular, (NFCIRC)/549 declarations on its civilian holdings, 4.85 MT of HEU were designated for civilian use,[18] leaving a military inventory of is 26 ± 6 MT of HEU and 6 ± 1.8 MT of WG-Pu. Relative to estimated production, these are large levels of uncertainty in current stockpiles, with 30% for WG-Pu and 19% for HEU.

The United Kingdom has publicly disclosed both its current stockpiles of fissile material, and production history for both HEU and plutonium.[19] In 1998 the Ministry of Defense listed 21.9 MT of HEU and 6.6 MT of WG-Pu as current defense stocks of fissile materials. While HEU remains for use in naval propulsion and outside of safeguards, 4.4 MT of WG-Pu was declared excess and placed under EURATOM safeguards.[20]

In 2006, HEU production was reviewed, and the then-current balance was 21.64 MT. This review, which involved a systematic audit of the HEU stocks, revealed a discrepancy of +0.22 MT, implying 0.22 MT less HEU on hand compared to the stockpile estimated from production and prior uses.[21] This discrepancy is attributed to missing records, difficulties in interpreting existing records, and measurement inaccuracies during early production years.

A similar review of plutonium production and inventory revealed a +0.29 MT difference between the book and audited inventories. This discrepancy, as in the case of HEU, was attributed to poor quality and completeness of records, potential missing records, and measurement and sampling uncertainties in the early years of the program due to the state of technology.[22]

The United States ceased production of WG-Pu and HEU in the 1960s, although some reactors primarily for tritium production remained active until the 1980s. By 1987, all DOE production reactors were shut down. In 1996, the United States released a report entitled *Plutonium: The First 50 Years,* in which the U.S. DOE detailed the production, acquisition, and utilization of plutonium from 1944 to 1994.[23] Based on this report, the United States has produced a total of 99.5 MT of WG-Pu.[24] In June of 2012, an updated report entitled *The United States Plutonium Balance, 1944–2009* was issued.[25] According to the updated report, the United States currently had 95.4 MT of plutonium in its inventory and an inventory discrepancy of −2.4 MT over its entire production history. This represents an improvement in accountability over the previous report with both an inventory decrease of 4.1 MT and an inventory discrepancy decrease of 0.4 MT. This inventory discrepancy is often reported as "materials unaccounted for" or simply MUF.

Uncertainty in the U.S. production of plutonium has been attributed to lack of advanced nuclear material management systems and the unavailability of computer-aided tools in the early days of the production program. In 2006,

the Federation of American Scientists, through the Freedom of Information Act, requested that DOE release the report entitled *Highly Enriched Uranium: Striking a Balance*, which documented the history of HEU production in the United States.[26,27] According to the report, the current U.S. stockpile for HEU was 740 MT with an inventory difference of +3.2 MT. This difference is attributed to lack of documentation, documents not designed for HEU accountability, and lack of sufficiently advanced measurement systems in early days of production. However, while the published report provided no explicit information on the accuracy of production estimates for HEU, it provided the HEU production history in kilograms to five significant figures.

Among the five Nuclear Weapons States, there is a wide range in both levels of information released and the degree of confidence or certainty among national estimates and declarations of fissile materials production, uses, and current stocks. While there is a general absence of formal statistical error (accuracy) assessments of production estimates, the *implied* accuracy of these estimates can vary from highly accurate, in the case of the U.S. HEU estimates, to highly uncertain, in the cases of HEU quantities in France, China, and Russia. In each case, the estimates have been derived from a records-based assessment to reconstruct production history from (presumably) the best available records for the many production processes. Furthermore, each report states that that record-based reconstruction is incomplete, inconsistent, or both.

Based on the current state of knowledge for United States and the United Kingdom, it appears that a records-based analysis can yield very accurate estimates for HEU production. Hence, the first step in fissile material transparency should be an in-depth, records-based accounting of HEU production, assuming such records exist.

GENERALIZATION OF THE NUCLEAR ARCHAEOLOGY CONCEPT

The scenario that originally suggested nuclear archaeology was one in which:

- Fissile material production had been the binding constraint on nuclear weapons stockpiles and thus nuclear force capabilities generally.
- There was great uncertainty about an adversary's historical production of fissile material.
- There was a low level of trust in an adversary's prospective production declaration.

All of the above features of the problem have changed substantially, and some new ones have emerged. This section suggests a generalization of nuclear archaeological objectives in this new context.

Beyond extension of methods to cover most or all production technologies, the primary sense in which the concept requires generalization is born in the fact that disarmament and weapons materials disposition is going to be a very long-term problem. Even with rapid recent progress on U.S.-Russian treaties limiting delivery vehicles and warheads, implementation of the New Strategic Arms Reduction Treaty (New START) may require 15 years. The future prospect for this agenda is one of more complex multilateral (or coupled bilateral) treaty frameworks, which are not yet even anticipated in any detail. Furthermore, the problem of materials disposition is also proving to be one that requires substantial investment and time; the U.S. mixed oxide program for weapons plutonium disposition is the most current example of this. While this means we cannot resolve the relevant uncertainties quickly, it also permits time for deliberate, cooperative application of existing techniques, and perhaps development of additional techniques.

Thus, today's context for nuclear archaeology is one of a long-term process rather than one of "initializing" fissile material stock estimates, as the problem was often framed in the early 1990s. One implication of this extended temporal context is that the relevant processes for certainty in fissile materials accounting extend beyond historical production to include disposition. The real figure of merit, as suggested in the Diakov and Podvig papers, is *remaining fissile material available for weapons use*, and the certainty with which we can bound this metric.

In statistical terms, we wish to minimize the error for an estimate of "material available for weapons use" (M_W), where

$$M_W = M_P - M_T - M_L - M_u, \tag{1}$$

In this equation, M_P is production, M_T is testing, M_L is loss (wastes), and M_u is non-weapons use (either as naval fuel for HEU, or "disposition" (e.g., burning plutonium as a mixed oxide, down blending HEU for use as reactor fuel, or converting to safeguarded stocks).

If we acknowledge that all of the terms in Eq. 1 are subject to uncertainty and treat them as random variables, the variance of the figure of merit M_W is

$$\begin{aligned} \mathrm{Var}(M_W) = {} & \mathrm{Var}(M_P) + \mathrm{Var}(M_T) + \mathrm{Var}(M_L) + \mathrm{Var}(M_u) - 2\mathrm{Cov}(M_P, M_T) \\ & - 2\mathrm{Cov}(M_P, M_L) - 2\mathrm{Cov}(M_P, M_u) + 2\mathrm{Cov}(M_T, M_L) \\ & + 2Cov(M_T, M_u) + 2\mathrm{Cov}(M_L, M_u) \end{aligned} \tag{2}$$

Equation 2 tells us that *all* of the uncertainty (variance) terms contribute equally to uncertainty in the figure of merit M_W, and that we must be concerned about the extent of correlation among these four terms. The first of these points indicates that to minimize $\mathrm{Var}(M_W)$, it is inefficient to invest great effort (for example) in scrupulous accounting to ensure that disposition of

plutonium stocks is controlled to the gram level if the uncertainties in other terms are very large by comparison. This statistical perspective makes it clear that nuclear archaeological methods should be applied to any and all of the terms on the right side of Eq. 2, to the extent that they contribute meaningfully to good estimates of (and confident upper bounds on) material available for weapons use.

The covariance terms in Eq. 2 would be zero if all of the estimates were statistically independent. The covariance terms should be assessed when a formal statistical model of any national production program is constructed to determine which ones apply.

PROSPECTIVE ACCURACY IN NUCLEAR ARCHAEOLOGY

This section addresses the prospective accuracy of measurement-based nuclear archaeological estimates for both plutonium and HEU production using Russian production as an example. Methods for quantifying Pu are well established from an accuracy perspective. Since established methodologies for HEU do not exist, the discussion takes the form of identifying the sources of error in the current estimate and their implications for an archaeology program. These examples *do not* imply that only Russian production histories warrant nuclear archaeological applications, but the context (largely due to the work of Diakov and Podvig) is well developed for these two cases.

Plutonium Estimates

Today's methods would support a broad-scale program to apply isotope ratio-based methods to a comprehensive estimate of historical plutonium production in either Russia or the United States. In both cases, the prospective accuracy of a measure-based estimate is better than current estimates. An assessment for applying isotope ratio measurements to Russian plutonium production indicated high prospective accuracy[28] and technical feasibility.[29] Table 5 provides the results of a calculation of the complex-level accuracy that such a program would be expected to provide for Russian WG-Pu production.[30] The error figures (last column in the table) were generated by applying typical graphite isotope ratio method (GIRM) percentage errors for a graphite reactor to the current (historically based) estimate for each reactor. The aggregation of the reactor errors to a total assumes independence. While the U.S. estimate might be slightly less precise because a substantial fraction of its production was in heavy-water moderated reactors, it would be of comparable accuracy.

To better visualize the impacts of such a program in the timeframes appropriate for disarmament, the statistical model described in the previous section is applied to a forecast of uncertainty for our figure-of-merit – the uncertainty

Table 5: Prospective accuracy of a graphite isotope ratio-based estimate for Russian WG-Pu.

Site	Reactor	Current plutonium estimate (Kg) [1]	GIRM SE (Kg)[2]
Mayak	A	6138	199
	AV-1	8508	276
	AV-2	8407	272
	AV-3	7822	253
	OK-180	53	2
			0
Seversk	I-1	8237	267
	IE-2	7453	241
	ADE-3	14020	454
	ADE-4	19460	631
	ADE-5	19144	620
			0
Zelenogorsk	AD	15433	500
	ADE-1	14184	460
	ADE-2	16317	529
Total		145175	1454

[1]Diakov A, "The History of Plutonium Production in Russia," *Science and Global Security* 19 (2011):28–45.
[2]Calculated as the quadratic sum of 2.55 percent standard error for the graphite isotope ratio method and 2 percent for reprocessing losses, or 3.208.

in plutonium available for weapons at any point in time. Figure 1(a) and 1(b) show how projected uncertainty in plutonium available for weapons would evolve in a "do-nothing" base case and in a hypothetical scenarios involving application of GIRM to all Russian production reactors. These figures are based on a history of production derived from Diakov,[31] as well as his estimates of the uncertainties associated with estimates of production. Both use his estimates of plutonium usage in weapons tests and the recent declaration of 34 MT of plutonium as surplus, and derive material available for weapons (blue curve) as the difference in cumulative production and use. Figure 1(a) is a projection of the current state of knowledge on Russian plutonium production to a program in which a disposition program is synchronized with a goal of complete nuclear disarmament by 2050. A key feature of Figure 1(a) is that all of the uncertainty in production is inherited on a permanent basis during the disposition phase. Figure 1(b) illustrates a scenario in which a comprehensive set of reactor estimates was performed in the next two decades.

One useful perspective illustrated from Figures 1(a) and 1(b) is the uncertainty in timing for the point at which nuclear capability is reduced to zero (or any other specified lower limit). In the base case, this window is over a decade

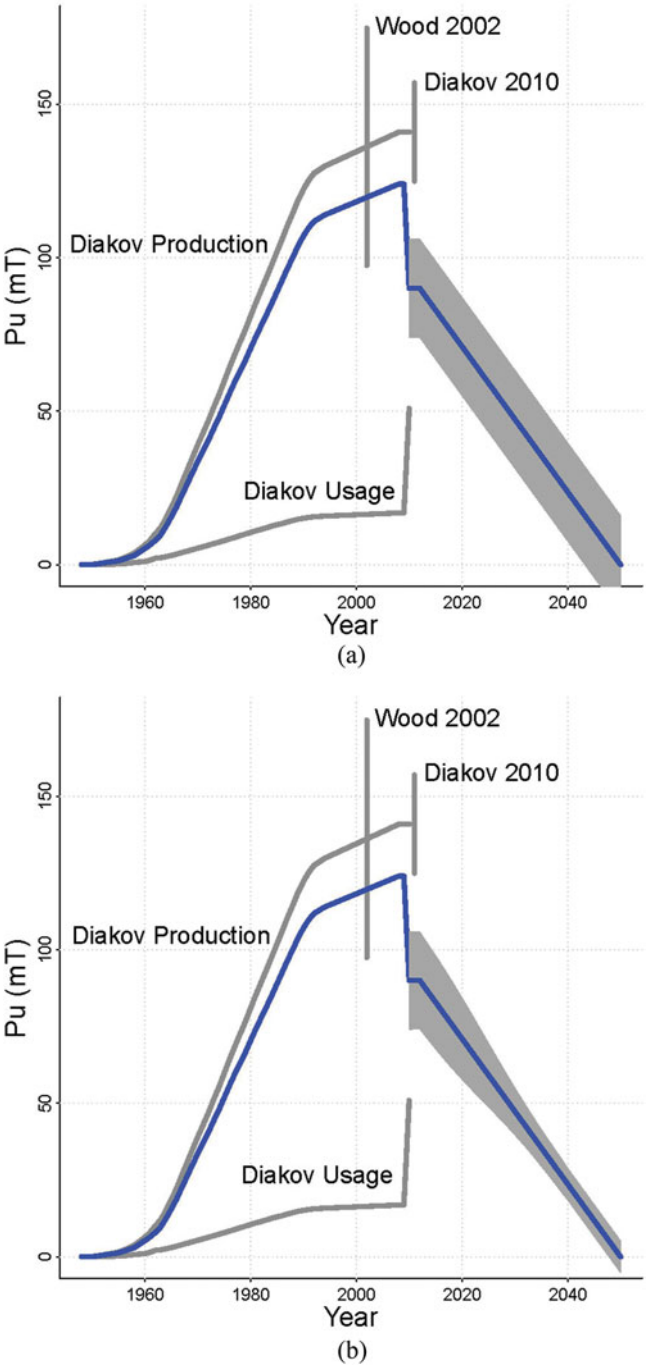

Figure 1: Plutonium production uncertainty for Russia: a) "do-nothing" and b) comprehensive GIRM program (color figure is available online).

long. In the case in which GIRM-based estimates are made for all production reactors, this window is about 3 years.

Nuclear Archaeology Strategy for HEU

To understand the prospective gains in applying archaeological methods to HEU production for weapons, it is necessary to begin with a baseline estimate and its sources of error. For this purpose, we use Podvig' s 2012 estimate for Russian weapons-grade HEU. Based on historical reconstruction of separative work unit (SWU) production, allocation to weapons and non-weapons uses, and uncertainty in tails and feed assays, the overall accuracy for Russian HEU is about 120 MT out of an estimate of 1250 MT.

By far the greatest portion of this uncertainty is due to the uncertainty in SWU production. Podvig uses an estimate of 5%, or 20 million Kg-SWU, out of an estimated cumulative production of 400 million SWU. This translates into an uncertainty of 100 MT, over 80% of the total 120 MT of uncertainty. Compared to this, other sources of error and uncertainty are minor. The allocation of the 400 million work units to HEU versus other products (LEU) is 136 ±9 million SWU, thus yielding 264 million ±22 million SWU applied to produce HEU. This corresponds to 110 MT of uncertainty under an assumption that these errors are uncorrelated.

Podvig attributes an error of ±5% to uncertainty in tails assays for uranium-235, which translates to 40 MT of production uncertainty. When added as an independent source of error, this increases the overall error from 110 to 120 MT.[32] Podvig mentions that his calculation assumes a feed assay of .667% (rather than natural feed at .71%), but appears to assign no uncertainty to this factor.

The implication for a nuclear archaeology strategy for HEU production is that even perfectly accurate uranium-235 assays of tails would gain only 7 to 10 MT of reduction in overall uncertainty, which corresponds to a reduction of about 6 to 8% in the current estimates.[33] A perfect estimate of the allocation of SWU between HEU and LEU would gain about 10 MT of uncertainty, but by far, the largest prospective gain would come from a better estimate of SWU production. For purposes of comparison, if nuclear archaeology methods could do for HEU what it can for plutonium (1% overall accuracy), this would give an error of just less than 10 million SWU for cumulative production, representing about 20 MT of HEU. Adding this to the 40 MT of error assumed for tails assay would give an overall error of 45 MT, which is very close to Podvig' s current (as yet unpublished) uncertainty estimate.

Assuming we could resolve tails uncertainty perfectly in addition to getting 1% accuracy for SWU production, the total error for Russian HEU production would be down to 20 MT, or less than 2% of production. This is an ambitious

target, but it still represents a large uncertainty when contrasted with the implied accuracy of the U.S. estimate.

A Measurements-Based HEU Estimate

The above analysis deals with the current uncertainty in terms of various sources of error. It defines potential measurement strategies or targets for a measurements-based archaeological regime; however, it says nothing about the prospective accuracy of such measurements-based methods.

In his original paper, Fetter discusses an estimation scheme for HEU production in which the ratio of uranium-234 to uranium-235 is measured in enrichment tails to indicate the product enrichment level. While he did not quantify the prospective accuracy of this technique, it is clear from the graph of this ratio as a function of product enrichment that very accurate tails characterization would be required to confirm product enrichment for HEU. While this is easily possible at the sample level, many samples would be needed to insure such accuracy held for the entire volume of tails produced over the life cycles of enrichment facilities. If this were possible, such an estimate could be combined with the mass of tails to estimate both the enrichment and mass of product.

A recent paper by Sharp[34] evaluates this scheme for simple cases. Sharp acknowledges that complex cases (such as the Russian enrichment complex) present many features which could make this method less useful. Since such an estimate could be very sensitive to details of the plant history, a rich statistical simulation model would be needed to assess its accuracy. Pending development of alternative methods for estimating HEU production, a study modeling the conditions that approximate the production conditions in both the United States and Russia would be very valuable.

Because further research and development on potential signatures for cumulative HEU production is in its very early stages,[35] it is not yet possible to quantify the accuracy of a production estimation method that exploits such signatures. This section explores the question briefly in a qualitative context.

In general, the problem of estimating cumulative production of HEU requires estimation of the cumulative production of SWUs in an enrichment facility. A SWU is a unit for the amount of separative work done in an enrichment process, and it is independent of the technological nature of the process. Thus gaseous diffusion, electromagnetic, centrifuge, and laser enrichment technologies must produce the same numbers of SWUs to yield a given quantity of HEU from a given input ("feed") of natural or other LEU material. However, the possible signatures and measurement strategies for these different technologies could be quite different. For all of these technologies, the rate of SWU production per unit time for a given design is generally well characterized from experiments conducted in developing and commercializing the technologies,

but often unpublished and considered commercially sensitive. Thus an estimate of the service histories of individual machines or barriers might provide a basis for an upper-bound estimate of production. Because there are many individual machines in a typical cascade, it may be possible to characterize *average* service history very accurately, if appropriate measurements can be found and exploited.

A more direct scheme would rely on the simple fact that uranium-238 and uranium-235 decay to thorium-230 and thorium-230, respectively, at a low but very well-known rate. For a given cascade configuration, the mass of uranium processed, and hence mass of decay products, are correlated with HEU output. These decay chains yield elements with chemical volatility significantly different from uranium, whose deposition on surfaces might be exploited to derive a cumulative production signature.

A scheme to exploit isotopic signatures in material deposited on enrichment plant components is now being experimentally tested at PNNL. The deposited material includes uranium isotopes and uranium decay daughters that are present in both concentrations and ratios that may be diagnostic for several aspects of plant history. While the presence of such signatures has long been recognized, they were considered too subtle for useful exploitation. The current methods under development at PNNL use ultra-trace-level spectroscopic analysis of components from production enrichment facilities, supplemented with samples produced under laboratory conditions, and modeling studies to translate the measured signatures into estimates of facility operational parameters. As outlined in Table 6, the signatures from the components may be useful in estimating the cumulative mass of material that was processed in the facility, enrichment level, number of enrichment campaigns, and time since last production campaign.

PNNL has completed proof-of-concept measurements for the averaged uranium isotopics and the depth profile in the corrosion layer. Initial results indicated the possibility of differentiating between uranium-235 enrichments with a high level of precision. Furthermore, it may be possible to associate isotopic content in the corrosion layer with a specific enrichment campaign based on depth, at least for a limited number of exposures. Figure 2 shows a depth profile of a sample sequentially exposed to two depleted uranium (UF_6) environments using Secondary Ion Mass Spectroscopy. This data show the feasibility of implementing a method for determining time-averaged enrichment of a centrifuge. To increase measurement confidence, PNNL is developing a method to determine the total time of exposure by measuring the thickness of the corrosion layer.

PNNL has also identified three additional signatures that have yet to be tested. These signatures rely on the ability to measure the uranium daughter products in multiple locations within a centrifuge using various techniques to determine cumulative throughput, time-averaged enrichment, and temporal

Table 6: PNNL's technical approach to evaluate potential signatures for verifying UF_6 production in centrifuge plants.

Potential signature	Verified information	Status of research
Averaged uranium isotopics in corrosion layer	Time-averaged enrichment of UF_6 (nonlinear corrosion rate)	Complete proof-of-principle
Depth profile of uranium isotopics in corrosion layer	Temporal variation in enrichment campaigns	Complete proof-of-principle
Corrosion layer thickness	Estimate exposure time to UF_6	In development
Cumulative number of decay products (protactinium and thorium atoms)	Cumulative mass throughput of centrifuge	Conceptual phase
Elemental ratio of decay products in rotor wall and corrosion layer	Time-averaged enrichment of UF_6	Conceptual phase
Depth profile of implanted decay products in rotor wall	Temporal variation in enrichment campaigns	Conceptual phase

variations in enrichment. Due to uranium's long half-life, surrogates would need to be exposed to uranium hexafluoride for a significant period of time to build up a measurable concentration of decay products. PNNL has identified potential surrogates to begin feasibility testing of these signatures.

Ultimately, the utility of these signatures for estimating cumulative uranium hexafluoride throughput may be limited by saturation of corrosion layers on plant components. PNNL will continue to assess the potential applicability and feasibility of these signatures using actual plant components.

Independently of the work on developing signatures in residual material on plant components, a formal statistical assessment of the tails characterization method proposed by Fetter should be conducted, for conditions reasonably approximating US, and if possible, Russian HEU production. This could be conducted as a cooperative study, and would involve little or no new measurements methods research.

The problem of estimating cumulative HEU production from measurements of plant components (thus in a way essentially independent of historical records) is harder than the same type of estimate for reactor-produced plutonium. It involves extrapolating a relatively subtle signature consisting of a small number of atoms. It may not be possible to develop a meaningful and accurate estimate based on measurements, but there is promise, and the research should allow an assessment of the feasibility of the methods within about a year.

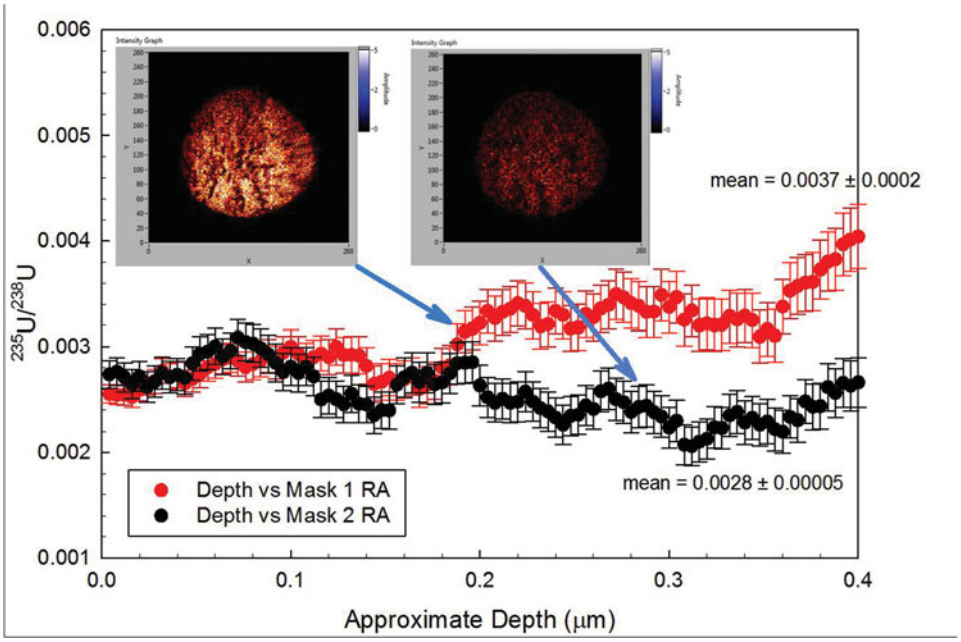

Figure 2: Isotopic analysis of UF_6 as a function of corrosion layer depth. This data shows a mixing layer of the two isotopic enrichments (black and red lines) which begin to separate into distinct layers with increasing depth which demonstrates the ability to differentiate between multiple enrichment campaigns. The micrographs represent the images derived from the secondary ion mass spectrometer measurements (color figure is available online).

SUMMARY AND FINDINGS

Significant technical progress has been made on nuclear archaeological methods since the concept was first advanced in the early 1990s. Several proof-of-principle experiments have demonstrated a technical basis for estimating cumulative plutonium production in the reactor types that account for the great majority of the world's WG-Pu stockpile. Several reports and papers were published on these methods during their development, and these documents are available in the open literature for independent scrutiny and verification. It is now feasible to plan and execute a program of nuclear archaeology that could reduce plutonium production uncertainty for both the United States and Russia. Such a program could serve as both a model for other countries and a first step toward confidence building in future bilateral or multilateral fissile material control treaties.

Even though technical progress on nuclear archaeological methods has promised more accurate estimates of plutonium production than we now have based on records alone, they have yet to be formally implemented. The extent of uncertainty characterizing the best record-based estimates of plutonium production is still substantial for all declared nuclear weapons states. The risk

posed by this uncertainty is that of weapons-useable material outside of formal control regimes. This risk becomes more important as nuclear stockpiles approach zero, or any other lower limit agreed on as a reasonable objective. Reduction of this risk requires implementation of archaeological measures in addition to their development.

The current technical state-of-the art for plutonium production estimation in graphite reactors is good enough to support a demonstration of the methods for an actual production reactor. While such an experiment is technically possible in any of the Hanford reactors, it would be much less costly if conducted prior to the encapsulation ("cocooning") of the reactor core. Currently, this opportunity exists at KE reactor for about another 12 months. PNNL is exploring such a demonstration with DOE, which could be conducted on a transparent bilateral or multilateral basis involving other interested countries.

Of current nuclear archaeological methods, only the isotopic ratio techniques for estimating plutonium production are mature enough for near-term application in a treaty-verification context. Even they may require additional cooperative development to assure treaty signatories that they present acceptable risks in verification use.

Similar methods that would allow accurate estimation for HEU production do not currently exist. This is due to several factors that include the difficult nature of the problem and the priority historically accorded to plutonium methods. Yet both the current uncertainty in informal estimates of HEU production and technical factors favoring HEU weapons over plutonium weapons by proliferant states and non-state actors, suggest that developing defensible methods for estimating HEU production should now become a priority. A useful first step in such a research agenda would be to clarify the best current estimates for Russian HEU production based on records and, thereby, improve upon Podvig' s recently published informal estimates.

The progress made in U.S.-Russian arms controls treaties has outstripped the formal agreements made to dispose of weapons-grade fissile materials. Even accounting for current fissile material disposition agreements, existing fissile material available for weapons far exceeds that needed for the current inventory. It is not clear if this decoupling of fissile materials and weapons can remain operative as weapons numbers decrease for the United States and Russia, but it is clear that this model is generally not valid for proliferant states, or for any non-state actors seeking nuclear capability. In these cases, fissile materials stockpiles could remain an active constraint on nuclear weapons capabilities.

The emphasis within the DOE nuclear archaeology program of developing methods that rely on measurements rather than records sought to maximize their value in verifying declarations. In today's political context, this same property may be even more important to allow production estimation in cases where no declarations are made.

A realistically long-term perspective on nuclear weapons reductions and fissile materials disposition suggests that the role of nuclear archaeology might be extended to estimation of use and disposition terms in certain cases, and that a focus on accurate estimation of *material remaining available for weapons use is* the correct objective for an archaeological program.

When archaeological capabilities are mapped against the existing and reasonably foreseeable policy landscape, several possible contributions emerge. In the context of a possible FMCT formulation, archaeological methods could supplement the existing separations-based definition of plutonium production to allow increased confidence that production was, in fact, cut off at the reactor stage. The principal roadblock to a broadened FMCT is the fissile material asymmetry between India and Pakistan. This could, at least partially, be addressed using nuclear archaeological methods.

Perhaps most importantly, nuclear archaeological methods including a suite of methods to allow estimation of HEU production, would enable a more complete fissile material control regime that addressed the extent and disposition of existing material stocks and their appropriate role in future weapons and weapons-limitation agreements.

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1. S. Fetter, "Nuclear Archaeology: Verifying Declarations of Fissile-Material Production," *Science & Global Security*, 3 (1993): 237–259.

2. B.D. Reid, D.C. Gerlach, P.G. Heasler, and J.V. Livingston, *Trawsfynydd Plutonium Estimate* (Richland, WA: Pacific Northwest National Laboratory, September 1997).

3. To the extent that estimates for several reactors were statistically independent, their errors would add by quadrature.

4. This paper was produced within the PNNL "Leadership Target" on *Quantification of Nuclear Facility Production*. The Nuclear Threat Initiative (NTI) has just launched a two-year "Verification Project" to explore the scope and value of possible verification technologies for both materials production and nuclear weapons. See also Matthew Sharp, "Applications and Limitations of Nuclear Archaeology in Uranium Enrichment Plants," *Science & Global Security* 21(2013): 70–92.

5. A. Diakov, "The History of Plutonium Production in Russia," *Science and Global Security* 19 (2011): 28–45.

6. James M. Acton, *Low Numbers A Practical Path to Deep Nuclear Reductions* (Washington, DC: Carnegie Endowment for International Peace, 2011) <http://carnegieendowment.org/files/low_numbers.pdf>.

7. Nuclear archaeology is defined as a set of methods for examining facilities to determine past nuclear material production or processing history, including type, quantity, timing, and other relevant attributes, seeking estimates of these attributes that are as independent as possible of operator declarations.

8. The information provided in the state-of-knowledge is based upon open-source, unclassified information only.

9. Reid et al., *Trawsfynydd Plutonium, op cit*.

10. J.P. McNeece, B.D. Reid, and T.W. Wood, *The Graphite Isotope Ratio Method (GIRM): A Plutonium Production Verification Tool*, PNNL-12095 (Richland, WA: Pacific Northwest National Laboratory, 1999).

11. In a blind experiment, PNNL predicted production of 3.63 Mt in a U.K. graphite moderated, gas-cooled reactor. Actual production as declared by the operator was 3.633 Mt.

12. While both sets of techniques rely on isotopic signatures, the HEU production signature also depends on chemical dynamics governing deposition of uranium-238 decay products in a complex and variable environment.

13. International Panel on Fissile Materials (http://fissilematerials.org/). Numbers for WG-Pu for the United States and United Kingdom are based on official data. Most numbers for reactor-grade plutonium are based on declarations submitted to IAEA. Other numbers are non-governmental estimates, often with large uncertainties. HEU amounts are 90% enriched HEU equivalent.

14. Ibid.

15. P. Podvig, "History of Highly Enriched Uranium Production in Russia," *Science & Global Security* 19 (2011): 46–67.

16. This is by no means an exhaustive account; no general Russian language review has been done.

17. H. Zhang, "China's HEU and Plutonium Production and Stocks," *Science & Global Security*, 19 (2011): 68–89.

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31. Ibid.

32. The authors calculate the overall error to be 117 MT.

33. This is based only of perfect information for uranium-235 and does not account for the informational value from uranium-234 characterization as proposed by Fetter, Nuclear Archaeology, 237–259, *op. cit.,* and exploited in the a recent paper by Sharp, Applications and Limitations of Nuclear Archaeology.

34. Sharp, "Applications and Limitations of Nuclear Archaeology in Uranium Enrichment Plants," 70–92, *op. cit*.

35. Work on this problem was deliberately delayed in favor of rapid development of methods for estimating plutonium production in the 1990s. Methods for HEU production estimation are being pursued at the scoping level in Laboratory-funded projects at PNNL.