

# Analysis of Fission Products— A Method for Verification of a CTBT During On-Site Inspections

Li, Bin<sup>a</sup>

---

If under a Comprehensive Test Ban Treaty a suspicious event can be accurately located, it should be possible to sample possible fission products through an on-site inspection. Such sampling, focused on analysis of the relative abundance of a few key isotopes, can be used to determine the time of a nuclear explosion to an uncertainty of a few hours or less even if the measurements are made several weeks after the event. The sampling could also reliably distinguish nuclear explosions from a deposit of nuclear waste.

## INTRODUCTION

The verification of a Comprehensive Test Ban Treaty (CTBT) would be implemented in two layers: remote international monitoring and on-site inspections (OSIs). The international monitoring system (IMS) would consist of a seismic network, a hydroacoustic network, a network of radionuclide monitors and a network of infrasound detectors. The addition to the IMS of a network of monitoring satellites and a network of electromagnetic pulse (EMP) sensors has also been proposed.<sup>1</sup> The IMS is designed to detect signals created by nuclear explosions which can travel long distances. Such a system may not be able to identify some events if the signals received by IMS are too weak. Further inspections closer to the events may therefore be needed to clarify the uncertainties.

Potential violators of the CTBT would prefer underground nuclear tests because such tests are harder to detect than atmospheric or underwater tests. If a suspicious underground event could be accurately located, samples of possible explosion products could be obtained by drilling at the site of the event. The existence of fission products could provide conclusive evidence of a nuclear explosion. However, the agreed four net-

---

<sup>a</sup> Beijing Institute of Applied Physics and Computational Mathematics. Work was completed as SSRC-MacArthur Foundation Peace and Security Fellow, Princeton University.

works of the IMS could only locate a suspicious event with an uncertainty of hundreds of square kilometers.<sup>2</sup> This area would need to be reduced by some on-site inspection methods.

In this paper, we assume that the positions of suspicious events can be accurately located, and therefore, the sampling of fission products is possible. We then focus on the analysis of fission products that might be present in samples obtained by drilling at the suspicious site. This paper provides a method to determine the time of nuclear explosions by measuring the relative abundance of isotopes in the samples. Based on the presumptive estimate of the explosion time, the proposed CTBT organization could judge whether or not there has been a violation of the CTBT. This method could also help distinguish a nuclear explosion from a deposit site of waste from nuclear reactors.

## THE PHYSICS OF UNDERGROUND NUCLEAR EXPLOSIONS

The fission core of a nuclear device is usually made up of highly-enriched uranium (U) 235 or plutonium (Pu) 239. Sometimes deuterium and tritium are added to the core to boost the yield. The neutrons generated by deuterium-tritium fusion have a higher energy—about 14 MeV—than most of those generated in fission. The fusion neutrons can even induce fission in U-238, which is also used in some nuclear devices. In a nuclear explosion, about 80 generations of fission take place in one microsecond, so all the fissions can be considered to occur at the same time if the fission products are measured weeks later. On the other hand, the fission in a nuclear reactor usually occurs over months or years of the operation of the reactor. The fission products initially consist of more than 300 nuclides of about 40 elements ranging from nickel to erbium. Almost all of them are radioactive and decay in various ways.

After a nuclear device is detonated underground, the huge amount of energy released vaporizes the hardware and adjoining rock to form a spherical cavity. The shock wave crushes and fractures the surrounding rock to a distance several times the radius of the cavity. As the cavity cools down, the mixture of the molten rock and weapon debris material solidifies at the bottom of the cavity. The solidus temperatures of most rocks are between 1,000°C and 1,200°C.<sup>3</sup> Some gaseous elements including noble gases (krypton and xenon) and elements with low boiling points (e.g., iodine, strontium and cesium) could remain outside of the molten rock. As the temperature and pressure in the cavity decrease, the roof of the cavity begins to fall in. The collapse of the overlying rock forms a rubble-filled chimney. An evasive test should avoid cratering and venting on the surface of the ground.<sup>4</sup>

The isotopes in the fission products are not uniformly mixed after an underground explosion. Almost all elements with high boiling points in the fission products are incorporated in the vitrified rock. The volatile elements and

**Table 1:** Initial percentages of some isotopes in the fission products from U-235 radiated by fission neutrons.**Non-volatile Isotopes**

Isotope	Zr-95	Mo-99	Ru-103	Ce-141	Ce-144	Nd-147
Yield <sup>a</sup>	6.299	6.015	3.650	5.718	4.872	2.168
Error	.3236	.3377	.5500	.2537	.2350	.1037
Half life <sup>b</sup>	64.0d	66.02h	39.4d	32.5d	284d	11.0d
Boiling point (°C)	4,377	4,612	3,900	3,426	3,426	3,068

**Volatile Isotopes**

Isotope	Kr-89	Sr-89	Xe-133m	Xe-133	Xe-135	Xe-137	Cs-137
Yield	4.330	4.507	.1840	6.630	6.300	5.650	6.360
Error	.2400	.3203	.0280	.9900	.9500	.3200	.6400
Half life	3.18m	50.5d	2.19d	5.25d	9.01h	3.82m	30.17y

a. The data of yields and measurement errors are taken from Wang Dao et al. eds., "Fission-Product Yield," Nuclear Data Center of Chinese Second Machine Production Ministry, Beijing, China, (February 1981).

b. The data of half lives and boiling points are taken from Lu Xiting et al. eds., "Handbook of Nuclear Data," Atomic Energy Publishing Co., Beijing, China, (1981).

those that have volatile precursors could be dispersed in the whole chimney and therefore have relatively low abundances in the vitrified rock. The noble gases and their daughter elements could even escape from the chimney through the fractures and faults in the surrounding rock.

## TIME-DEPENDENCE OF CONTENTS OF FISSION PRODUCTS

Most isotopes in the fission products have extra neutrons, so they tend to decay to more stable isotopes through beta emission accompanied by gamma emission. Some isotopes in their metastable states could either decay to other isotopes through beta emission or release their extra energy through internal transitions. Neither of these types of decays change the mass distribution of the fission products. Typically, the mass distribution curve of the fission products has a camel-type curve: two peaks and a valley at mass number of about 115. Table 1 lists some isotopes in the fission products of U-235 that are near the two peaks. The initial yields of isotopes near the two peaks do not depend much on the materials fissioned (U-235, Pu-239 or U-238) or on the energy of the neutrons that caused the fission (see table 2). The yields of the isotopes at the valley, however, depend sharply on these variables. The decay chains of some isotopes are given in figure 1.

**Table 2:** Initial percentages of some isotopes in the fission products of U-235, Pu-239 and U-238 by fission neutrons and 14-MeV neutrons.<sup>a</sup>

	Fission neutrons			14-MeV neutrons		
	U-235	Pu-239	U-238	U-235	Pu-239	U-238
Zr-95	6.299	4.669	5.124	5.164	-	4.994
Mo-99	6.015	5.912	6.780	5.067	-	5.601
Ru-103	3.650	6.434	4.995	3.641	-	4.565
Ag-111	.03098	.4191	.07853	1.207	2.073	1.130
Cd-115	.02371	.08216	.03955	0.8854	-	.6594
Ce-141	5.718	4.570	5.850	4.660	-	4.172
Ce-144	4.872	3.482	4.593	3.676	2.870	3.997
Nd-147	2.168	2.023	2.604	1.751	1.890	1.998

a. The data are taken from Wang Dao, "Fission Product Yield." The data unavailable in this book are marked by "-" in the table.

The content of an isotope at time  $t$  can be described by a differential equation:

$$\frac{dC_n}{dt} = -\lambda_n C_n + \lambda_{n-1} C_{n-1} \quad (1)$$

where  $n$  represents the  $n$ th isotope in a decay chain,  $n-1$  represents its precursor;  $C_n$  and  $C_{n-1}$  are their contents at time  $t$ ;  $\lambda_n$  and  $\lambda_{n-1}$  are the decay constants, which are  $\ln(2)$  divided by their half lives. This equation describes the situation in which a nuclide has only one decay branch. Some metastable isotopes have two decay branches: beta decay and internal transition. In this case, two more terms are added to the right side of equation (1).

By solving differential equations like equation (1) for isotopes in a decay chain, we can get the content of any isotope at any time:

$$C_n(t) = A_{n1} \exp(-\lambda_1 t) + A_{n2} \exp(-\lambda_2 t) + \dots + A_{nn} \exp(-\lambda_n t) \quad (2)$$

where the coefficients  $A_{n1}, A_{n2}, \dots, A_{nn}$  are determined by the initial contents of the isotopes in the decay chain  $C_1(0), C_2(0), \dots, C_n(0)$  and their decay constants  $\lambda_1, \lambda_2, \dots, \lambda_n$ .

Decay:		beta		beta		beta	
Isotope	Kr-89	→	Rb-89	→	Sr-89	→	Y-89
Half life	3.18m		15.2m		50.5d		stable
Decay:		beta		beta			
Isotope	Zr-95	→	Nb-95	→	Mo-95		
Half life	64.0d		35.0d		stable		
Decay:		beta		beta			
Isotope	Mo-99	→	Tc-99	→	Ru-99		
Half life	66.02h		$2.14 \times 10^5$ y		stable		
Decay:		IT		beta			
Isotope	Xe-133m	→	Xe-133	→	Cs-133		
Half life	2.19d		5.25d		stable		
Decay:		IT (99%+)		beta		beta	
Isotope	Xe-135m	→	Xe-135	→	Cs-135	→	Ba-135
Half life	15.6m		9.01h		$3 \times 10^6$ y		stable
Decay:		beta		beta			
Isotope	Xe-137	→	Cs-137	→	Ba-137		
Half life	3.82m		30.17y		stable		
Decay:		beta		beta			
Isotope	Ce-144	→	Pr-144	→	Nd-144		
Half life	284d		17.3 m		nearly stable		
Decay:		beta		beta			
Isotope	Nd-147	→	Pm-147	→	Sm-147		
Half life	11.0d		2.623y		nearly stable		

Figure 1: Some decay chains in the fission products.

For the first two isotopes in a decay chain, the contents are given by:

$$C_1(t) = C_1(0) \exp(-\lambda_1 t)$$

$$C_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} C_1(0) \exp(-\lambda_1 t) + \frac{C_2(0)\lambda_2 - C_2(0)\lambda_1 - C_1(0)\lambda_1}{\lambda_2 - \lambda_1} \exp(-\lambda_2 t)$$

(3)

If we know the initial contents of the isotopes and their decay constants in a decay chain, we can calculate their contents at any time. This fact is used later in the paper to make estimates of fission time from later measurements.

**Table 3:** Radioactive emissions of some fission products.<sup>a</sup>

Isotope	Gamma radiation energy in MeV <sup>b</sup> (emission probability) <sup>c</sup>	Beta radiation energy in MeV <sup>d</sup> (emission probability)
Zr-95	0.724 (45%), 0.757 (55%)	0.396 (55%), 0.360 (43%)
Nb-95	0.766 (100%)	0.160 (100%)
Mo-99	0.739 (12.6%), 0.140 (88.7%), 0.181 (6.29%)	1.21 (84%), 0.450 (14%)
Tc-99	0.0987	0.292
Ru-103	0.497 (86.4%)	0.225 (91%), 0.117 (5.3%)
Ce-141	0.145	0.582 (30%), 0.444 (70%)
Ce-144	2.186 (7%), 1.489 (3%), 0.696 (1.3%), 0.134 (11.1%)	0.316 (767%), 0.160 (20%)
Nd-147	0.531 (12%), 0.319 (2%), 0.0911 (27.2%)	0.810 (83%), 0.369 (15%)
Pm-147	0.1218	0.2245
Sr-89	0.908	1.448
Cs-137	Emission rate too low to detect	0.2;
Cs-137	0.0322	0.5116 (94%), 1.176 (6%)

- a. The data are taken from Lu Xinting, "Handbook of Nuclear Data," and OTA, "Environmental Monitoring for Nuclear Safeguards," OTA-BP-ISS-168, (September 1995), p. 16.
- b. The gamma radiation energy in this table refers to the central position of a peak in the gamma energy spectrum.
- c. The emission probability indicates the average probability of the emission for the decay of one nucleus. Some nuclei can emit two gamma rays. In this case, the sum of the emission probabilities of all gamma decay branches could be greater than one.
- d. The beta radiation refers to the maximum energy of a beta decay branch.

A radionuclide usually has one or more peaks in its gamma ray energy spectrum, with characteristic positions and intensities. The combined gamma spectrum of a mixture of the fission products can have many peaks, whose positions indicate the existence of the isotopes and whose intensities indicate their relative amounts.

Every branch of beta decay has a continuous spectral energy distribution with a maximum energy cutoff. The spectra of the beta decays of most isotopes in the fission products are well known. Therefore, it is also possible to estimate the relative amounts of isotopes in the fission products by measuring the combined beta spectrum. The parameters of characteristic gamma and beta decays of some isotopes are listed in table 3.

After we get the relative amounts of two radionuclides in a sample by measuring the intensities of their characteristic radiation, we can then derive the time of the fission from equations (2) or (3).

## EVENT IDENTIFICATION AND TIMING

Samples taken from a suspicious site can be analyzed in various ways. The first thing to do is to examine the physical features of the samples. The high temperature of an underground nuclear explosion will form vitrified rock at the bottom of the cavity. If there has been a nuclear explosion, we should be able to find vitrified rock by drilling. A repository for waste from nuclear reactors would not have such a feature. This could be used as a criterion to identify a nuclear explosion.

The second step would be to measure the gamma and beta radiation from the samples. A high radioactivity would strongly suggest a nuclear fission source.

The third step would be to estimate the abundances of the isotopes in the samples by measuring the intensities of the characteristic radiation. Based on the principles described earlier, we could also determine the time when the fission occurs if the analyzed isotopes are uniformly mixed. As mentioned, some noble gases, volatile elements and their daughter elements will have been depleted in the vitrified rock. We should avoid using such elements to estimate the fission time.

We therefore have several criteria for choosing isotopes with which to estimate the time of the fission. The first criterion is to choose elements with high boiling points to make sure that most of them are incorporated into the vitrified rock. The second criterion is that the chosen isotopes should have high yields in the fission products. The third criterion is that they should not be natural isotopes, which could not be distinguished from existing isotopes in the environment. The fourth criterion is to choose isotopes with appropriate half lives. The on-site inspection teams could arrive within days to weeks of a suspicious event detected by the IMS. We should have some isotopes with half lives of days to weeks with which to measure the time. We also need some isotopes with half lives of years or even longer. Their contents do not change much from days to weeks, so they could be used to calibrate the measurement. The isotopes in group one of table 1 (Zr-95, Mo-99, Ru-103, Ce-141, Ce-144 and Nd-147) and some of their daughter isotopes (Nb-95, Tc-99, Pm-147) meet the above standards, so these isotopes should be the focus of the measurements.<sup>5</sup>

The measurement of the gamma and beta radiation from the samples could be done at the inspection site. If the measurement accuracy is insufficient, the samples could be taken to laboratories for measurement by mass spectrometers, which could give much higher accuracy. Some isotopes (e.g., Cs-135) have very low emission rate for detection. Spectrometers therefore provide a good way to measure their abundances.

The determination of the time of the fission has the following uses for CTBT verification.

## Discrimination of Nuclear Explosions

The fission in a reactor lasts a long time—from months to years. Isotopes with half lives shorter than weeks would have a reduced presence in the waste. So, if the abundances of the isotopes with short lives in the samples are less than those expected in nuclear explosions, the drilling sample could be from a deposit of reactor waste. In contrast, the fission in a nuclear explosion occurs almost at the same time and the time of the fission could be calculated by measuring ratios of abundances of various pairs of isotopes. Each ratio could give an estimate of the time of the fission. If all the estimates of the fission time as well as the time of a suspicious event detected by the IMS are coincident with each other, the event could be confirmed as a nuclear explosion.

## Violation Judgment

The sites of underground nuclear explosions conducted before the entry into force of the CTBT could also be found by the inspection team. The determination of the time of the explosion could help the CTBT organization judge whether or not it was a violation. To avoid too much classified information being revealed, the measured energy intervals of beta and gamma radiation could be limited to several “windows,” transparent to only the characteristic peaks of specified isotopes.

## ACCURACY IN DETERMINATION OF FISSION TIME

The error in the measurement of the intensities of gamma peaks depends on the level of the background and the total counts of the gamma emissions. We can estimate the errors under some assumptions.

Assume: (1) that the yield of the explosion is 1 kiloton, which means 57 grams of U-235 were fissioned in the explosion; (2) a 10-gram sample is taken from 1,000 tons of vitrified rock containing radionuclides; and (3) the detection efficiency is 10 percent. Then the count rate of gamma emissions from a specified decay of an isotope is:

$$\begin{aligned}
 D &= \frac{57}{235} \times 6.023 \times 10^{23} \times \frac{10}{10^9} \times 10\% C(t) \lambda P \\
 &= 1.46 \times 10^{14} C(t) \lambda P
 \end{aligned}
 \tag{4}$$

where  $C(t)$  is the abundance of the isotope at time  $t$ , given by equations (2) or (3);  $\lambda$  is its decay constant,  $P$  is the emission probability of the given radiation branch. The calculated count rates of 0.14-MeV gamma of Mo-99



**Table 4:** Uncertainties in determination of fission time by measuring the gamma radiation of Mo-99 and Tc-99.

$t$ (days) <sup>a</sup>	1	7	14	21	28	70
$C_1(t)$	$4.68 \times 10^{-2}$	$1.03 \times 10^{-2}$	$1.77 \times 10^{-3}$	$3.03 \times 10^{-4}$	$5.19 \times 10^{-5}$	$1.32 \times 10^{-9}$
$C_2(t)$	$1.34 \times 10^{-2}$	$4.98 \times 10^{-2}$	$5.84 \times 10^{-2}$	$5.98 \times 10^{-2}$	$6.01 \times 10^{-2}$	$6.01 \times 10^{-2}$
$D_1$ (1/hour)	$6.36 \times 10^{10}$	$1.40 \times 10^{10}$	$2.4 \times 10^9$	$4.12 \times 10^8$	$7.06 \times 10^7$	1,789
$D_2$ (1/hour)	723	2,690	3,152	3,231	3,244	3,246
$\Delta C_1(t)/C_1(t)$	$4.00 \times 10^{-5}$	$8.44 \times 10^{-5}$	$2.04 \times 10^{-4}$	$4.93 \times 10^{-4}$	$1.19 \times 10^{-3}$	0.236
$\Delta C_2(t)/C_2(t)$	0.372	0.193	0.178	0.176	0.176	0.176
$\Delta t$ (hours)	7.89	15.22	16.49	16.72	16.82	39.23

- a. Note that  $t$  is the time between the nuclear fission and the measurement of the gamma radiation;  $C_1(t)$  is the content of Mo-99 at time  $t$ ;  $C_2(t)$  is the content of Tc-99 at time  $t$ ;  $D_1$  is the count rate of 0.14-MeV gamma emissions of Mo-99 at time  $t$ ;  $D_2$  is the count rate of 0.0987-MeV gamma emissions of Tc-99 at time  $t$ ;  $\Delta C_1(t)/C_1(t)$  is the uncertainty in determination of the content of Mo-99 at time  $t$ ;  $\Delta C_2(t)/C_2(t)$  is the uncertainty in determination of the content of Tc-99 at time  $t$ ;  $\Delta t$  is the uncertainty in determination of the fission time by measuring the contents of Mo-99 and Tc-99. In the calculation, the duration of the measurement is assumed to be one hour.

and 0.0987-MeV gamma of Tc-99 at different times are given in table 4. All needed parameters are listed in table 1, figure 1 and table 3.

If the background level  $B$  is 50 times the height of the pure gamma peak,<sup>6</sup> the measurement accuracy of the gamma ray intensity is:

$$\frac{\Delta C(t)}{C(t)} = \frac{\sqrt{2Bd}}{Dd} = \frac{\sqrt{2 \times 50Dd}}{Dd} = \frac{10}{(Dd)^{1/2}} \quad (5)$$

where  $d$  is the duration of each measurement of gamma radiation. We assume that each measurement takes 1 hour ( $d = 1$  hour). The calculated measurement accuracies for Mo-99 and Tc-99 at different times are given in table 4.

The abundances of the isotopes in the samples of fission products could also be measured using mass spectrometers. These measurements require more time and money than radioactivity measurements, but can achieve very high accuracy.

When we calculate the time of fission, we need to measure the relative abundances of at least a pair of isotopes in the samples. The measured pair of isotopes could be in the same decay chain. For example, we can measure the abundances of Zr-95 and its daughter Nb-95, Mo-99 and its daughter Tc-99, and Nd-147 and its daughter Pm-147. The initial abundances of the three daughter isotopes (Nb-95, Mo-99 and Pm-147) are zero, so we can simplify equation (3) as follows:

$$C_1(t) = C_1(0) \exp(-\lambda_1 t)$$

$$C_2(t) = \frac{\lambda_1}{\lambda_1 - \lambda_2} C_1(0) [\exp(-\lambda_2 t) - \exp(-\lambda_1 t)]$$

By solving this equation, we get:

$$t = \frac{1}{\lambda_1 - \lambda_2} \ln \left[ 1 + \frac{\lambda_1 - \lambda_2 C_2(t)}{\lambda_1 C_1(t)} \right]$$

The differential of  $t$  can be written:

$$dt = \frac{1}{\lambda_1 - \lambda_2} \{ 1 - \exp [ (\lambda_2 - \lambda_1) t ] \} \left[ \frac{dC_2(t)}{C_2(t)} - \frac{dC_1(t)}{C_1(t)} \right]$$

A conservative estimate of the uncertainty in  $t$  is then:

$$\Delta t \leq \frac{1}{\lambda_1 - \lambda_2} \{ 1 - \exp [ (\lambda_2 - \lambda_1) t ] \} \left[ \frac{\Delta C_2(t)}{C_2(t)} + \frac{\Delta C_1(t)}{C_1(t)} \right] \quad (6)$$

where  $\Delta C_1(t)/C_1(t)$  and  $\Delta C_2(t)/C_2(t)$  are the relative measurement errors of the abundances, which are given by equation (5). In this case, the calculation uncertainty depends on the relative measurement errors of the abundances, measurement time and the decay constants.

For a pair of isotopes in different decay chains, the uncertainties in the initial abundances could also contribute some to the final results. If the two measured isotopes  $a$  and  $b$  are first ones in their decay chains, the ratio of their abundances is:

$$\frac{C_a(t)}{C_b(t)} = \frac{C_a(0)}{C_b(0)} \exp [ (\lambda_b - \lambda_a) t ]$$

so we can derive the time  $t$ :

$$t = \frac{1}{\lambda_a - \lambda_b} \ln \left[ \frac{C_a(0) C_b(t)}{C_b(0) C_a(t)} \right]$$

and its differential:

$$dt = \frac{1}{\lambda_a - \lambda_b} \left[ \frac{dC_a(0)}{C_a(0)} - \frac{dC_b(0)}{C_b(0)} + \frac{dC_a(t)}{C_a(t)} - \frac{dC_b(t)}{C_b(t)} \right]$$

**Table 5:** Uncertainties in determination of fission times.

$t$ (days) <sup>a</sup>	1	7	14	21	28	70
Measured isotopes	$\Delta t$ (hour)					
Zr-95/Nb-95	0.03	0.114	0.198	0.287	0.383	1.24
Mo-99/Tc-99	7.89	15.22	16.49	16.72	16.82	39.23
Nd-147/Pm-147	0.104	0.287	0.411	0.510	0.603	1.49
Mo-99/Nd-147	102	102	102	102	102	132
Ce-141/Ce-144	461	461	461	461	461	461

- a. Note that  $t$  is the time between the fission and the measurement of the radiation;  $\Delta t$  is the uncertainty in the determination of the time by measuring the gamma radiation. The branch ratio used in the calculation for each isotope is the biggest one among all of its gamma decay branches. The duration of measurement is assumed to be one hour.

so the uncertainty in time in this case can be estimated:

$$\Delta t \leq \frac{1}{|\lambda_a - \lambda_b|} \left[ \frac{\Delta C_a(0)}{C_a(0)} + \frac{\Delta C_b(0)}{C_b(0)} + \frac{\Delta C_a(t)}{C_a(t)} + \frac{\Delta C_b(t)}{C_b(t)} \right] \quad (7)$$

where  $\Delta C_a(t)/C_a(t)$  and  $\Delta C_b(t)/C_b(t)$  are the measurement errors of abundances given by equation 5,  $\Delta C_a(0)/C_a(0)$  and  $\Delta C_b(0)/C_b(0)$  are the relative uncertainties of the initial abundances.

There are two sources of uncertainty in the initial abundances. The tabulated initial abundances themselves have some errors, which are about 5 percent to 15 percent (see table 1). The second source of error is that we do not know the configuration of the fissile material and the energy of the incident neutrons, which could change the initial abundances of the isotopes in group one of table 1 by 10 percent to 25 percent (see table 2). So the maximum combined uncertainty of initial yields is 40 percent:

$$\frac{\Delta C(0)}{C(0)} = 40 \% \quad (8)$$

From equations (6) and (7), we can calculate the uncertainties in the determination of the fission time by measuring the intensities of gamma decays of fission the products. The needed parameters are listed in table 1, figure 1, and table 3. The obtained results for different cases are listed in tables 4 and 5.

The biggest uncertainty in table 5 is 461 hours (19 days). So for the purposes of CTBT verification, all uncertainties listed in table 5 are acceptable for determining the time of nuclear explosions. If we combine the results of different pairs of isotopes, we can get even smaller uncertainties.

## ATMOSPHERIC AND GROUND SURFACE SAMPLING IN ON-SITE INSPECTIONS

A global network of radionuclide detectors for the IMS has been suggested to monitor particulate radionuclides by the Radionuclide Expert Group to the Ad Hoc Committee on a Nuclear Test Ban Working Group on Verification. The global monitoring of noble gases is also being considered by some of the experts in the group.<sup>7</sup>

The noble gases and particulate radionuclides from nuclear explosions would have higher densities at the test site. Some noble gases could escape from the explosive chimney to the atmosphere through venting or through fractures and faults in the rock created by the explosion. Examples are Kr-89, Xe-133, Xe-133 m (metastable Xe-133), Xe-135 and Xe-137. The detection of these noble gases could help locate a event. Some of the daughter isotopes of these noble gases are also radioactive. For example, Sr-89, Cs-135 and Cs-137. They would remain near the test site because their precursors have short half lives and can not travel very far. The inspection team could trace the event by detecting Sr-89, Cs-135 and Cs-137.

Some neutron-activated isotopes and their daughter isotopes (e.g., C-14, Ar-37) could also have higher abundances on the test site. The detection of these isotopes could be considered as a method to locate an underground explosion.

## CONCLUSIONS

The analysis of fission products in the samples taken from drilling can reliably determine the time of nuclear explosions. It can also be used to distinguish nuclear explosions from a deposit of reactor waste. The detection of some radionuclides could help locate nuclear explosions.

For this method to work, there must first be accurate location of a suspicious event. This is a remaining challenge.

## ACKNOWLEDGMENTS

The author thanks Professor Du Xiangwan of China Academy of Engineering Physics, Professor Frank von Hippel, Dr. Hal Feiveson and Dr. Suzanne Jones for their comments and suggestions to improve this work.

## NOTES AND REFERENCES

1. "Comprehensive Test Ban Treaty: Now or Never—A Report of the 1995 Conference on Disarmament Negotiations," ACRONYM No. 8, (October 1995).
2. See the report by the Expert Group on Auxiliary Seismic Stations (December 8, 1995), Report of Infrasound Expert Group (December 15, 1995), Report of Hydroacoustic Expert Group (December 8, 1995), and Report of Radionuclide Expert Group (December 15, 1995). These expert groups are organized by the Ad Hoc Committee on a Nuclear Test Ban Working Group on Verification based in Geneva.
3. Toulouskian, Y.S., et al. edited, "Physical Properties of Rocks and Minerals," (McGraw-Hill Book Company 1981), p. 382.
4. The details of the process and effects of underground nuclear tests. See Jozef Goldblat and David Cox edited, "Nuclear Weapon Tests: Prohibition or Limitation?," (Oxford University Press 1988), pp. 75–91.
5. Molybdenum has volatile oxide  $\text{MoO}_3$  with boiling point of  $1,155^\circ\text{C}$ ; ruthenium has volatile oxide  $\text{RuO}_4$  with boiling point of  $40^\circ\text{C}$  (see David R. Lide et al. edited, "CRC Handbook of Chemistry and Physics," 76th Edition, (CRC Press 1995-1996), pp. 4–72 and pp. 4–81). According to R.A. Lorentz's calculation (see "Status of Valiation of the Corsor Computer Code Used for the Accident Source Term Reassessment Study" in T. S. Kress Compiled, "Review of the Status of Valiation of the Computer Codes Used in the Svere Accident Source Term Reassessment Study," OAK Ridge National Laboratory (1985), pp. 286 and 301), about  $2 \times 10^{-7}$  fraction of ruthenium releases to the air every minute at  $1,200^\circ\text{C}$ ; molybdenum has an even lower release rate at the same temperature. So, when we estimate their abundances in the explosion products, we can ignore the losses of ruthenium and molybdenum in the vitrified rock caused by the release of their volatile oxides.
6. According to the CTBT text, the solid samples would be taken by drilling several weeks after the conduct of a suspicious event. At this time, the solid isotopes that still have strong gamma emission must meet the following three requirements: (1) The initial yield of the decay chain is greater than 1 percent; (2) the half-life of the isotope or that of its parent isotope must be longer than several days so that the amount of the isotope has not decreased too much; (3) the half-life time of the isotope must be shorter than several years so that it has a big decay rate. About nine isotopes (Zr-95, Nb-95, Mo-99, Ru-103, Ce-141, Ce-144, Nd-147, Sr-89 and Pm-147) meet the above standards (see table 1 and figure 1). They have about 17 gamma spikes that have emission probabilities greater than 1 percent (see table 3). Their widths are greater than 1 keV, the energy resolution of germanium detector at energies of MeV. (See Robert Mozley and Oleg F. Prilutsky, "Radiation Detectors and Backgrounds," in Frank von Hippel and Ronald Z. Sagdeer, eds., *Reversing the Arms Race*, (Gordon & Breach Science Publishers, New York, 1990). So these 17 gamma spikes are distinguishable by germanium detectors. (See Fudan University edited, "Methods of Nuclear Experiments," p. 186.) Assuming that the intensities of the 17 spikes are comparable, the ratio of a peak to the combination of all these Compton platforms is about  $40/17 = 2.4$ . If we consider that the emission probabilities of some isotopes could be 100 times smaller than others (1 percent vs. 100 percent, see table 3), the ratio of a peak to the combined Compton background should be greater than 0.02.
7. See endnote 2.