OFF-SITE AIR SAMPLING ANALYSIS AND NORTH KOREAN NUCLEAR TEST

Hui Zhang

Kennedy School of Government
Harvard University
79 John F. Kennedy Street
Cambridge, MA 02138

ABSTRACT

While some people in the international community were skeptical about whether North Korea had actually tested a nuclear device on Oct.9, 2006, the U.S. Director of National Intelligence stated decisively on Oct.16, “Analysis of air samples collected on October 11, 2006 detected radioactive debris which confirms that North Korea conducted an underground nuclear explosion.” Moreover, some experts suggested that such air sampling analysis would be able to determine if the fissile material was plutonium or uranium. In this paper, I will explore what information could have been obtained from off-site air sampling analysis. Specifically, I will examine how to use the activity ratios of xenon isotopes to identify the North Korean nuclear test and whether the off-site air sampling analysis would be able to distinguish a test from a plutonium-bomb and a HEU-bomb.

Introduction

On October 9, 2006, North Korea conducted its nuclear test. While a number of seismic stations around the world detected this seismic activity, given the low yield of the test, many raised questions about whether the seismic signal was caused from a nuclear explosion or if a test had failed. The suspicion of whether North Korea had actually tested a nuclear was cleared up when United States Director of National Intelligence released on October 16, 2006 the statement that, “Analysis of air samples collected on October 11, 2006 detected radioactive debris which confirms that North Korea conducted an underground nuclear explosion in the vicinity of P’unggye on October 9, 2006. The explosion yield was less than a kiloton.” Moreover, on October 17, 2006, the New York Times reported that “American intelligence agencies have concluded that North Korea’s test explosion last week was powered by plutonium that North Korea harvested from its small nuclear reactor, according to officials who have reviewed the results of atmospheric sampling since the blast.” Some experts suggested that air sampling analysis would be able to determine if the fissile material was plutonium or uranium. In the followings, I explore what information could have been obtained from off-site air sampling analysis. In particular, I examine how to use the activity ratios of xenon isotopes to identify the North
Korean nuclear test and whether the off-site air sampling analysis would be able to distinguish a test from a plutonium-bomb and a HEU-bomb.

Detection of Radioactive Xenon

For an underground nuclear test, the noble gas fission products (e.g. krypton and xenon) are most likely vented above ground. Here we focus on the detection of radioactive xenon for off-site sampling analysis. The krypton isotopes leaked from North Korea’s small test would be hard to be detected, since the krypton isotope with shorter half-life (less than five hours) would be decayed out when the US collected the samples two days after the test, and the small amount of krypton-85 (with half-life of 11 years) would not be able to picked out from the already large background in the atmosphere contributed by the spent fuel reprocessing. In practice, the International Monitoring System (IMS) established by the Comprehensive Nuclear-Test-Ban Treaty has been mandated to establish a worldwide network of detector systems capable of detecting the four radioxenons: Xe-131m, Xe-133m, Xe-133, and Xe-135 (see table 1 for details). Since the yield of Xe-131m is much smaller than Xe-133, and Xe-135, Here we focus on Xe-133m, Xe-133, and Xe-135 for this North Korea’s test.

Table 1: Half-lives, fission yield, and MDC of xenon fission products

<table>
<thead>
<tr>
<th></th>
<th>Half-life</th>
<th>MDC (mBq/SAC)</th>
<th>Uranium-235 (fission yield %)</th>
<th>Plutonium-239 (fission yield %)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>independent</td>
<td>cumulative</td>
</tr>
<tr>
<td>Xe-131m</td>
<td>11.9d</td>
<td>10</td>
<td>2.4x10-7</td>
<td>0.045</td>
</tr>
<tr>
<td>Xe-133m</td>
<td>2.19d</td>
<td>4</td>
<td>0.0042</td>
<td>0.19</td>
</tr>
<tr>
<td>Xe-133</td>
<td>5.24d</td>
<td>0.5-1</td>
<td>0.0015</td>
<td>6.72</td>
</tr>
<tr>
<td>Xe-135</td>
<td>9.10h</td>
<td>0.5-1</td>
<td>0.12</td>
<td>6.6</td>
</tr>
</tbody>
</table>

Note: MDC--minimum detectable concentration. CTBT Preparatory Commission stipulates that the MDC of Xe-133 be less than 1 mBq per standard-cubic-meter of air (mBq/SCA) for a 24-hour sampling period. The fission yield data source: Lawrence Berkeley Laboratory, “Fission Product Yields,” [http://ie.lbl.gov/fission.html](http://ie.lbl.gov/fission.html) (fast-pooled neutron fission data).

An air sampling of xenon would be dependent on a number of factors including, e.g. how much xenon isotopes would be released into atmosphere (related to total inventory, leak rate, etc); meteorology conditions and dilutions (atmospheric dispersion), timely access to
the plume (e.g. decay factor and plume location), the background radionuclide concentration, and the minimum detectable concentration.

For a nuclear test with a yield around 1 kiloton (the most likely North Korean yield), the total activity of radioactive Xe-133m, Xe133 and Xe135 is estimated as shown in Figure 1. Furthermore, I estimate the concentration of xenon over the Sea of Japan at 48 hours after the test when the US collected the samples. Atmospheric dispersion model—HYSPLIT is used to simulate the xenon plume. Assuming that, 1) a release source at 41.294N, 129.094E (as the U.S. Geological Survey recorded) and at a height from ground level to 10m, 2) The release start time ranging from half an hour to five hours (Based on USGS provided local time--10: 35 am), and the release duration ranging from one hour to five hours respectively, 3) the leak rate of the xenon is of a few percent, then we estimate that the concentrations of xe-133 and xe-135 at 48 hours over Japan sea are 0.4–4 mBq/SCA (xe-133) and 1.5–15 mBq/SCA (xe-135) respectively. Both would be around or above their minimum detectable concentration (around 0.5-1 mBq/SAC). Thus they would be detectable. However, the concentrations of xe133m and xe131m could be too low for detection. In addition, based on the simulation of the xenon plume, it could be estimated that the plume was not yet transported over the Sea of Japan one day after the test. This could explain why the US airplane collected the useful samples until two days after the test.

![Fig.1: Activity of radioactive xenon from a one-kiloton plutonium or HEU explosion](image)
Fig. 2: Plume over the Sea of Japan at 48 hours after the North Korea’s test simulated by NOAA HYSPLIT MODEL. Concentration (at one unit) averaged between 0m and 1000m.

Identifying a Nuclear Explosion

Based on above analysis, the detectable fission products among the US samples taken two days after the test were most likely xe-133 and xe-135. Then, how can both be used to identify a nuclear explosion?

The detection of xe-133 or xe-135 alone could be difficult to identify a nuclear explosion, because both could also be leaked from South Korean or Japanese civilian nuclear reactors operations. However, we could use the activity ratio of xe-135 to xe133 to identify a nuclear test. The figure 3 shows the activity ratios of xe-135 to xe133 for four cases (plutonium-device explosion, HEU-device explosion, light water reactor (LWR) and fast breeder reactor (FBR)). As shown in the figure, the activity ratio of xe135 to xe133 from a nuclear explosion (either plutonium or HEU device) is much higher than that from a LWR or FBR. For example, xe135 to xe133 ratio from a nuclear explosion would be three times higher than that from the reactor leakage at 48 hours. Finally, we might need to make sure that there was no leakage from the nearby reactors since the explosion and if a leakage, there was no contribution to the samples. In addition, the activity ratio for the case of a
reprocessing plant would be zero, because $\text{xe}135$ would be decay out after lest several weeks cooling. In short, using the activity ratio of $\text{xe}-135$ to $\text{xe}133$ would be able to identify a nuclear test.

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**Fig. 3:** The activity ratio of $\text{xe}-135$ to $\text{xe}-133$ for four cases: pu-bomb, HEU-bomb, LWR, and FBR
Can Off-site Air Sampling Determine the Fissile Material Used in the North Korean Test?

One day after the U.S. office of the Director of National Intelligence confirmed North Korea indeed conducted a nuclear test, the report of *New York Times* further indicated the atmospheric sampling could determine the test was powered by plutonium. Some nuclear experts suggested so as well. As Harold P. Smith wrote, “Any nuclear explosion creates radioactive noble gases, notably xenon and krypton, that do not combine with other elements in the geologic structure. Therefore, they can more easily leak to the surface and into the atmosphere where they can be detected beyond national boundaries. Because at least two different gases escape, it is possible for radio-chemists to determine if the fissile material was plutonium or uranium, which of course is exactly what happened. (Press reports said the material was plutonium.”

Indeed, we believe the North Korea tested a plutonium device, given the fact that North Korea already had enough separated plutonium for at least several weapons, and it would be years away from producing enough HEU for one or two bombs if it had a dedicated HEU program. However, we are skeptical about the statement that measurements of radioactive noble gases alone can determine the fissile material used in the North Korean test, particularly if detected as much as two days after a test.

In order to examine whether the off-site air sampling analysis would be able to distinguish a test from plutonium-bomb and HEU-bomb, here I consider the relative changes of ratio of $\text{xe-135}$ to $\text{xe-133}$ for plutonium- and HEU-explosion cases, i.e. $R_{\text{Pu}}^{\text{HEU}}(\text{xe135}/\text{xe133}) - R_{\text{HEU}}^{\text{HEU}}(\text{xe135}/\text{xe133})/(R_{\text{HEU}}^{\text{HEU}}(\text{xe135}/\text{xe133})$ (where $R_{\text{Pu}}^{\text{Pu}}(\text{xe135}/\text{xe133})$ is the ratio of $\text{xe-135}$ to $\text{xe-133}$ for plutonium explosion case; $R_{\text{HEU}}^{\text{HEU}}(\text{xe135}/\text{xe133})$ is the ratio of $\text{xe-135}$ to $\text{xe-133}$ for HEU explosion case) (see Figure 4). As shown in figure 4, the relative changes of ratio of $\text{xe-135}$ to $\text{xe-133}$ within a few hours (i.e. the sampling time) would be of a few tens percentage, which would be distinguishable in the practical measurement. However, the relative changes of ratio of $\text{xe-135}$ to $\text{xe-133}$ at 48 hours sampling time (when US acquired the samples) was dropped quickly to about 0.1 %. Meanwhile, at the best measurement conditions, it is estimated that the relative measurement error of the ratio of $\text{xe135}$ to $\text{xe-133}$ at 48 hours sampling time would be at least over 10%, which would be least 100 times higher than that of the relative changes of ratio of $\text{xe-135}$ to $\text{xe-133}$ at 48 hours sampling time. Consequently, the use of the ratio of $\text{xe1335}$ to $\text{xe-133}$ would not be able to separate the Pu-bomb from HEU bomb.

In addition, for the case with significant “fractionation” effect, i.e. the xenon isotopes are separated from their parent iodine isotopes, also it can be concluded that the ratio ($\text{xe1335}$ / $\text{xe-133}$) would not be able to used to separate the Pu-bomb from a HEU bomb. Indeed, it would be difficult to exclude the “fractionation” effect due to different releases of some of the xenon precursors (iodine) because of their different condensation temperatures, solubilities in water and glassified rock, etc. In short, measurements of radioactive noble gases alone would be not able to determine the fissile material used in the North Korean test, particularly if detected as much as two days after a test.
However, it should be noted that if the unfissioned materials (e.g. plutonium debris) are vented and collected, it would be easy to determine whether the explosion was from plutonium or HEU device. However, unlike the noble gases, the unfissioned materials would be hard to escape into the atmosphere under a normal underground test. Even some plutonium debris are vented, as for the North Korea’s case, it would be hard to be transported far away for off-site air sampling. Finally, beyond seismic approach for North Korea case, using off-site air sampling would be not able to estimate the yield.

![Graph](image)

**Fig.4: The relative changes of ratio of xe-135 to xe-133 for plutonium- and HEU-explosion cases** (i.e. $R_{Pu}^{He} (xe_{135}/xe_{133}) - R_{HEU}^{HEU} (xe_{135}/xe_{133})$)/$R_{HEU}^{HEU} (xe_{135}/xe_{133})$, where $R_{Pu}^{Pu} (xe_{135}/xe_{133})$ is the ratio of xe-135 to xe-133 for plutonium explosion case; $R_{HEU}^{HEU} (xe_{135}/xe_{133})$ is the ratio of xe-135 to xe-133 for HEU explosion case.

In conclusion, radioactive xe-133 and xe-135 were most likely detectable from the US samples taken two days after the test. Using the activity ratio of xe135 to xe133 would be able to identify a nuclear explosion. However, the sampling analysis of the radioactive noble gases would be not able to distinguishing a test from a Pu-bomb and a HEU-bomb.
References and Notes


8 Shanker and Sanger, “North Korean fuel identified as plutonium,” op.cit.

9 Smith, “Nuclear Forensics and the North Korean Test,” op.cit.


11 A detailed estimation could be found at, Hui Zhang, “The North Korean Nuclear Test and Nuclear Forensics,” (to be submitted).

12 More details could be found at, Hui Zhang, “The North Korean Nuclear Test and Nuclear Forensics,” (to be submitted).