Low-level radioxenon measurements for Comprehensive Nuclear-Test-Ban Treaty verification purposes

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Overview

- The Comprehensive Nuclear Test Ban Treaty – some history...
- Some basics of a nuclear explosion
- Underground nuclear explosion and noble gases
- The International Noble Gas Experiment (INGE)
- Conclusions
1939  (2 August) Albert Einstein sends „his“ famous letter to President Roosevelt

1942  (June) the secret “Manhattan Project” starts

1945  (16 July) first human detonated nuclear explosion: „Trinity“ Test (21kt, New Mexico, USA)

1945  (August) nuclear weapons destroy Hiroshima (15kt) and Nagasaki (20kt)

1946  (July) USA starts its nuclear weapon test programme in the Pacific (Bikini Atoll etc.)

1949  (August) first Soviet nuclear weapon test (Semipalatinsk, Kazakhstan)

1954  (February) Fallout from the 15 Megaton BRAVO-Test contaminates the Japanese Fishing boat „Lucky Dragon“
Some history (2)

1954  India (PM Nehru) proposes at the UN to forbid all nuclear explosions

1958 - 1962  Experts Conferences in Geneva (main problematic: how to monitor?)

1963  „Partial Test Ban Treaty“ open for signature (PTBT)

1976 - 1981  Trilateral negotiations (USA, USSR, UK)

1985  USSR announces a test moratorium

1994 - 1996  Multilateral negotiations in Geneva after “Cold War”


1997  (April) “Provisional Technical Secretariat“ in Vienna

2007  (May) 177 countries signed the CTBT, 138 ratified the Treaty, 34 of the “list of 44” after 2410 nuclear tests took place…

Italy signed on 24 September 1996 and ratified on 1 February 1999
The CTBT Verification System

Italy contributes with an Auxiliary Seismic Station in Enna, Sicily and a Radionuclide Laboratory.
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Some basics…

**Critical masses**

<table>
<thead>
<tr>
<th></th>
<th>Mass for bare sphere</th>
<th>Mass with reflectors</th>
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</thead>
<tbody>
<tr>
<td>U-235</td>
<td>50 kg</td>
<td>20 kg</td>
</tr>
<tr>
<td>Pu</td>
<td>20 kg</td>
<td>3 kg</td>
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</table>

**Fissile**

<table>
<thead>
<tr>
<th></th>
<th>Mass for bare sphere</th>
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<td>Pu</td>
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</tbody>
</table>

**Fertile**

<table>
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<tr>
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<th>Mass for bare sphere</th>
<th>Mass with reflectors</th>
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<tr>
<td>U-238</td>
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</tbody>
</table>

**Gun device**

- ~1000 m/s good enough for U-235

**Implosion**

- ~5000 m/s needed for Pu to avoid preinitiation

Pu-240 emits ~1000 neutrons per gram and second

**Little Boy**

**Fat Man**
Some basics…

- Conventional chemical explosive
- Sub-critical pieces of uranium-235 combined

**Gun-type assembly method**

- High-explosive lenses
- Plutonium core compressed

**Implosion assembly method**
A little bit of physics…

- During fission of uranium or plutonium in a nuclear reactor, thermal (slow) neutrons are used, whereas during a nuclear explosion the fission is induced by fast neutrons. The full fission sequence in a device is finished within a microsecond.

- There is little time for complex activation build-up in a nuclear explosion, whereas there is sufficient time for production of many activation products in a nuclear reactor.

- These differences produce different radionuclide abundances. Since a nuclear blast produces different radionuclide abundances, nuclide ratios may be used for source identification.
Fission yield curves

Fission yield in % for several nuclear explosion relevant nuclides
(Fission yield is a function of the fissioning nuclide and the incident neutron energy)
Cumulative fission yields

<table>
<thead>
<tr>
<th>Fission Product</th>
<th>Half-life</th>
<th>Time unit</th>
<th>$^{235}\text{U}_f$</th>
<th>$^{235}\text{U}_{\text{he}}$</th>
<th>$^{238}\text{U}_f$</th>
<th>$^{238}\text{U}_{\text{he}}$</th>
<th>$^{239}\text{Pu}_f$</th>
<th>$^{239}\text{Pu}_{\text{he}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{131m}\text{Xe}$</td>
<td>11.934 d</td>
<td>d</td>
<td>0.05</td>
<td>0.06</td>
<td>0.05</td>
<td>0.06</td>
<td>0.05</td>
<td>0.07</td>
</tr>
<tr>
<td>$^{133m}\text{Xe}$</td>
<td>2.19 d</td>
<td>d</td>
<td>0.19</td>
<td>0.29</td>
<td>0.19</td>
<td>0.18</td>
<td>0.24</td>
<td>0.42</td>
</tr>
<tr>
<td>$^{133}\text{Xe}$</td>
<td>5.243 d</td>
<td>d</td>
<td>6.72</td>
<td>5.53</td>
<td>6.76</td>
<td>6.02</td>
<td>6.97</td>
<td>4.86</td>
</tr>
<tr>
<td>$^{135}\text{Xe}$</td>
<td>9.14 h</td>
<td>h</td>
<td>6.6</td>
<td>5.67</td>
<td>6.97</td>
<td>5.84</td>
<td>7.54</td>
<td>6.18</td>
</tr>
</tbody>
</table>

- Cumulative fission yields in % for six fission modes relevant to nuclear explosions, induced by fission spectrum neutrons (f) and high energy neutrons (14.7 MeV) (he)

- $^{133}\text{Xe}$ has high production rates and a not too short half-life. Therefore this xenon isotope is the one most observed in environmental samples
Energy from a 1 kton nuclear explosion

- 1 kiloton (kton) nuclear explosion is equal to an explosion of 1000 tons of TNT, which equals $10^{12}$ calories = $4.2 \times 10^{12}$ Joules

- The average total energy released in fission of one uranium-235 or plutonium-239 atom is 200 MeV = $3.2 \times 10^{-11}$ J

- This energy takes the form of kinetic energy of the fission fragments, instantaneous gamma-ray energy, kinetic energy of fission neutrons, beta particles from fission products, gamma rays from fission products and neutrinos from fission products
Activity of $^{133}\text{Xe}$ released per kton

- ~180 MeV is immediately available as energy from each fission event which is equal to ~1.45 $10^{23}$ fissions per kton

- Activity $A = \lambda \times N(t) = \ln(2) \times \frac{N(t)}{T_{1/2}}$

  $A = \text{activity [in e.g. Bq]}$
  $\lambda = \text{decay constant}$
  $N(t) = \text{number of atoms}$
  $T_{1/2} = \text{half-life}$

- Depending on the fission material ($^{235}\text{U}$, $^{233}\text{U}$ or $^{239}\text{Pu}$) between $1.08 \times 10^{16} \text{ Bq}$ and $1.33 \times 10^{16} \text{ Bq}$ of $^{133}\text{Xe}$ will be created in a 1 kton nuclear explosion
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Nuclear tests in different environments

Atmospheric Nuclear Tests
- Infrasonic waves
- Radionuclides: particulates & gases
- Possible seismic/hydro-acoustic coupling
- Noise sources: natural and cultural background, meteors, volcanoes, weather, air/spacecraft

Underground Nuclear Tests
- Seismic waves
- Radionuclides: vented gases
- Possible hydro-acoustic/infra-sound coupling
- Noise sources: natural and cultural background, earthquakes, volcanoes, chemical explosions

Underwater Nuclear Tests
- Hydroacoustic waves
- Radionuclides: vented gases
- Possible seismic/infra-sound coupling
- Noise sources: natural and cultural background, earthquakes, volcanoes, chemical explosions, whales
Identification of Nuclear Explosions

A nuclear explosion creates two sources of radioactive material:

- **Fission products**: these are direct products from the nuclear reaction. If remotely measured, they can give information on the material used inside the nuclear device.

- **Activation products**: this is radiation from the surrounding material from the device, formed by the neutrons that were created in the nuclear device. If these are measured, they can give a good indication of the environment where the explosion took place.

  - Atmospheric, underwater, surface and near surface (**non-evasive**) explosions: can be identified via particulate monitoring.

  - Underground and underwater (**evasive**) explosions: no aerosols, but only noble gases are released in the atmosphere via cracks, diffusion etc.
Noble gases: the “smoking gun” of an underground nuclear explosion

Noble Gases:

- **Argon-37**: activation product. Decays with no photons

- **Krypton-85** \((t_{1/2} = 10.7 \text{ y})\): world-wide high background

- **Xenon**’s: four isotopes are suitable for CTBT verification \((^{131m}\text{Xe}, ^{133m}\text{Xe}, ^{133}\text{Xe}, \text{ and } ^{135}\text{Xe})\)

  - An underground nuclear test creates seismic waves, possibly hydro-acoustic /infra-sound coupling

  - Radionuclide monitoring provides the only direct method with the potential to establish whether a nuclear explosion has occurred

  - A contained underground nuclear explosion may emit radioactive noble gases into the atmosphere
Noble gases releasing scenarios after a test

• Unintentional release of radioactive matter to the atmosphere due to failure of the containment system (containment failure) (0 - 100 %)

• Early venting due to high pressure of the explosion and other dynamic effects (pushes gas through cracks and fissures in the bedrock) (~10 %)

• Venting due to opening of tunnels for measurements of the test or recovery of measurement material can happen days till weeks after the event (controlled tunnel purging)

• Drilling of holes etc. (operational releases)

• Sucking of gases (among others, radioxenon) from deposits in the walls of cracks and fissures by low pressure weather systems (late-time seeps) (~1 %)
Radioxenon signal from the event to IDC

EVENT

Atmospheric transport to IMS station (1-21 days)

Total: ~ 3 - 24 calendar days

< 24-hour collection

1-2-hour processing

23-hour nuclear measurement

Transmission via GCI

IDC
Sources of Radioactive Xenon

- **Nuclear Reactors**: mainly $^{133}\text{Xe}$
- **Fuel reprocessing plants**: mainly $^{131m}\text{Xe}$
- **Hospitals**: mainly $^{131m}\text{Xe}$ and $^{133}\text{Xe}$
- **Nuclear Explosions**: mainly $^{135}\text{Xe}$, $^{133m}\text{Xe}$ and $^{133}\text{Xe}$

Differentiate between Nuclear Explosions and Reactor Emissions:

Several approaches are under study – more work to be done!
Xenon ratios for source characterisation with four isotopes

The plot separates into two domains due to duration of the neutron interaction:
- few microseconds in an explosion
- continuous in a reactor

(Ref.: Martin Kalinowski)

Critical area:
_signal caused by medical isotope production_
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Objective of the International Monitoring System (IMS) Noble Gas Network:
At least 90% detection capability within 14 days after a nuclear explosion in the atmosphere, underwater or underground for a 1 kton nuclear explosion.

Nuclides of interest:
$^{131m}\text{Xe} \ (11.9 \ \text{d}), \ ^{133m}\text{Xe} \ (2.19 \ \text{d}), \ ^{133}\text{Xe} \ (5.24 \ \text{d}), \ ^{135}\text{Xe} \ (9.10 \ \text{h})$

Minimum detectable concentration: $< 1 \ \text{mBq/m}^3$ for $^{133}\text{Xe}$
The International Noble Gas Experiment

» 4 different systems
  • **SPALAX** (France): high-resolution gamma spectra
  • **ARSA** (USA): two-dim. beta-gamma coincidence spectra
  • **SAUNA** (Sweden): two-dim. beta-gamma coincidence spectra
  • **ARIX** (Russian Fed.): two-dim. beta-gamma coincidence spectra (originally: beta-gated gamma spectra)

» Different Project Phases:
  • **I** at developers' sites
  • **II** all systems in Freiburg, Germany (2000 - 01)
  • **IIIa** systems at IMS stations with developers (2001 - 03)
  • **IIIb** systems at IMS stations with station operators (2003 - 04)
  • **IIIc** clusters of noble gas stations (2004 - …)
The INGE Project systems

**ARIX** (Russian Fed.)
β-gated γ spectra

**ARSA** (USA)
two-dim. β-γ coincidence spectra

**SAUNA** (Sweden)
two-dim. β-γ coincidence spectra

**SPALAX** (France)
high-resolution γ spectra
Noble gas stations map (by mid 2007)
The Measuring Principle

- **Sampling** of air at “high” volume air flow (> 0.4 m³/h)
- **Cleaning** (removal of aerosols, water, Radon, CO₂, …)
- **Extraction** of Xenon gas from air with high efficiency (adsorption of Xenon onto charcoal followed by thermal desorption of Xenon)
- **Detection** and **calculation** of the isotopic composition
- **Measuring** the stable Xenon volume (e.g. Gas Chromatograph)
- Sample into **archive bottle**
Identifying nuclides (γ-spectra, I)

- Ge escape peak (71 keV)
- $^{133}$Xe (81 keV)
- $^{133}$Xe (160.1 keV)
- $^{131m}$Xe (163.9 keV)
- $^{133m}$Xe (233.2 keV)
- $^{135}$Xe (249.8 keV)

Spectrum from CAX05 collection stop 12 Sept. 2003
Identifying nuclides (γ-spectra, II)

$^{131m}\text{Xe}$ and/or $^{131m}\text{Xe}$ X-Rays
(29.8 keV)

$^{133}\text{Xe}$ X-Rays
(31.6 keV)

$^{133}\text{Xe}$
(81 keV)

$^{131m}\text{Xe}$
(163.9 keV)
Not present

$^{133m}\text{Xe}$
(233.2 keV)
Not present

$^{135}\text{Xe}$
(249.8 keV)
Not present

Spectrum from DEX33
collection stop 24 Aug. 2005
Sample Collection Stop 10 June 2002: \((0.77 \pm 0.11)\) mBq/m\(^3\) of \(^{133m}\)Xe and \((4.26 \pm 0.35)\) mBq/m\(^3\) of \(^{133}\)Xe
Environmental monitoring, e.g. Schauinsland mountain (Freiburg), Germany (DEX33)
Environmental monitoring, in very low background regions, e.g. Yellowknife, Canada (CAX16)

Evidence of $^{131m}$Xe and $^{133}$Xe presence in a remote region
Environmental monitoring, in very low background regions, e.g. Spitsbergen, Norway (NOX49)

Summed spectra, to get a better sensitivity
Finding the needle in the haystack...

- **No radioxenon expectation**: no radioxenon isotopes present (e.g. at Tahiti in the Southern Pacific)

- **Regular but low radioxenon background**: regular presence of $^{133}\text{Xe}$ and or $^{131}\text{mXe}$ at very low concentrations (less than 1 mBq/m$^3$) (e.g. on the Arctic station of Longyearbyen, Spitsbergen)

- **Regular radioxenon background of $^{133}\text{Xe}$** (~ 1 - 100 mB/m$^3$) and occasionally other isotopes at low level are seen (e.g. in the European station on the Schauinsland mountain, Germany)

- **High radioxenon background with many isotopes**: all isotopes are regularly present at different activity concentrations (up to few Bq/m$^3$) (e.g. the station of Ottawa, which is surrounded by nuclear power plants and a large radiopharmaceutical production facility)
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Conclusions

✔ Radioxenon measurements play a key role in the verification of the CTBT in discovering clandestine underground nuclear explosions;

✔ The new technologies and operational systems are emerging at good speed;

But, a lot of work ahead:

@Enable
✔ in building more stations;

@Enable
✔ improving the methods;

@Enable
✔ further understanding the signatures to properly interpret its sources;

...to make the world a safer place to live...
Some references


Some references


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• Saey P.R.J. Ultra-Low-Level Measurements of Argon, Krypton and Radioxenon for Treaty Verification Purposes, submitted to ESARDA Bulletin

• Saey P.R.J., M. Bean, A. Becker et al. Intercontinental transport of radioxenon from the Korean Peninsula to Yellowknife, Canada in late October 2006, submitted to Geophysical Research Letters
Grazie !

Questions ?