



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

Some history (1)



- (2 August) Albert Einstein sends "his" famous letter to President Roosevelt
- (June) the secret "Manhattan Project" starts
- (16 July) first human detonated nuclear explosion: "Trinity" Test (21kt, New Mexico, USA)



- (August) nuclear weapons destroy Hiroshima (15kt) and Nagasaki (20kt)
- (July) USA starts its nuclear weapon test programme in the Pacific (Bikini Atoll etc.)
- (August) first Soviet nuclear weapon test (Semipalatinsk, Kazakhstan)
- (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"
- **1996** (24 September) "Comprehensive Nuclear Test Ban Treaty" opens for signature in New York
- **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

50 kg for bare sphere and 20 kg with reflectors **U-235** 20 kg for bare sphere and 3 kg with reflectors

Fissile Fertile

Pu

U-235, Pu-239, U-233 U-238 U-238-> U-239-> Np-239-> Pu-239, Th-232 Th-232-> Th-233-> Pa-233-> U-233

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



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

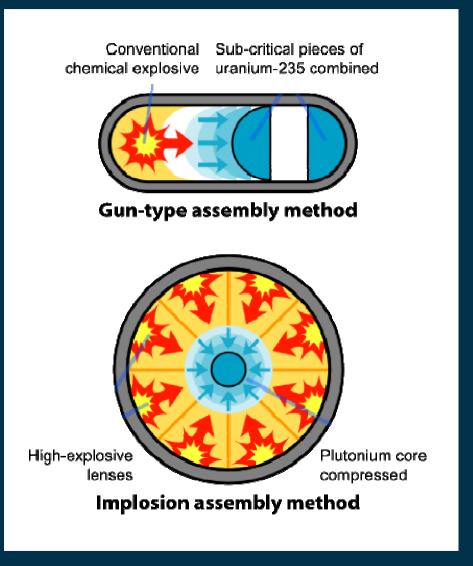
Pu-240 emits ~1000 neutrons per gram and second

Little Boy

Fat Man

Some basics...





A little bit of physics...

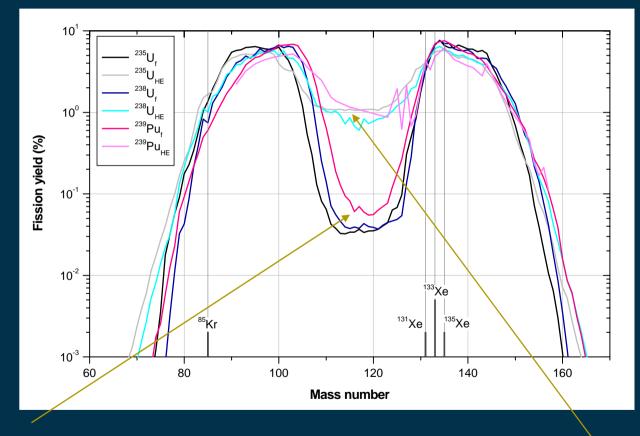


- During fission of uranium or plutonium in a nuclear reactor, thermal (slow) neutrons are used, whereas during a nuclear explosion the fission is induced by fast neutrons. The full fission sequence in a device is finished within a microsecond.
- There is little time for complex activation build-up in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



Fission induced by thermal (fission spectrum) neutrons

Fission induced by high energy neutrons (14.7 MeV)

Cumulative fission yields



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

- 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)
- ¹³³Xe has high production rates and a not too short half-life. Therefore this xenon isotope is the one most observed in environmental samples



- 1 kiloton (kton) nuclear explosion is equal to an explosion of 1000 tons of TNT, which equals 10¹² calories = 4.2 10¹² Joules
- The average total energy released in fission of one uranium-235 or plutonium-239 atom is $200 \text{ MeV} = 3.2 \ 10^{-11} \text{ J}$
- 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



- ~ 180 MeV is immediately available as energy from each fission event which is equal to ~ $1.45 \ 10^{23}$ fissions per kton
- Activity $A = \lambda \times N(t) = \ln(2) \times N(t) / T_{1/2}$

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

 Depending on the fission material (²³⁵U, ²³³U or ²³⁹Pu) between 1.08 10¹⁶ Bq and 1.33 10¹⁶ Bq of ¹³³Xe will be created in a 1 kton nuclear explosion

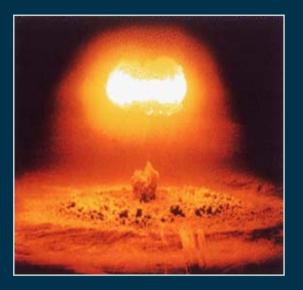
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Nuclear tests in different environments







Atmospheric Nuclear Tests

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

Underground Nuclear Tests

- Seismic waves
- Radionuclides: vented gases
- Possible hydro-acoustic/infrasound 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



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 (nonevasive) 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.



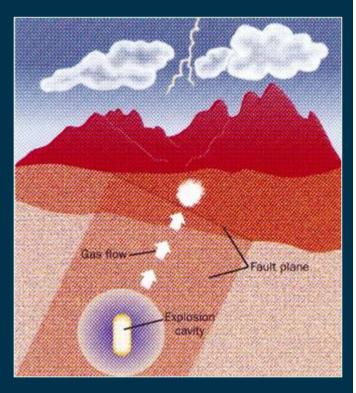
Noble Gases:

- ▲ Argon-37: activation product. Decays with no photons
- **Krypton-85** (t $_{1/2}$ = 10.7 y): world-wide high background
- ▲ Xenon's: four isotopes are suitable for CTBT verification (^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe)
- An underground nuclear test creates seismic waves, possibly hydroacoustic /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

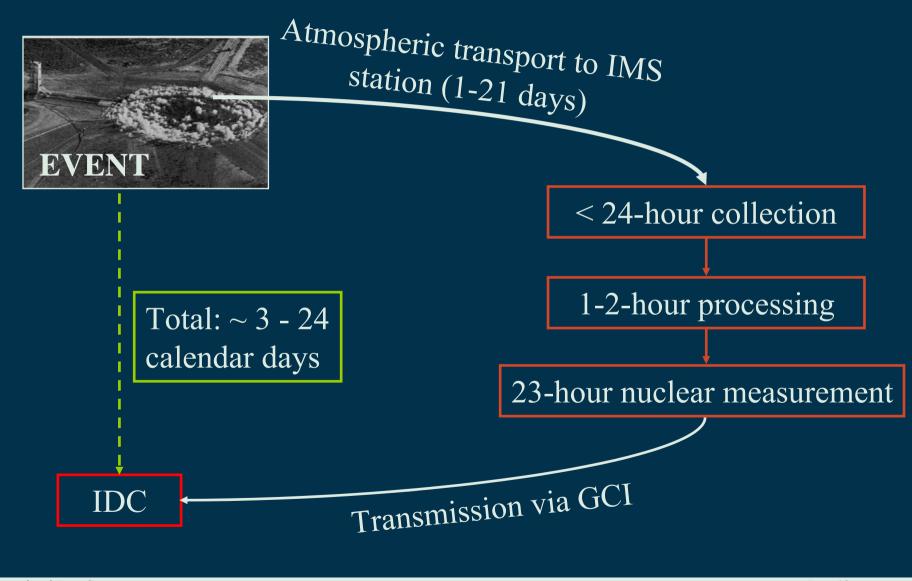


- Unintentional release of radioactive matter to the atmosphere due to failure of the containment system (containment failure) (0 100 %)
- Early venting due to high pressure of the explosion and other dynamic effects (pushes gas through cracks and fissures in the bedrock) (~ 10 %)
- Venting due to opening 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





Sources of Radioactive Xenon

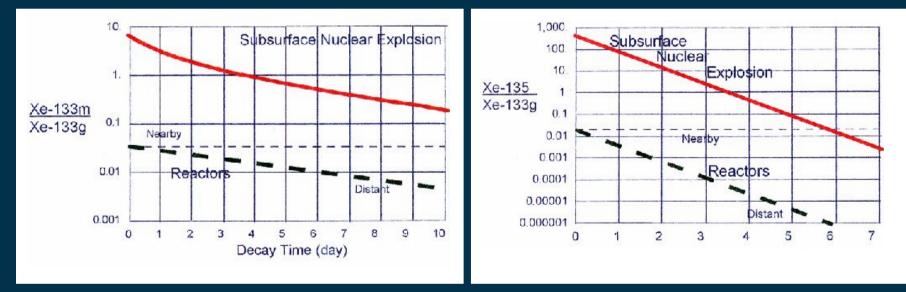


- Nuclear Reactors: mainly ¹³³Xe
- ▲ Fuel reprocessing plants: mainly ^{131m}Xe
- ▲ Hospitals:

mainly ^{131m}Xe and ¹³³Xe

Nuclear Explosions: mainly ¹³⁵Xe, ^{133m}Xe and ¹³³Xe

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

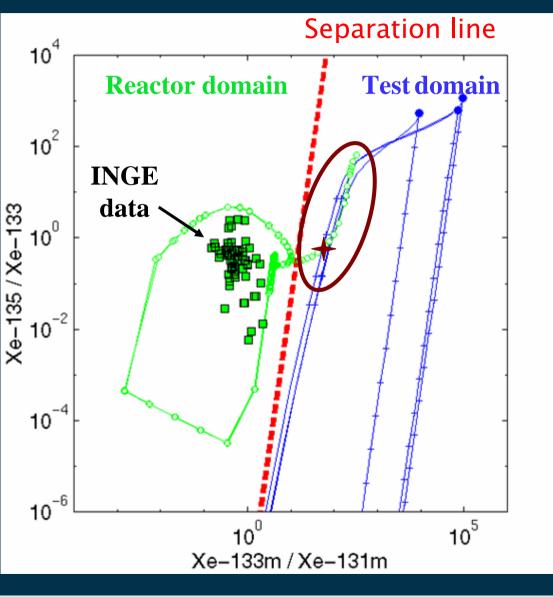


The plot separates into two domains due to duration of the neutron interaction:

- 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}Xe (11.9 d), ^{133m}Xe (2.19 d), ¹³³Xe (5.24 d), ¹³⁵Xe (9.10 h)

▲ Minimum detectable concentration: $< 1 \text{ mBq/m}^3$ for ^{133}Xe



- ➡ 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





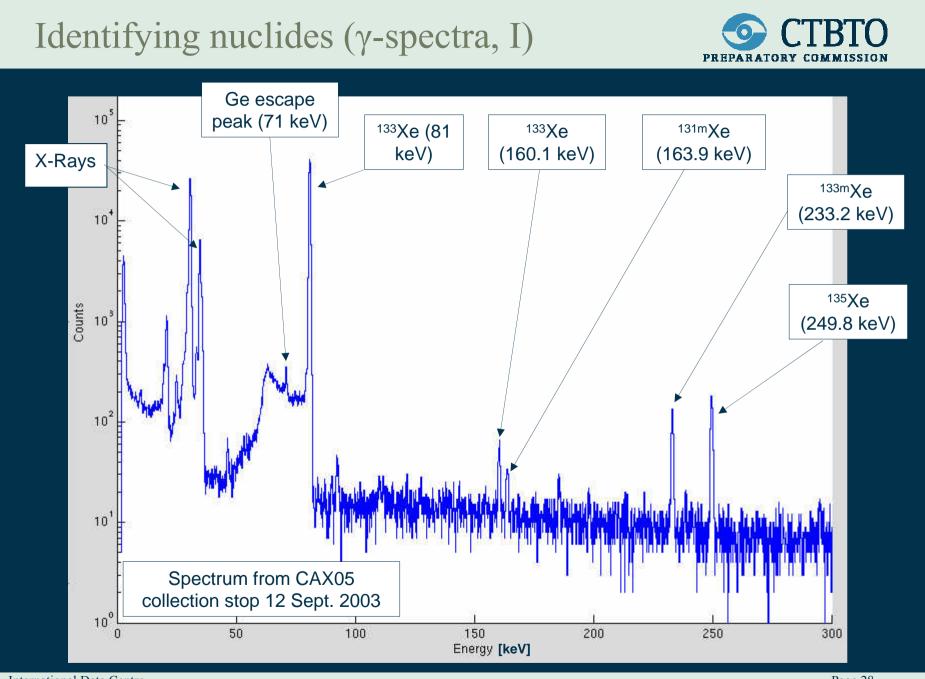
Noble gas stations map (by mid 2007)





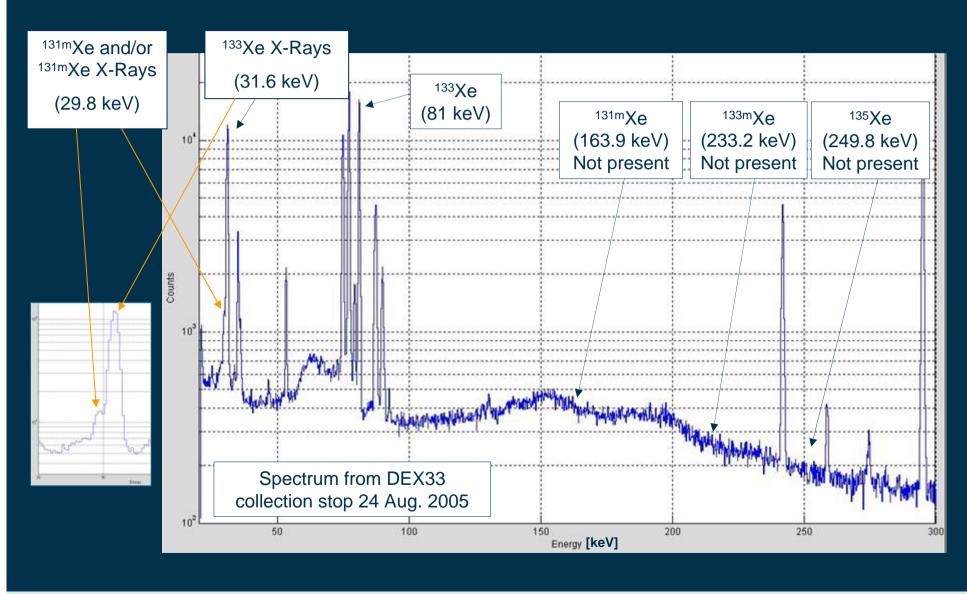


- **Sampling** of air at "high" volume air flow (> $0.4 \text{ m}^3/\text{h}$)
- **Cleaning** (removal of aerosols, water, Radon, CO_2 , ...)
- Extraction of Xenon gas from air with high efficiency (adsorption of Xenon onto charcoal followed by thermal desorption of Xenon)
- **Detection** and **calculation** of the istotopic composition
- **•** Measuring the stable Xenon volume (e.g. Gas Chromatograph)
- Sample into **archive bottle**



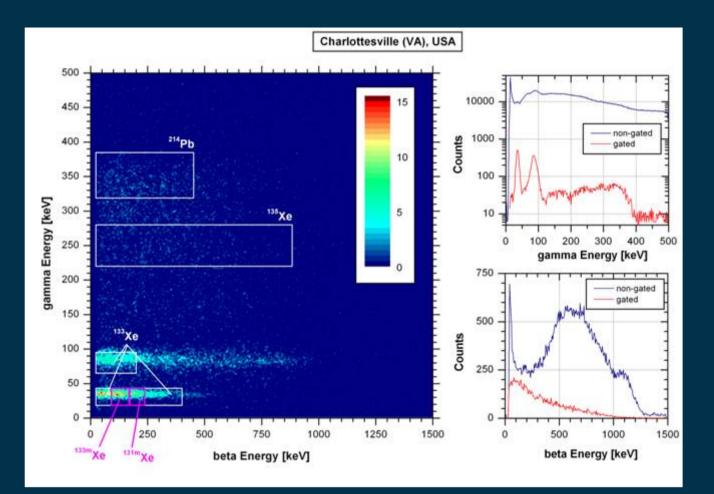
Identifying nuclides (γ-spectra, II)





Identifying nuclides (β - γ spectra)

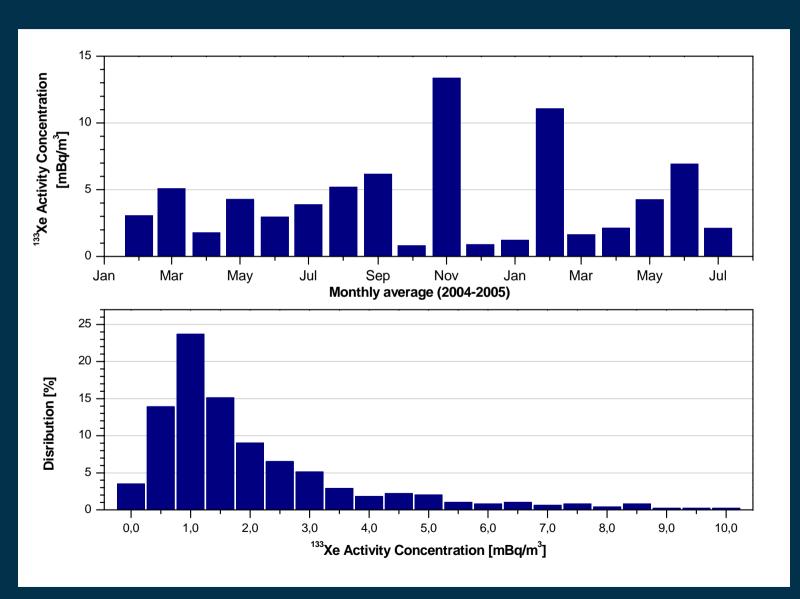




Sample Collection Stop 10 June 2002: $(0.77 \pm 0.11) \text{ mBq/m}^3$ of $^{133\text{m}}$ Xe and $(4.26 \pm 0.35) \text{ mBq/m}^3$ of 133 Xe

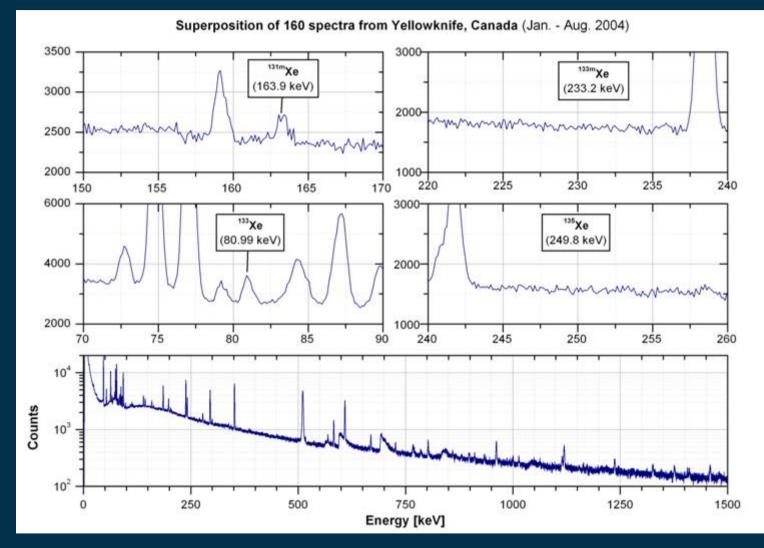
Environmental monitoring, e.g. Schauinsland mountain (Freiburg), Germany (DEX33)





Environmental monitoring, in very low background regions, e.g. Yellowknife, Canada (CAX16)





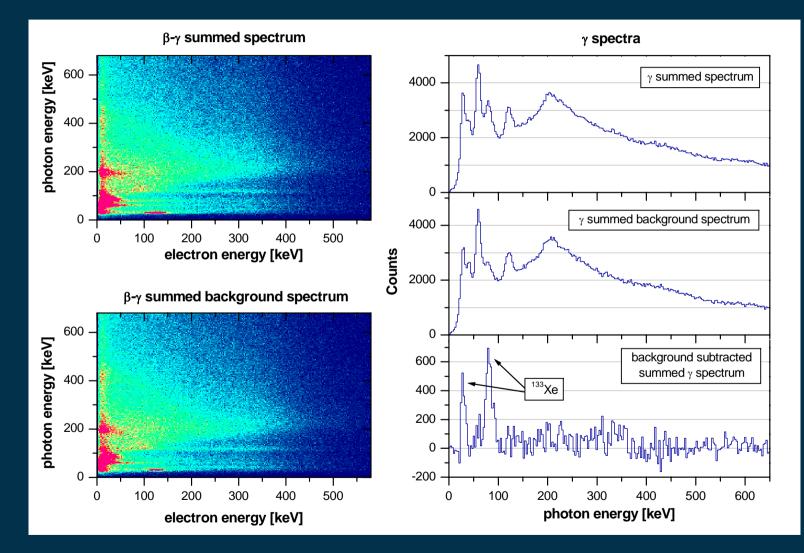
Evidence of ^{131m}Xe and ¹³³Xe presence in a remote region

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Page 32

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 ¹³³Xe and or ^{131m}Xe at very low concentrations (less than 1 mBq/m³) (e.g. on the Arctic station of Longyearbyen, Spitsbergen)
- Regular radioxenon background of ¹³³Xe (~ 1 100 mB/m³) 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/m³) (e.g. the station of Ottawa, which is surrounded by nuclear power plants and a large radiopharmaceutical production facility

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- ✓ 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:
- ➔ in building more stations;
- ➡ improving the methods;
- ➡ further understanding the signatures to properly interpret its sources;

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

Some references



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