Emissions from Fission-Based Medical Isotope Production and their Effects on the International Monitoring System

TW Bowyer
Pacific Northwest National Laboratory

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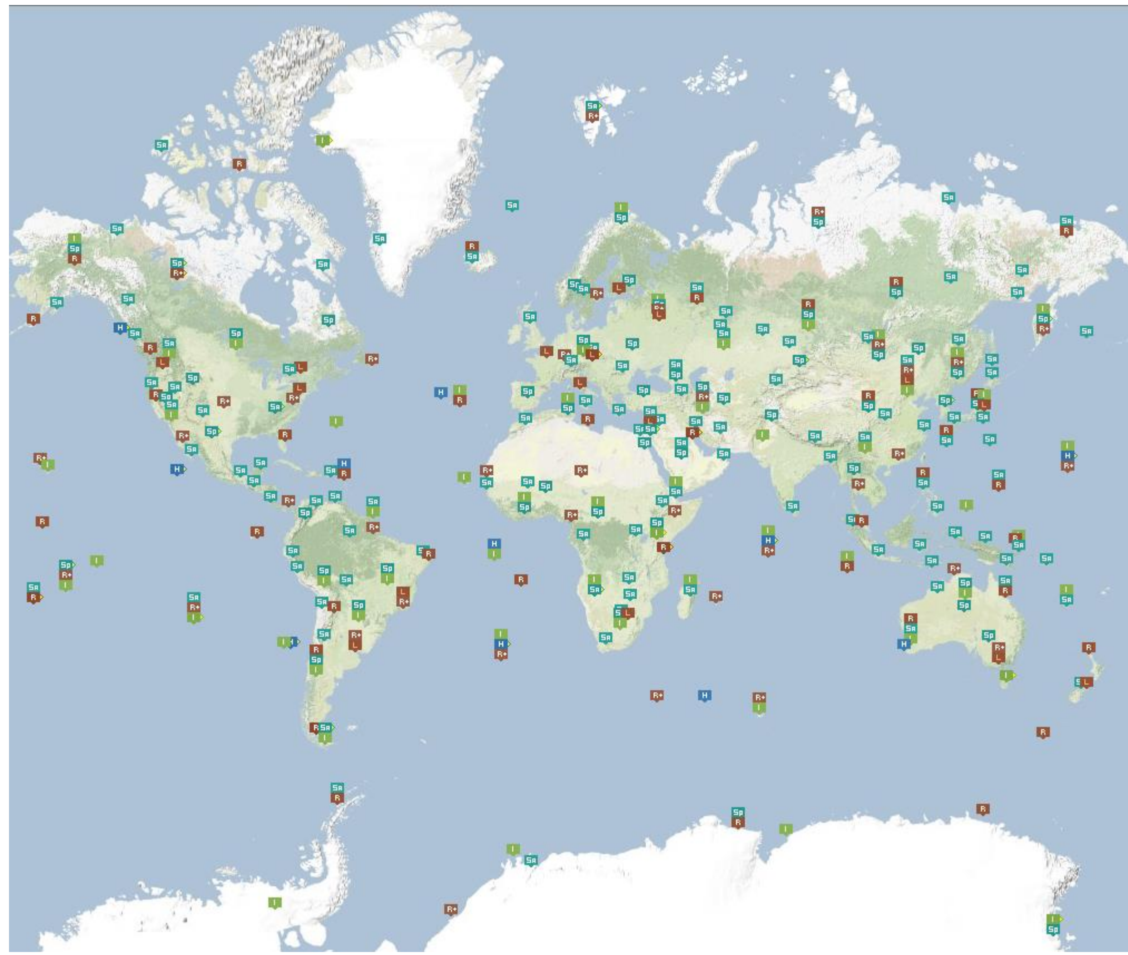
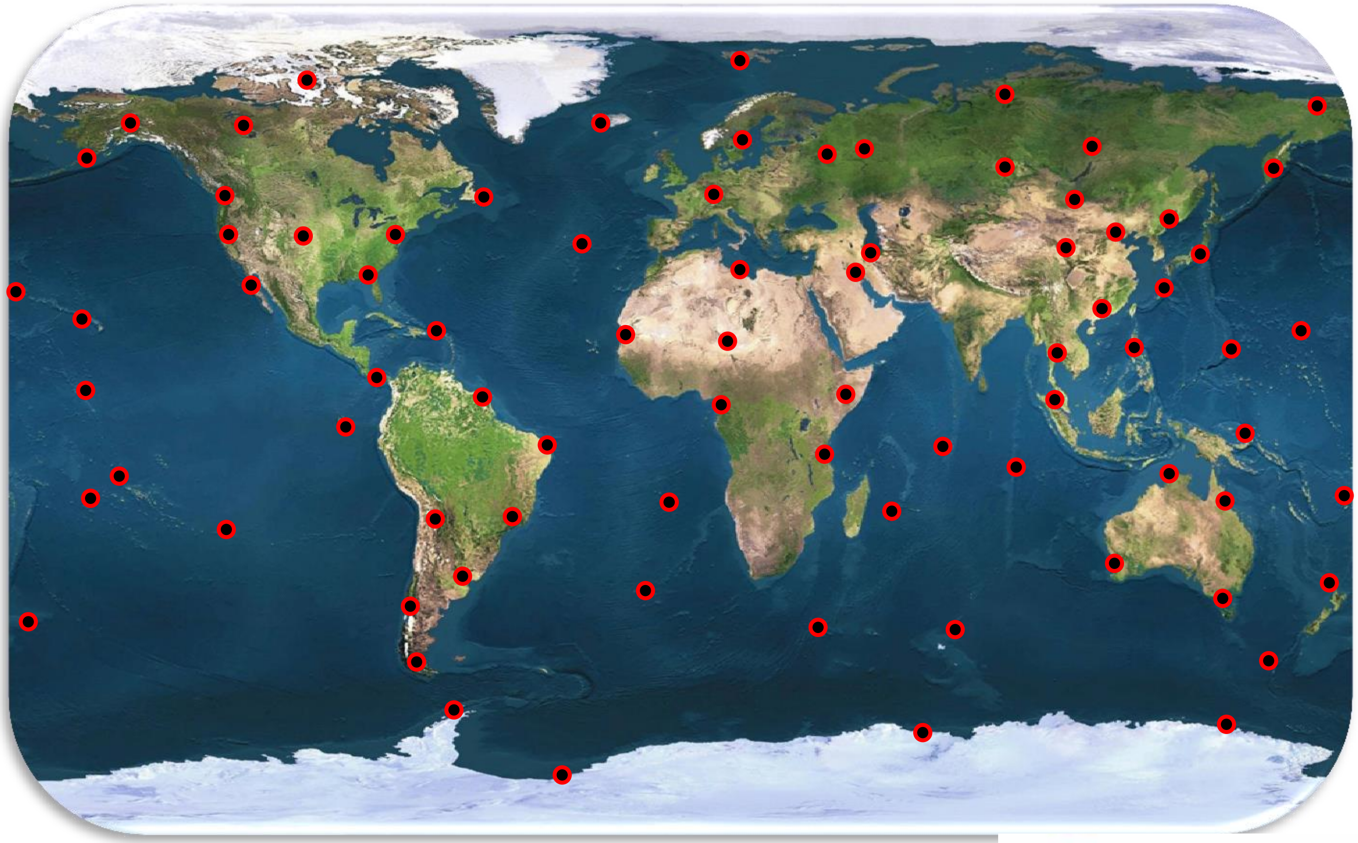
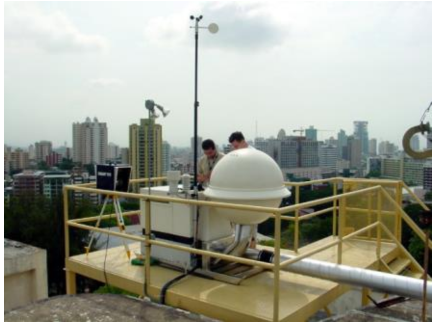
The International Monitoring System

- The International Monitoring System (IMS) is a highly sensitive network capable of detecting small-scale underground nuclear explosions
- The IMS will ultimately consist of 321 stations with the following sensors:
  - Seismic
  - Hydroacoustic
  - Infrasound
  - **Airborne radionuclide**
    - **XENON**
    - **PARTICULATES**



Radionuclide Stations in the IMS

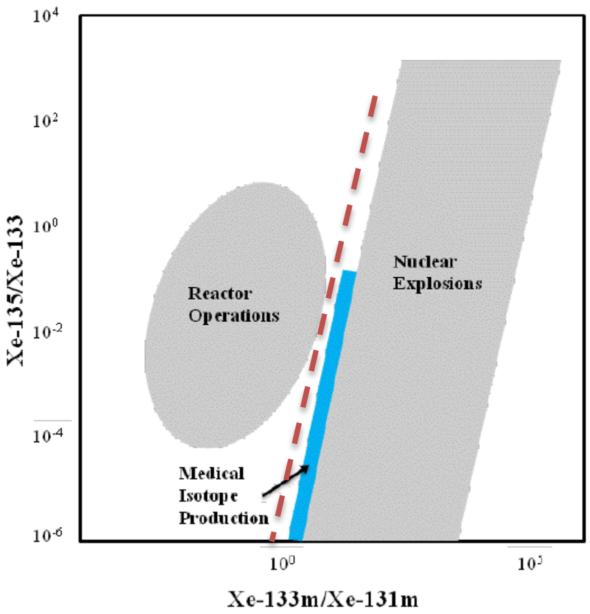




Backgrounds for RN Stations

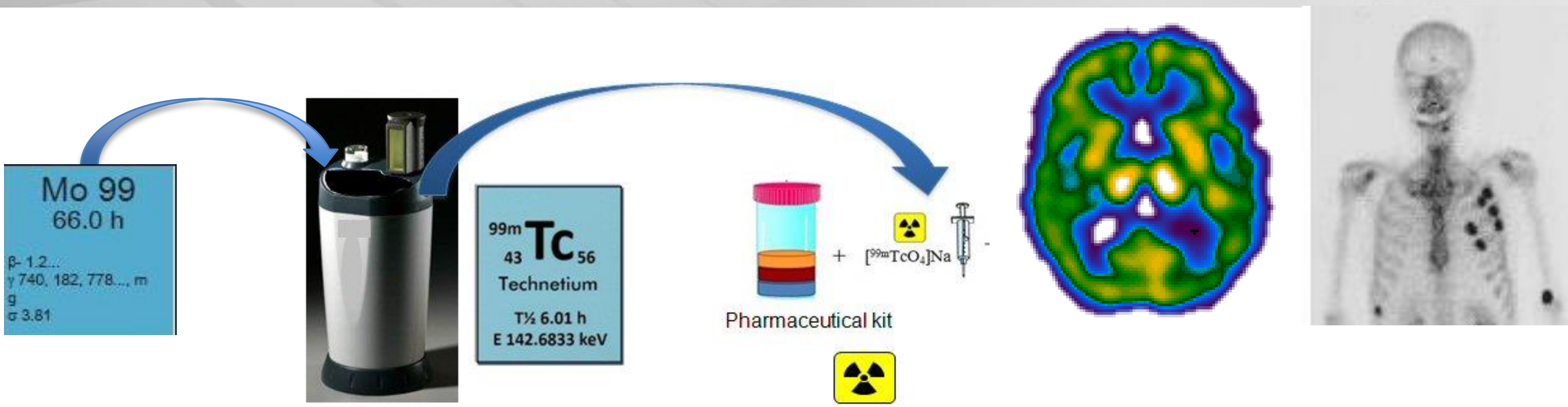
- **Particulate measurements** - There have been a number of events that were not screened out and no clear explanation given; medical isotopes were thought to be the cause
  - Detections of ^{140}La, ^{131}I, etc.

- **Xenon measurements –** Every day xenon, largely from medical isotope production, is observed
  - Multiple isotopes of xenon are observed

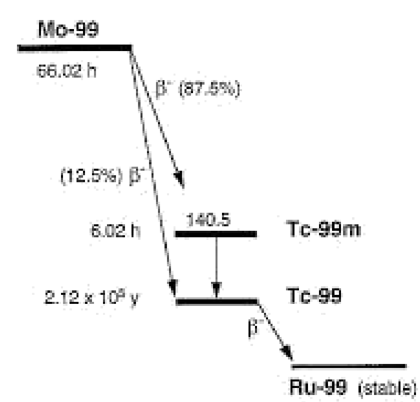


Medical Isotope Production is a major background source for CTBT Noble Gas measurements

^{99}Mo/^{99m}Tc Use in Medicine

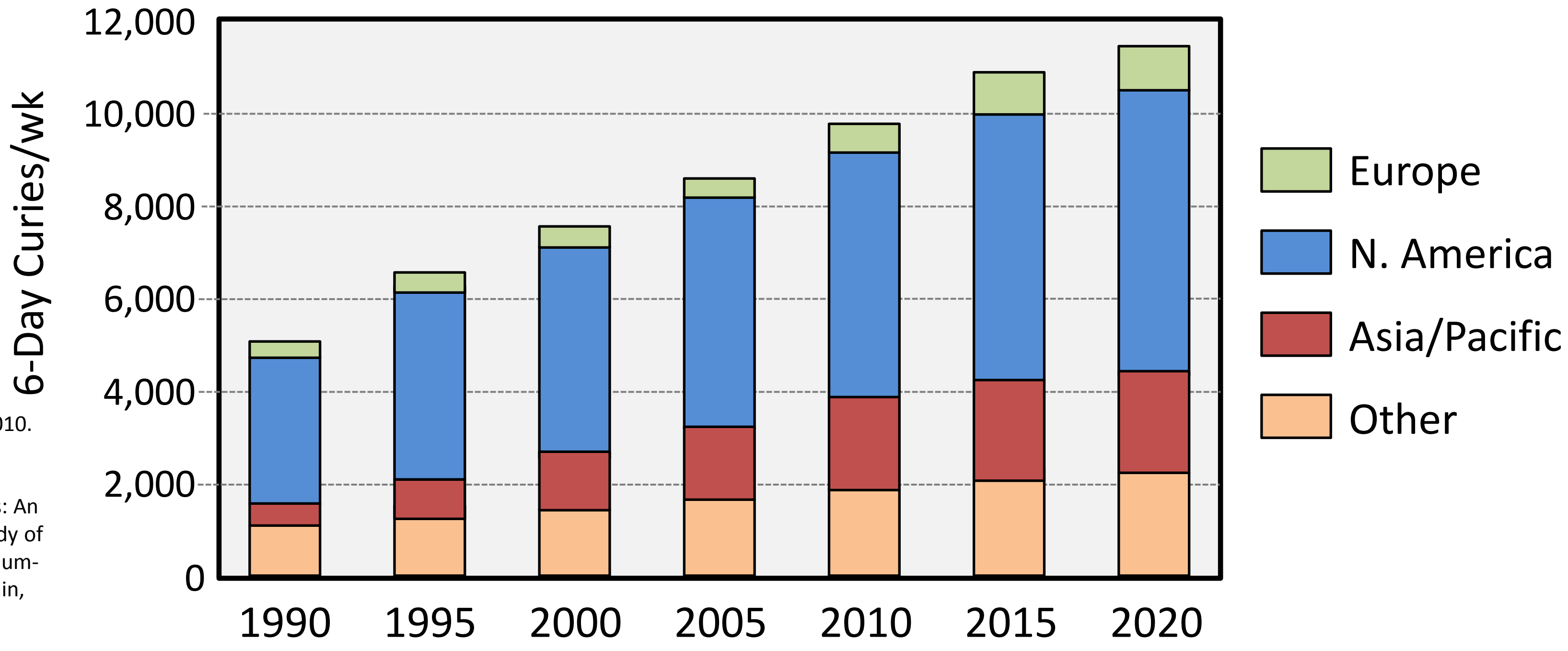


- ^{99m}Tc used in nuclear medicine (^{99m}Tc comes from ^{99}Mo decay)
  - Primary radioisotope used in the world
  - 80% of nuclear medicine diagnostic procedures
  - >30 million procedures annually[1] (once every second)
- Short half-lives of ^{99}Mo (^{99m}Tc) means no stockpiling



Global ^{99}Mo Demands

- Demand is increasing
- Typical production facility makes 100-5000 “6-day Ci/week”
- Figure depicted here is the estimate from 2010
- More recent information indicates that developed country Mo-99 production may be equilibrating, but increasing in developing countries

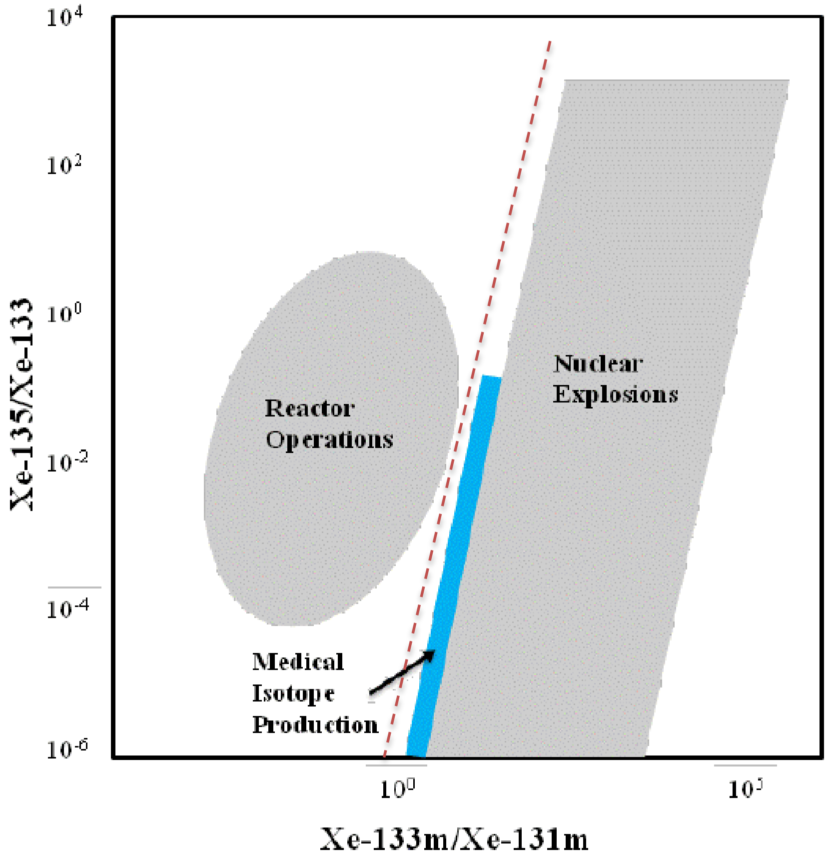


OECD-NEA, 2010. The Supply of Medical Radioisotopes: An Economic Study of the Molybdenum-99 Supply Chain, NEA 6967.

Emissions from Medical Isotope Production and Interference with IMS Measurements

- Emissions from nuclear explosions can be anywhere from 0 to 100% of the inventory
  - 0 – 10^{16} Bq per kiloton; well contained explosions are 'low'
- Emissions from medical isotope production are 10^9–10^{13} Bq /day
  - Isotopes released are similar to explosions
- Isotopes emitted tend to create a 'fog' of ^{133}Xe

Discrimination Plot



XENON-133 IS DETECTED IN SOME LOCATIONS EVERY DAY FROM ISOTOPE PRODUCTION

Contribution to Background Comparisons

Factor	Fission-Based Medical isotope production	Nuclear explosions	Nuclear power reactors
Fuel/target type	LEU	Pu/HEU	LEU
Duration of Irradiation	Short	'Immediate'	Long
Major nuclides released	^{133}Xe, …	^{133}Xe, ^{135}Xe	^{133}Xe
Release amounts	Daily releases of 10^{9}-10^{13} Bq/day	1 kT $\rightarrow$ 10^{16} Bq produced; Much less is likely from underground explosions (<10^{11}-10^{12} Bq)	10^{9} Bq/reactor/day

Fission-based production of ^{99}Mo produces fission gases including ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe

- *Neither neutron activation: n + ^{98}Mo $\rightarrow$ ^{99}Mo, nor accelerator production, e.g., ^{100}Mo(ϒ,n)^{99}Mo produces fission gases*
- *^{133}Xe emissions can be entirely eliminated by using activation or accelerator methods in lieu of fission methods*

These radioxenon isotopes are also used to detect nuclear explosions

The International Community detects this "background" on a regular basis under auspices of the Comprehensive Nuclear-Test-Ban Treaty (CTBT)

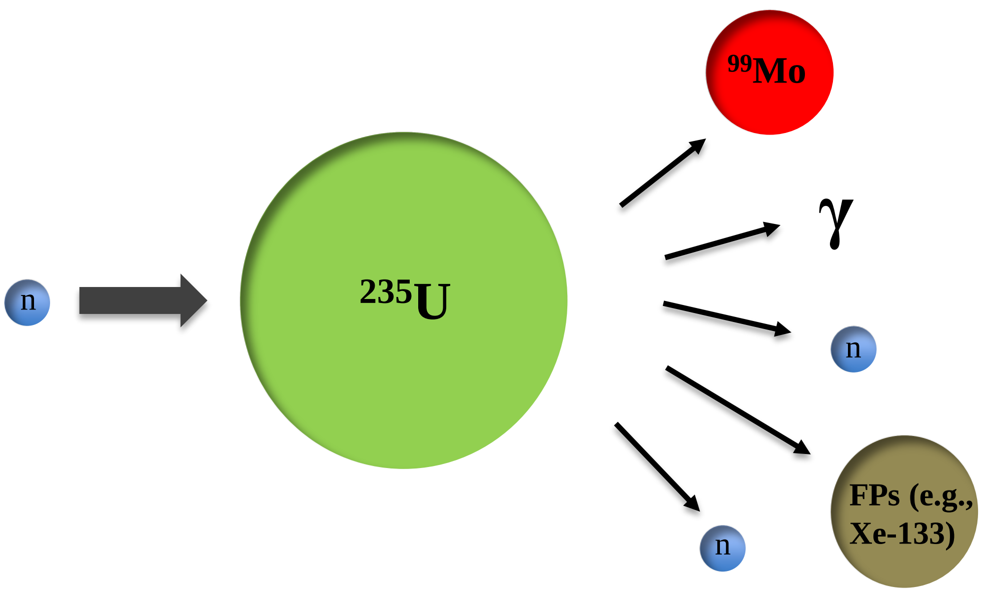
Background influence

- The production of fission-based medical isotopes is similar in many ways to a nuclear explosion
  - Irradiation of uranium, followed by dissolution as soon as possible

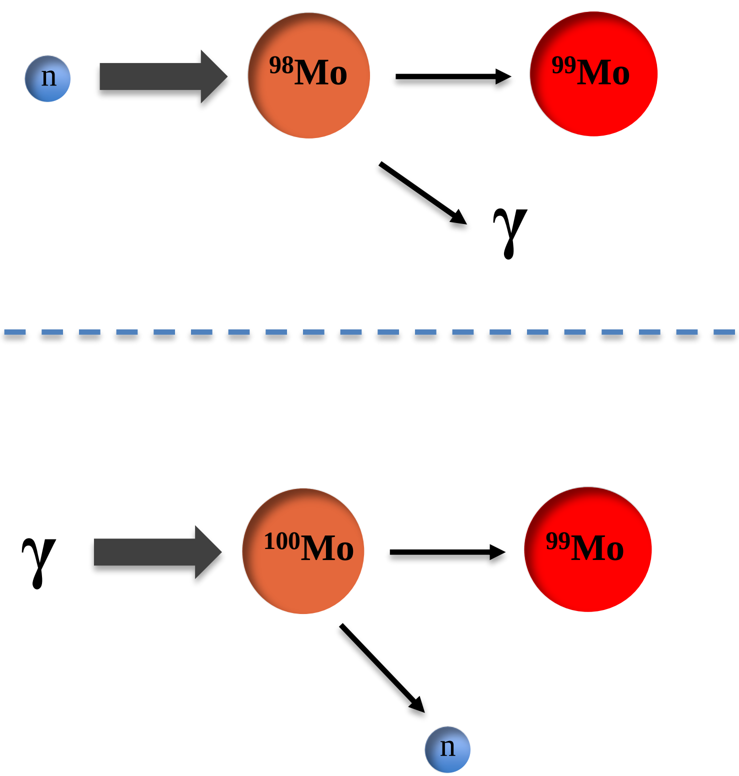
- A constant presence of xenon causes a background that can be subtracted, but this “fog” is the same isotope we are looking for and therefore the **<u>statistical precision</u>** to which we can subtract it is affected

Fission vs. Activation

Mo-99 Production Using Fission Also Produces Xe-133



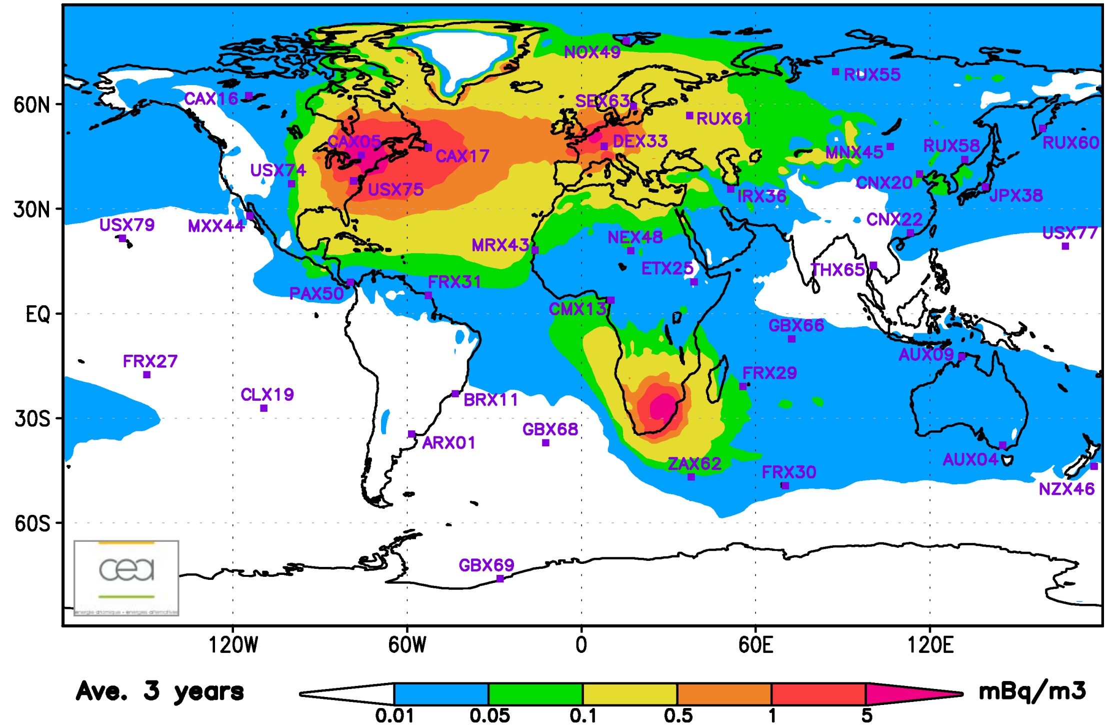
Alternate Production Using Neutron Activation or Accelerator Does Not Produce Xe-133



A Few Major Producers Dominate Worldwide Emissions

- ^{133}Xe isotopes created cause a daily background (aka “Xenon Weather”) that must be subtracted

- Some tools exist to track and account for this background, but the situation is worsening because of globalization of production and could worsen if fission based production increases



Achim, 2010

What is the size of this effect?

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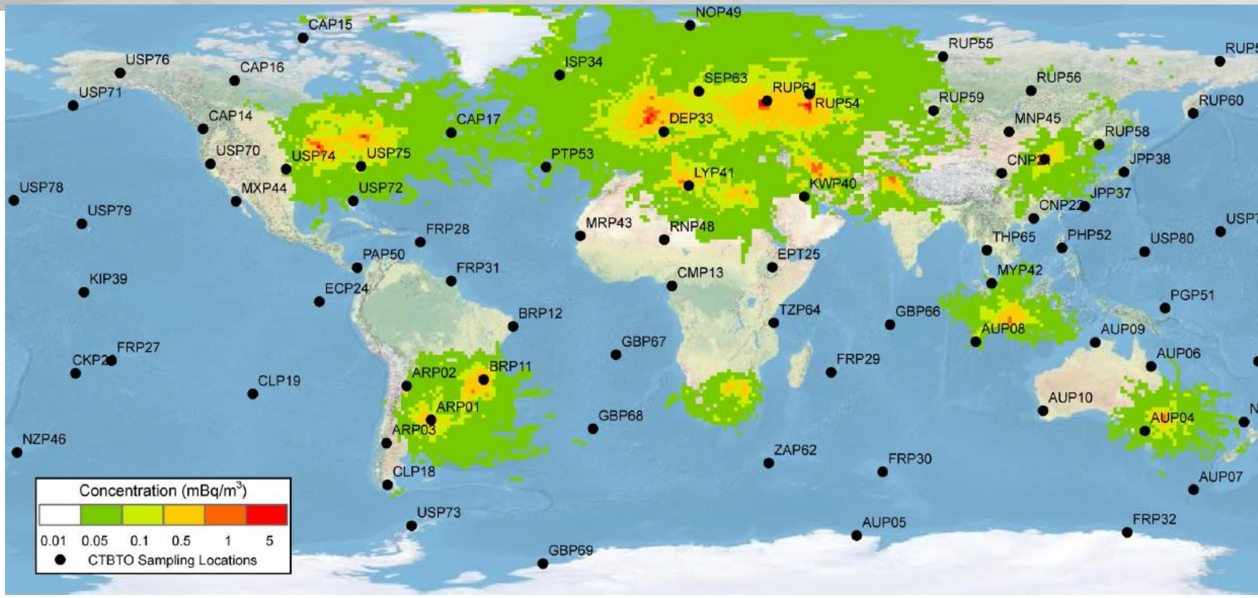
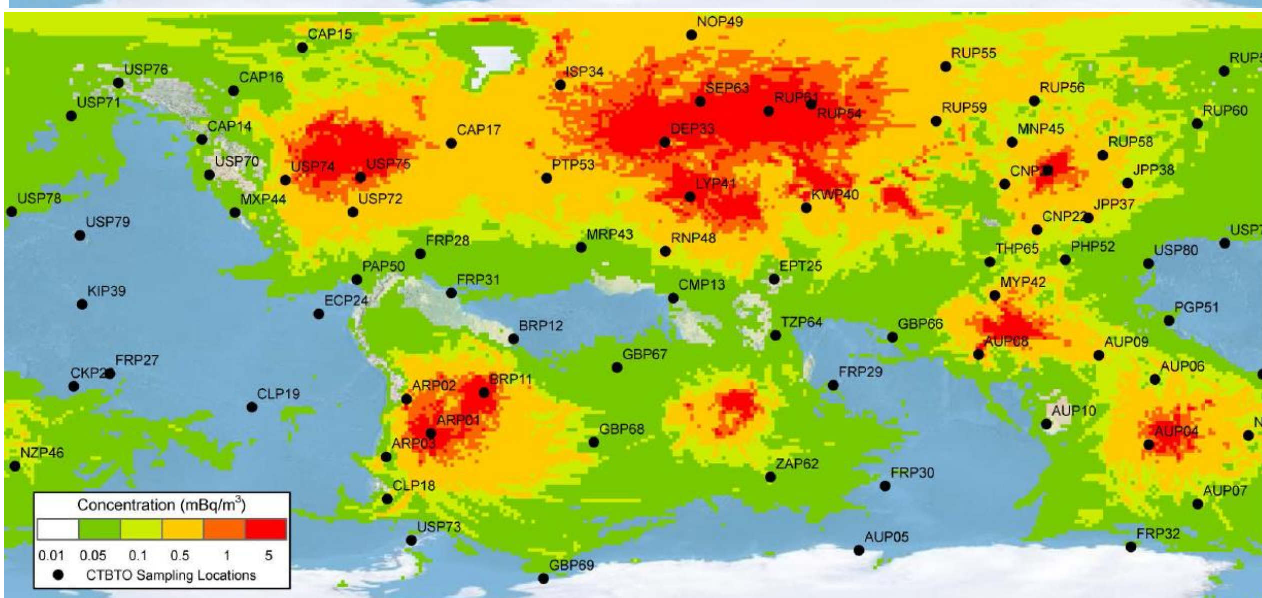


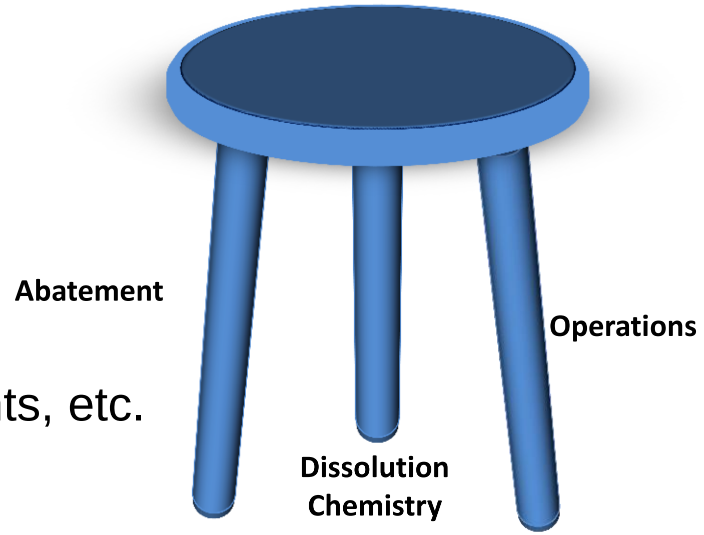
"Hopeful case" (w/ action)
Theoretical releases
of $5x10^9$ Bq/day of ^{133}Xe



"Bad case" (no action)
Theoretical releases
of $1x10^{12}$ Bq/day of ^{133}Xe

Factors Affecting Xenon Releases

- The amount of potential radioisotope emissions are affected by the amount of ^{99}Mo produced, and
- For a given production, the amount of emissions are affected primarily by 3 factors:
  - Dissolution chemistry
    - Alkaline v. acidic
  - Abatement control systems
  - Operational issues
    - Leaky valves, seals, etc.
    - Standard operating procedures, accidents, etc.



TW Bowyer, RF Kephart, PW Eslinger, JI Friese, HS Miley, PRJ Saey. 2013. "Maximum Reasonable Radioxenon Releases from Medical Isotope Production Facilities and Their Effect on Monitoring Nuclear Explosions." *J. of Environmental Radioactivity*

What Does This Look Like?

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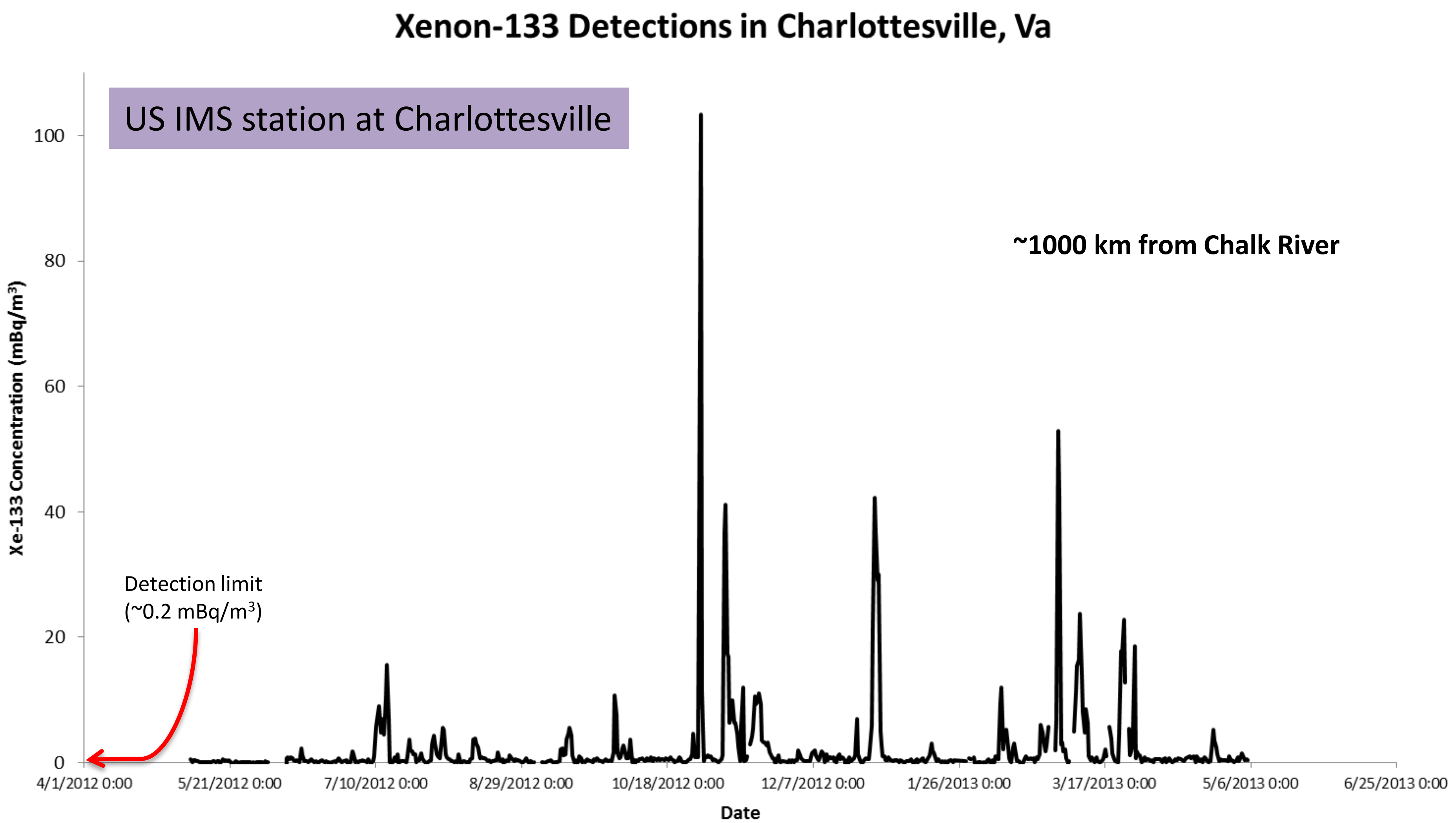

Recent Xe Detection Reported at the Takasaki IMS Station

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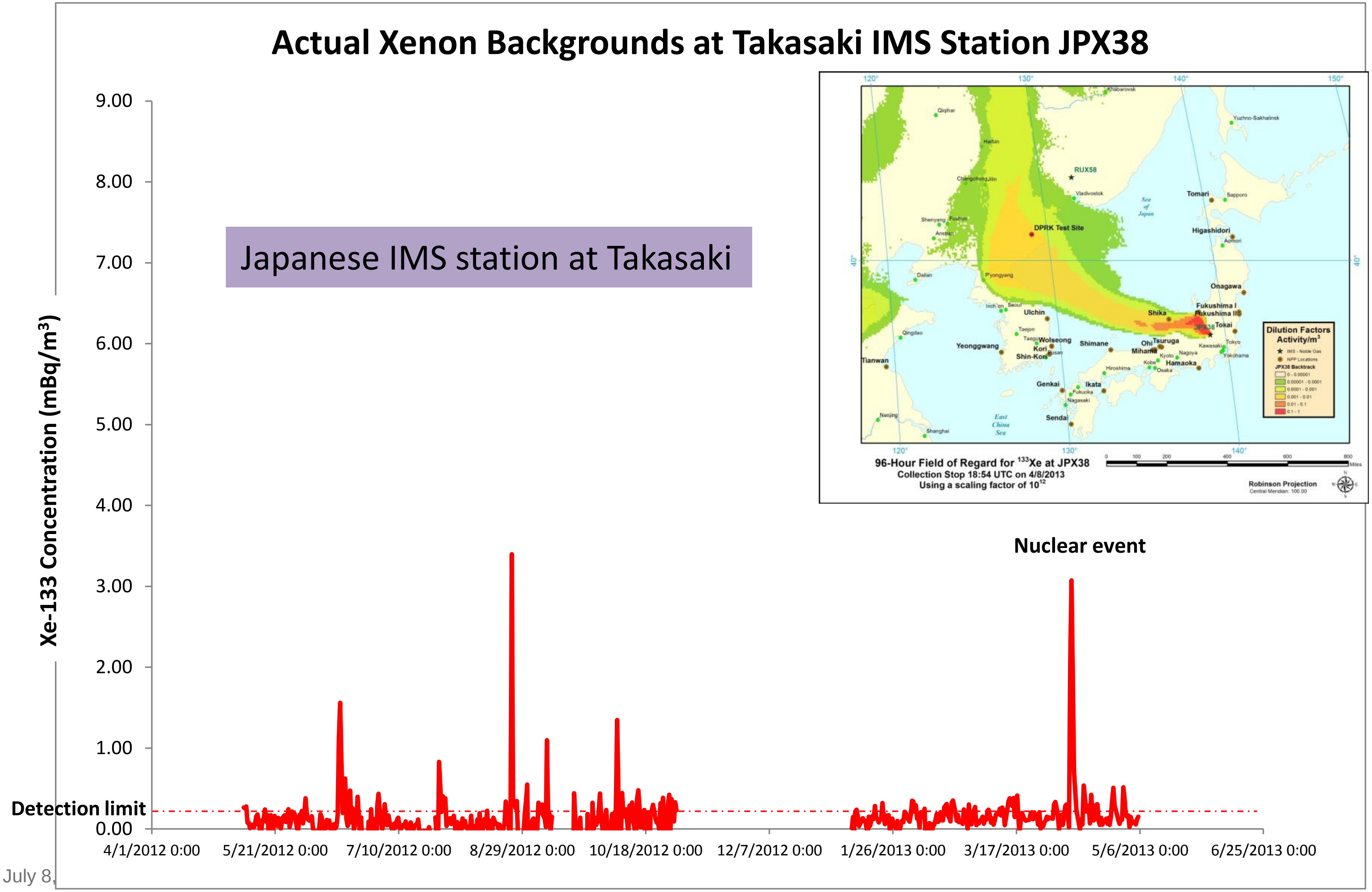

Medical Isotope Simulations for Takasaki

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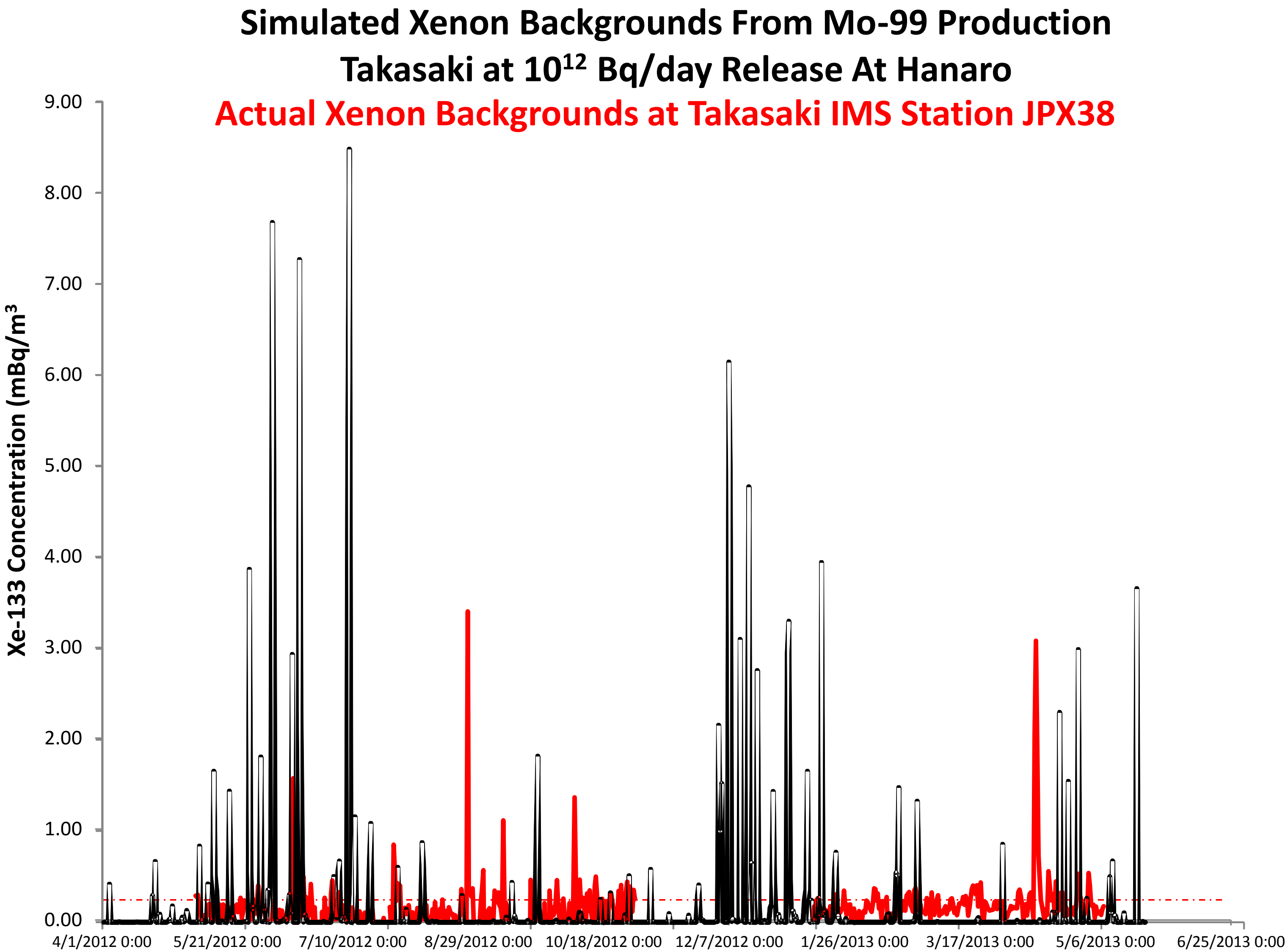

Graphical View-Yearly Detections at 10^{12} Bq/day

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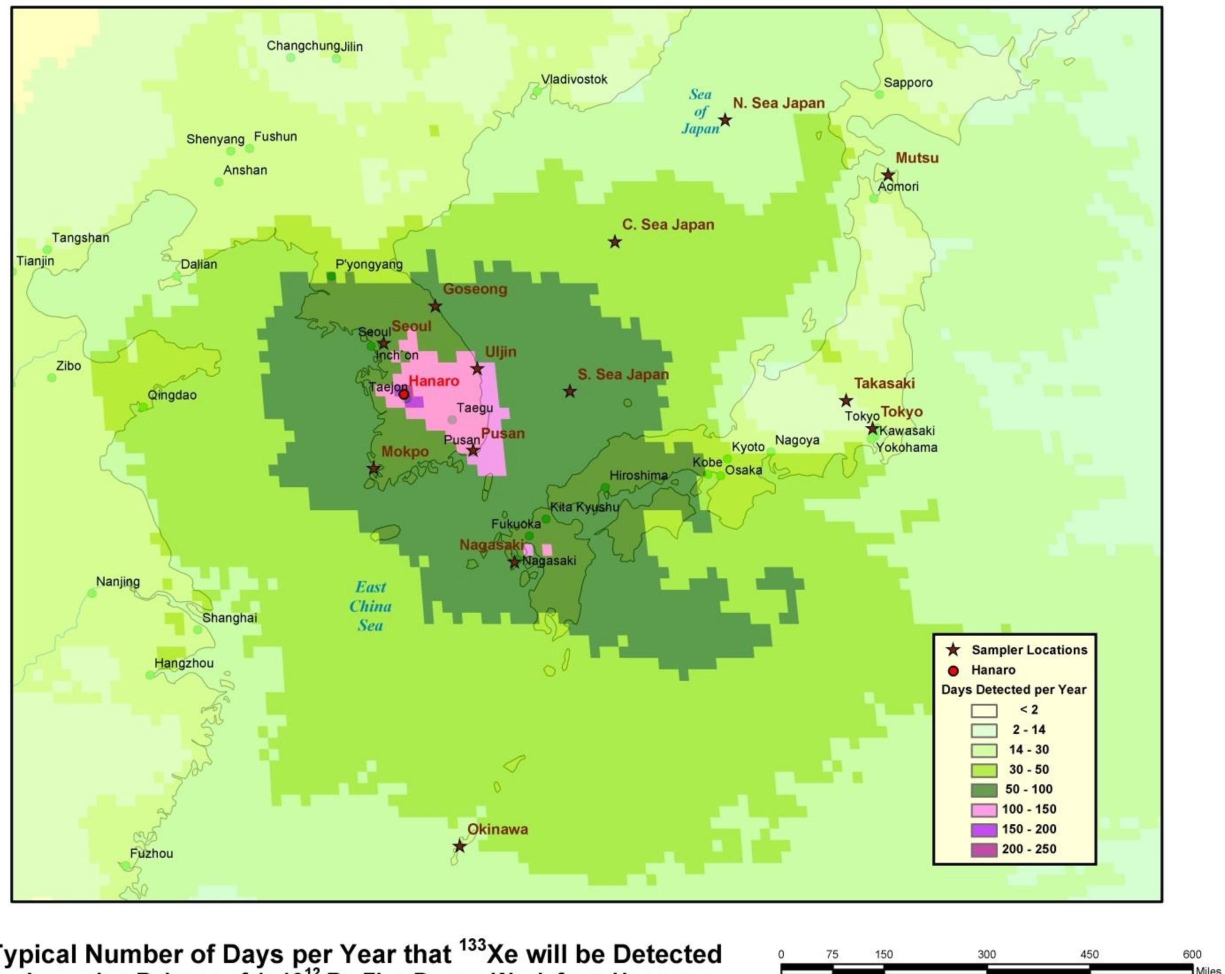

Typical Number of Days per Year that ^{133}Xe will be Detected
Assuming Release of 1×10^{12} Bq Five Days a Week from Hanaro
MDC = 0.2 mBq/m^3

KAERI is working with the international community to aggressively address their xenon emissions!

Amount of emissions we hope to be below 10^{12} Bq/day!

How much Xe-133 can be emitted and not adversely affect nuclear explosion monitoring?

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- Calculations performed and validated indicate that for most locations, emissions in the range of ~<u>**$5x10^9$ Bq/day**</u> are acceptable, and within the realm of possibility for producers (i.e., it can be done)

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Maximum reasonable radioxenon releases from medical isotope production facilities and their effect on monitoring nuclear explosions

Theodore W. Bowyer[a,*], Rosara Kephart[a], Paul W. Eslinger[a], Judah I. Friese[a], Harry S. Miley[a], Paul R.J. Saey[b]

[a] Pacific Northwest National Laboratory, National Security Division, 902 Battelle Blvd, P.O. Box 999, MSIN K8-27, Richland, WA 99354, USA
[b] Vienna University of Technology, Atomic Institute of the Austrian Universities, Stadionallee 2, 1020 Vienna, Austria

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ABSTRACT

Fission gases such as ^{133}Xe are used extensively for monitoring the world for signs of nuclear testing in systems such as the International Monitoring System (IMS). These gases are also produced by nuclear reactors and by fission production of ^{99}Mo for medical use. Recently, medical isotope production facilities have been identified as the major contributor to the background of radioactive xenon isotopes (radioxenon) in the atmosphere (Stocki et al., 2005; Saey, 2009). These releases pose a potential future problem for monitoring nuclear explosions if not addressed. As a starting point, a maximum acceptable daily xenon emission rate was calculated, that is both scientifically defendable as not adversely affecting the IMS, but also consistent with what is possible to achieve in an operational environment. This study concludes that an emission of 5×10^9 Bq/day from a medical isotope production facility would be both an acceptable upper limit from the perspective of minimal impact to monitoring stations, but also appears to be an achievable limit for large isotope producers.

1. Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) bans explosions from any environment, including in the atmosphere, underwater, and underground. An international system was designed and is now under construction intended for the verification of the treaty. This International Monitoring System (IMS) is comprised of 321 stations (UNGA, 1996; Dahlman et al., 2009) at various locations across the globe. The data collected by this system of sensors is transmitted to an International Data Center, which analyzes the data every day. The IMS comprised of four types of technologies designed to detect nuclear explosions conducted in the various environs. For detection of underground nuclear explosions, seismic sensors are used to detect vibrations in the ground, hydrophones are used to detect water pressure pulses from underwater detonations, microphones are used to detect low frequency sounds from atmospheric detonations, and a number of radionuclide sensors are used to detect airborne nuclear debris that can be emitted from either atmospheric, underwater, or underground detonations.

The seismic, hydrophonic and acoustic sensors cannot discriminate nuclear explosions from phenomena such as the use of conventional explosives. Radionuclide sensors on the other hand, can identify nuclear explosions and discriminate between conventional explosions and actual nuclear explosions. The radionuclide technology of the International Monitoring System (IMS) consists of radionuclide particulate detectors (based on high resolution gamma spectrometry) and radioxenon detectors (based on high resolution gamma spectrometry or beta-gamma coincidence counting). The particulate systems are designed to identify debris from an atmospheric, shallow underground or shallow underwater nuclear explosion, whereas the radioxenon systems can also measure fission gases originating from deep underground or deep underwater explosions. Radioactive xenon has been used effectively for over a decade in the IMS, and various technologies and algorithms have been used to detect and discriminate faint radioxenon emissions from other anthropogenic sources such as reactor operations and medical isotope production (Kalinowski et al., 2010).

Each year millions of procedures utilizing medical isotopes are performed to address issues such as heart studies and other critical activities. Among the isotopes used for medical procedures, ^{99m}Tc, produced as decay product from ^{99}Mo, is by far the most prevalent (IAEA, 1989). There are two main production methods of ^{99}Mo; neutron capture on ^{98}Mo and through uranium fission. In the latter production route, the uranium targets are dissolved, followed by chemical separation to obtain a purified ^{99}Mo product (IAEA, 2004).

* Corresponding author. Tel.: +1 509 372 6401.
E-mail address: ted.bowyer@pnnl.gov (T.W. Bowyer).

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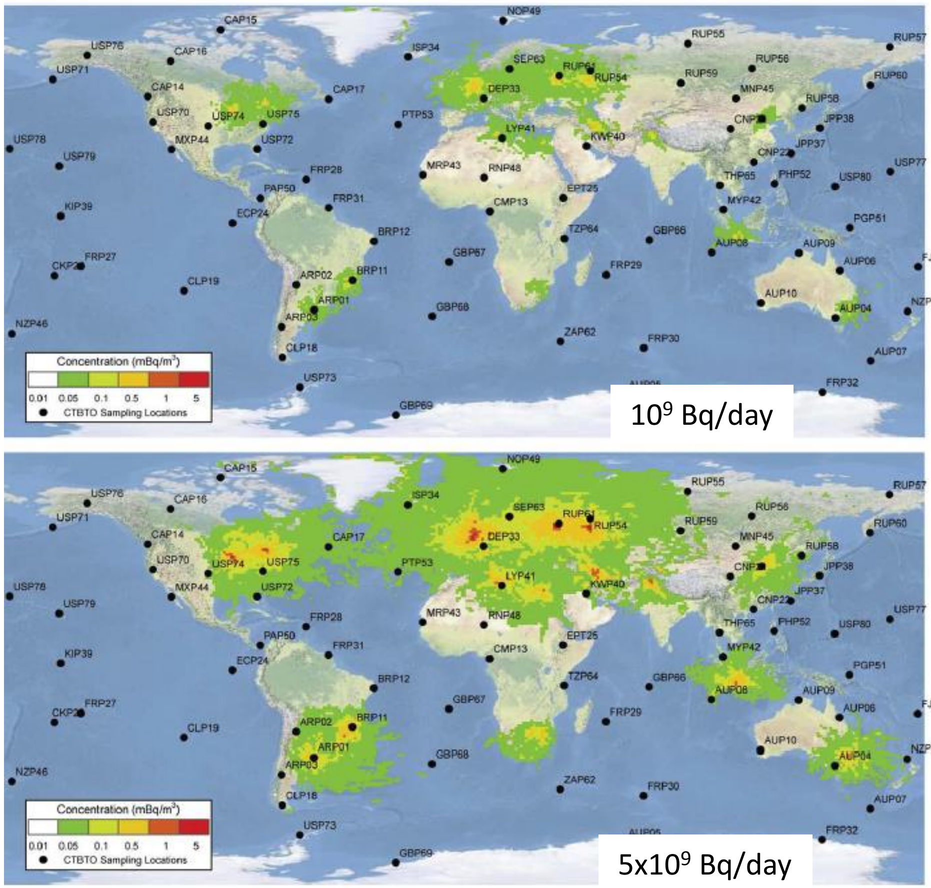
Global maximum calculated daily concentrations of Xe-133 for various releases



What Can We / Should We Do About This?

- Engage and raise awareness
  - Hopefully producers – especially new ones - will be able to build in emissions control
  - Some producers have already agreed to engage and may officially adopt emission controls to levels needed by the IMS

- Develop tools to better allow for discrimination of emissions
  - This will never be sufficient, since ^{133}Xe emitted will always create a "fog"

- Supply stack monitoring data to the IDC
  - Data on a regular basis will allow for better discrimination/backtracking of *current* producers

- Why should a producer work with the CTBTO?
  - The CTBTO can work with producers to assure confidentiality of data and to inform the public if there is an issue

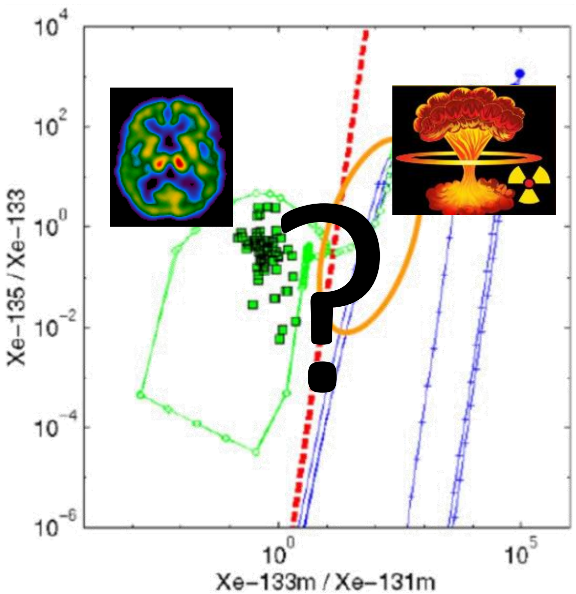
CTBTO Executive Secretary-Elect Lassina Zerbo and IRE CEO-General Manager Jean-Michel Vanderhofstadt sign a low-emissions pledge during the recent S&T2013 Conference in Vienna.

Specific Actions

- Identify and share information about all fission-based producers
- Encourage interaction between producers and CTBT community (WOSMIP)
- Encourage producers to keep emissions low, provide stack monitoring data to the IDC
- Encourage scientific investigations to understand emissions, measure background, find ways to exploit current data, and explore ways to keep emissions low

Summary

- ^{99}Mo is an important medical radionuclide and the demand is growing
- Effluents from ^{99}Mo production are observed in the IMS
- One of the most problematic effluent streams from ^{99}Mo production is gaseous xenon
- More knowledge about the processes used in ^{99}Mo production will lead to a more robust understanding of IMS detections



WOSMIP 2015

May

Brussels, Belgium

Workshop on Signatures of Medical and Industrial Isotope Production