CTBTO
PREPARATORY COMMISSION

COMPREHENSIVE NUCLEAR-TEST-BAN TREATY ORGANIZATION

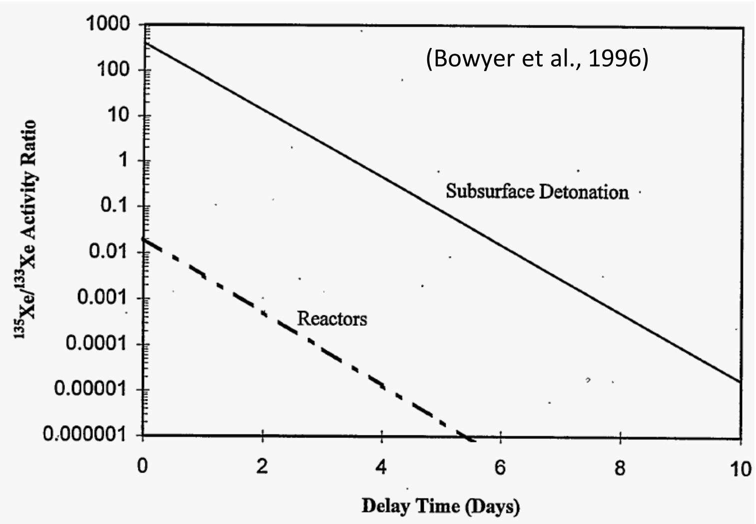
Review of Xe-135 observations, specifically in Japan, and possible sources that might explain high Xe-135/Xe-133 ratios

Martin Kalinowski, Jonathan Baré, Yuichi Kijima, Jolanta Kuśmierczyk-Michulec, Boxue Liu, Robin Schoemaker, Anne Tipka

IDC/CTBTO, Vienna, Austria

Introduction

- An elevated level of the $^{135}Xe/^{133}Xe$ activity ratio is known to be an important indicator for a prompt release from an underground nuclear explosion.
- De Geer (2012) use the outstanding high $^{135}Xe/^{133}Xe = 4.085$ observed in May 2010 to support the hypothesis of a nuclear test by the DPRK. (No seismic evidence was ever found.)
- However, there are frequent real observations of high $^{135}Xe/^{133}Xe$ activity ratios
- Therefore, understanding all sources of ^{135}Xe is vital for nuclear explosion monitoring.



of South Korea's ruling Grand National Party.[24,25] Kim claimed that the Korea Institute of Nuclear Safety had detected a xenon-133 concentration of 2.45 mBq/m^3 and 10.01 mBq/m^3 of xenon-135 at Geojin, and that "The concentration ratio of the noble gas [xenon] . . . had remained below 0.55 since 2007, but suddenly jumped to 4.085 at 1:07 a.m. on May 15." The significant levels of fairly short-lived xenon-135 ($T_{1/2} = 9.14$ hours) indicated a quite recent event.[26] A similar xenon signal had not been previously detected at Geojin.[27]

Overview

$^{135}Xe/^{133}Xe$ isotopic activity ratios observed in the atmosphere

- IMS noble gas systems
- Background campaign in Japan

$^{135}Xe/^{133}Xe$ isotopic activity ratios of various sources

- nuclear explosions
- nuclear facilities

Conclusions

- Comparison of IMS observations with various sources
- Usefulness of the $^{135}Xe/^{133}Xe$ ratio for nuclear explosion monitoring

Overview

$^{135}Xe/^{133}Xe$ isotopic activity ratios observed in the atmosphere

- IMS noble gas systems
- Background campaign in Japan

$^{135}Xe/^{133}Xe$ isotopic activity ratios of various sources

- nuclear explosions
- nuclear facilities

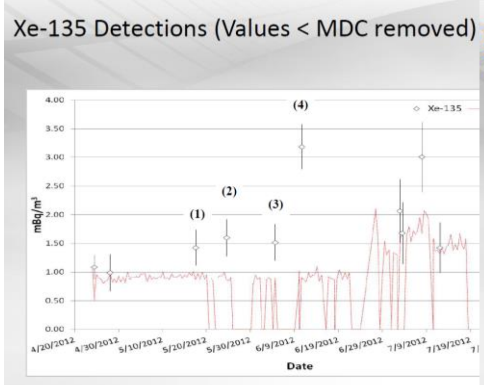
Comparison

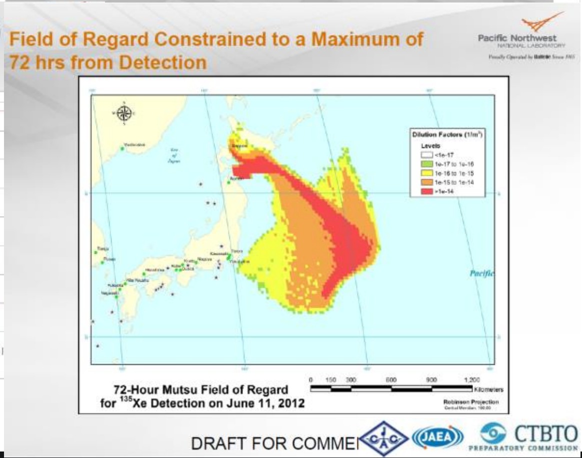
- Comparison of IMS observations with various sources
- Usefulness of the $^{135}Xe/^{133}Xe$ ratio for nuclear explosion monitoring

Early observations by PNNL

- ^{135}Xe is sometimes observed without detection of ^{133}Xe in the same sample or with very high $^{135}Xe/^{133}Xe$ activity ratio.

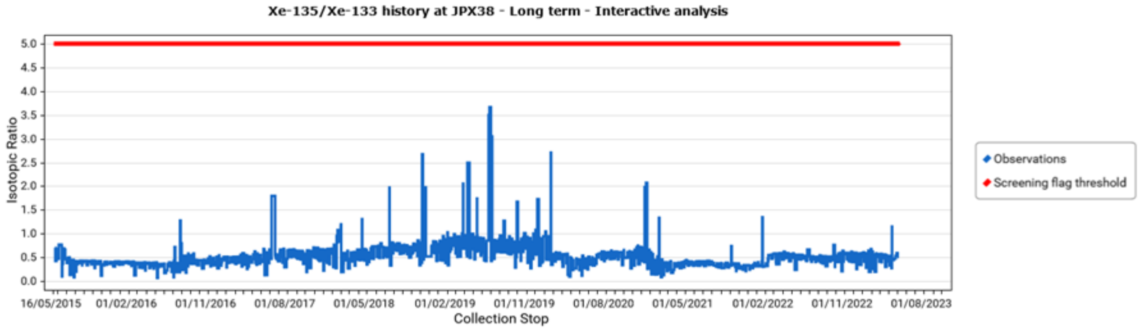
- This was reported first at SnT2011 by Ted Bowyer.
- The possible source still remains an unsolved question.

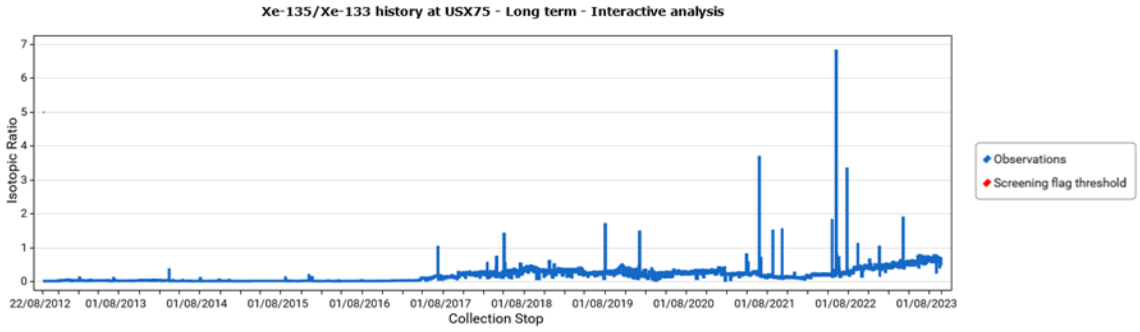




High $^{135}Xe/^{133}Xe$ ratio observations

- A baseline level of the isotopic activity ratio can be observed with occasional outliers several times larger than the baseline.
- At JPX38: elevated frequency and average level of $^{135}Xe/^{133}Xe$ activity ratios in 2019.
- At USX75, there is a long-term trend of increasing levels of $^{135}Xe/^{133}Xe$ activity ratios since 2017.





High $^{135}Xe/^{133}Xe$ ratio observations

- Elevated levels of ^{135}Xe are observed at some locations (IMS and background campaigns) but not at all IMS NG locations.
- 50% of all IMS NG systems never see $^{135}Xe/^{133}Xe > 0.5$
- Only few sites have many observations of $^{135}Xe/^{133}Xe > 0.5$

SITE	Sum of Ratio>0.5	Sum of binary>0.5	Sum of DA_XE135
Mutsu, Japan	108.15	52	11.54
Jakarta, Indonesia	39.08	34	73.18
Charlottesville, VA, USA	37.71	18	10.35
Spitsbergen, Norway	27.68	13	2.87
Stockholm, Sweden	26.94	24	30.58
Horonobe, Hokkaido, Japan	20.46	9	1.76
Wake Island, USA	10.91	3	0.53
Melbourne, VIC	9.97	4	2.06
Darwin, NT, Australia	8.06	2	0.38
Rio de Janeiro, Brazil	6.01	2	0.48
Takasaki, Gunma, Japan.	5.73	3	0.75
Tristan da Cunha, UK	5.31	2	0.31
Oahu, HI, USA	3.49	1	0.19
Ashland, KS, USA	3.37	1	0.28

High $^{135}Xe/^{133}Xe$ ratio observations

- Taking into consideration that there is a false alarm rate of about 5% of all samples we are keeping only sites that have more than 4 times $^{135}Xe/^{133}Xe > 0.5$

- This leaves only 3 IMS sites and 3 background campaign sites

SITE	Sum of Ratio>0.5	Sum of binary>0.5	Sum of DA_XE135
Mutsu, Japan	108.15	52	11.54
Jakarta, Indonesia	39.08	34	73.18
Charlottesville, VA, USA	37.71	18	10.35
Spitsbergen, Norway	27.68	13	2.87
Stockholm, Sweden	26.94	24	30.58
Horonobe, Hokkaido, Japan	20.46	9	1.76
Wake Island, USA	10.91	3	0.53
Melbourne, VIC	9.97	4	2.06
Darwin, NT, Australia	8.06	2	0.38
Rio de Janeiro, Brazil	6.01	2	0.48
Takasaki, Gunma, Japan.	5.73	3	0.75
Tristan da Cunha, UK	5.31	2	0.31
Oahu, HI, USA	3.49	1	0.19
Ashland, KS, USA	3.37	1	0.28

Overview

^{135}Xe/^{133}Xe isotopic activity ratios observed in the atmosphere

- IMS noble gas systems
- Background campaign in Japan

^{135}Xe/^{133}Xe isotopic activity ratios of various sources

- nuclear explosions
- nuclear facilities

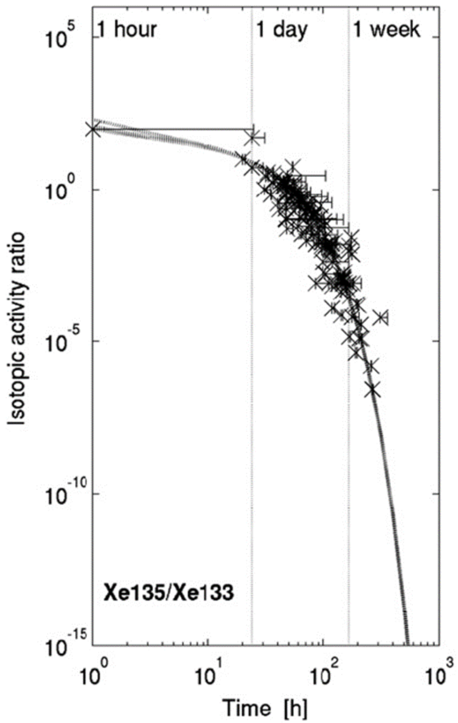
Comparison

- Comparison of IMS observations with various sources
- Usefulness of the ^{135}Xe/^{133}Xe ratio for nuclear explosion monitoring

Nuclear Explosions

- Nuclear explosions have very high $^{135}Xe/^{133}Xe$ activity ratio:

- Initially >1000
- After 1 day ~ 10
- After 2 days ~ 1

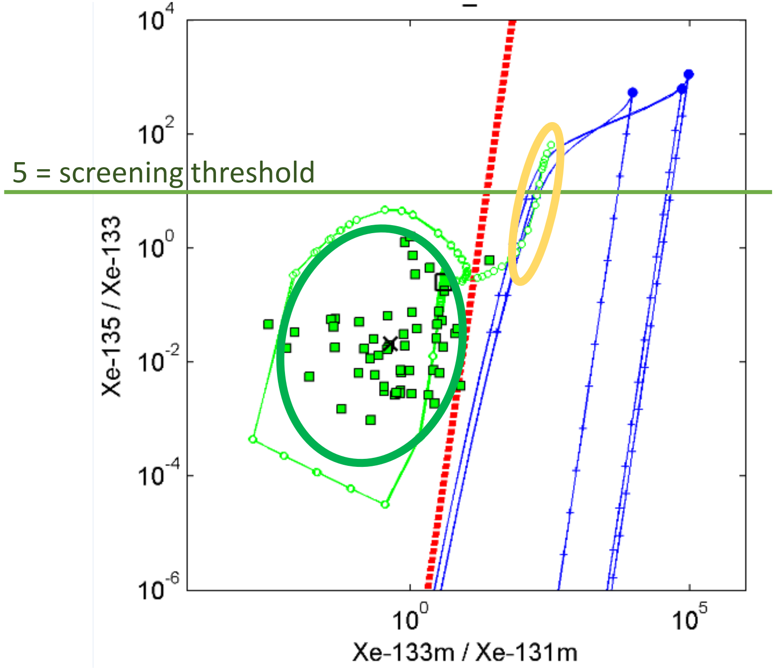


Kalinowski/Pistner (2006)
Solid curves: simulation of nuclear tests
Crosses: Releases at Nevada Test Site

Nuclear power plants (NPPs) at steady power

- Nuclear power plants have isotopic ratio of Xe-135 to Xe-133 from fission that are typically well below 5, i.e. below the screening threshold for the nuclear explosion domain (see green horizontal line).

- Medical isotope production is expected to cause observations in the nuclear explosion domain.



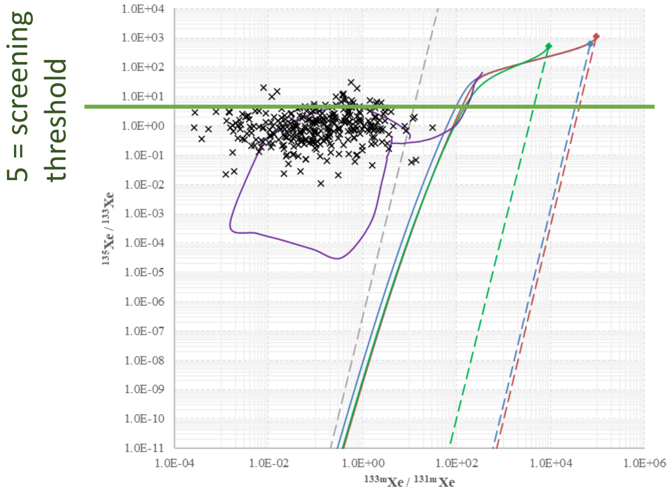
Normal observations from fission sources have Xe-135/Xe-133 activity ratios well below the screening threshold of 5.

Medical isotope production facilities

- The surprise was to find observations from medical isotope production in the nuclear reactor domain.

- Observations at Jakarta, Indonesia, 14 km from BaTek
  Very high of $^{135}Xe/^{133}Xe$ activity ratios

- Confirmed by STAX stack release data.

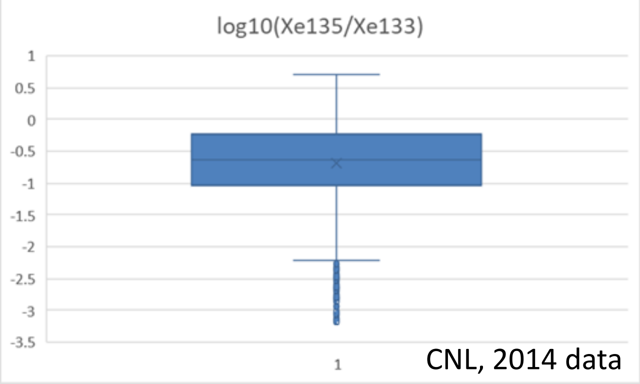


Jakarta

Medical isotope production facilities

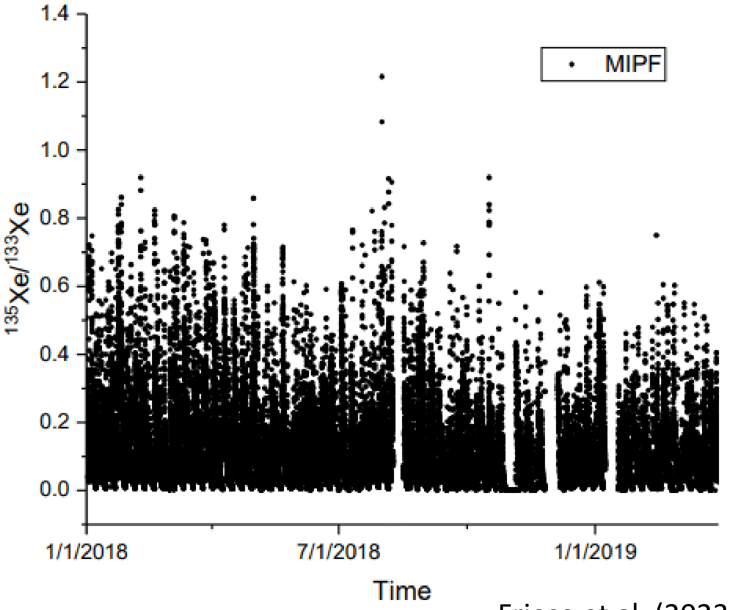
Stack release data:

- Very rarely $^{135}Xe/^{133}Xe$ activity ratios >1
- Applies to ANSTO, CNL, and IRE due to release delay



CNL, 2014 data

5 = screening threshold

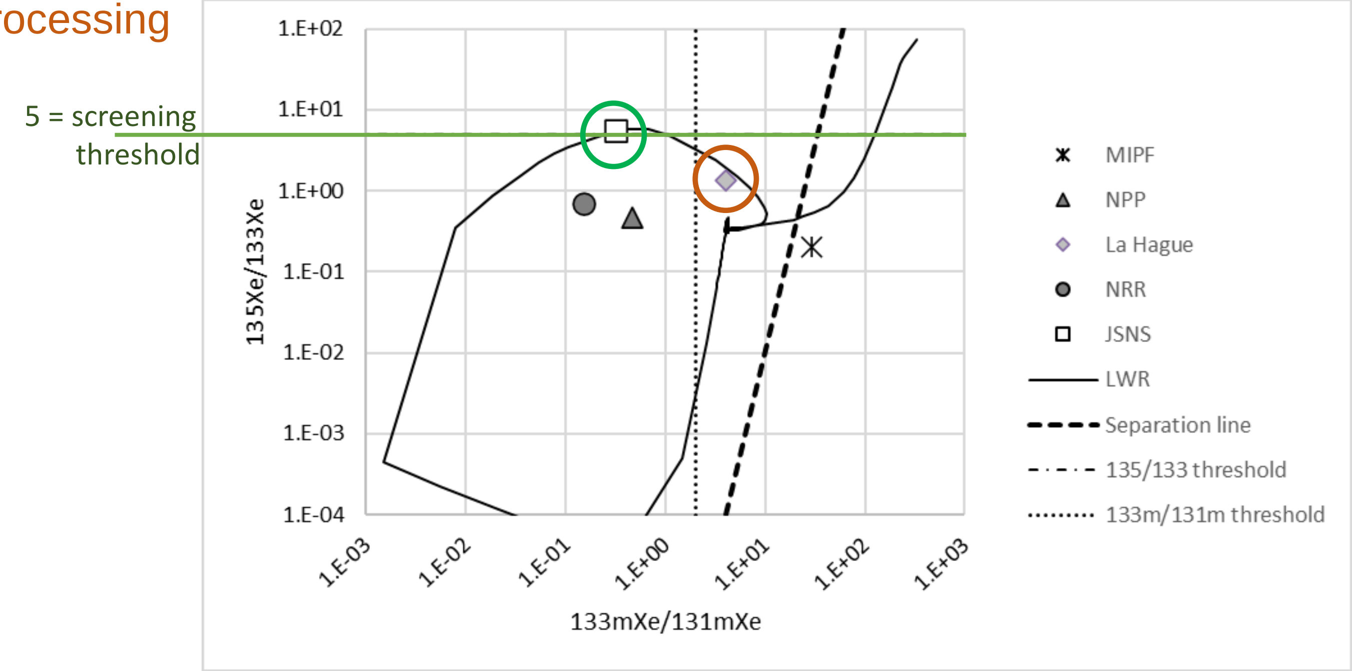


Friese et al. (2023)

Comparing different nuclear facilities

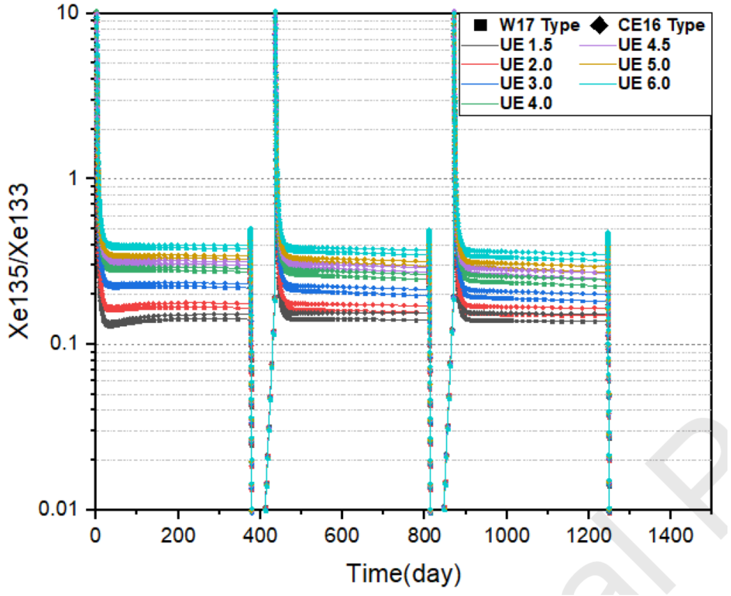
Activation sources (HFIR, Spallation Neutron Sources)

Spent nuclear fuel reprocessing



(see presentation by Tatiana Boitsova)

$^{135}Xe/^{133}Xe$ activity ratio as a function of uranium enrichment



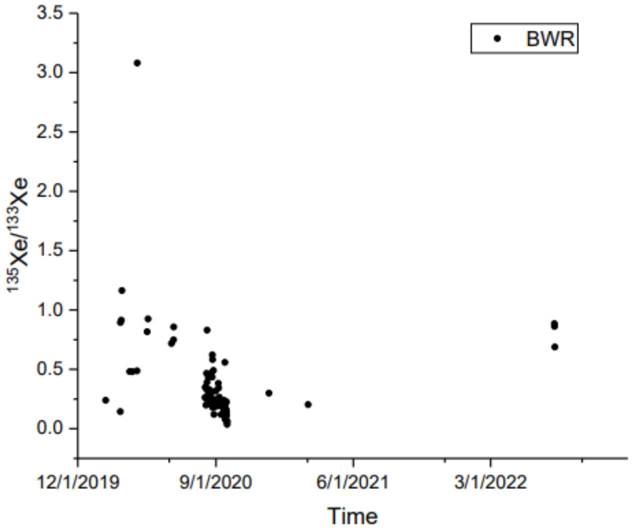
Ser Gi Hong, Geon Hee Park, Sang Woo Kim, Yu Yeon Cho (2023). An extensive characterization of xenon isotopic activity ratios from nuclear explosion and nuclear reactors in neighboring countries of South Korea. Nuclear Engineering and Technology, NET 2646.

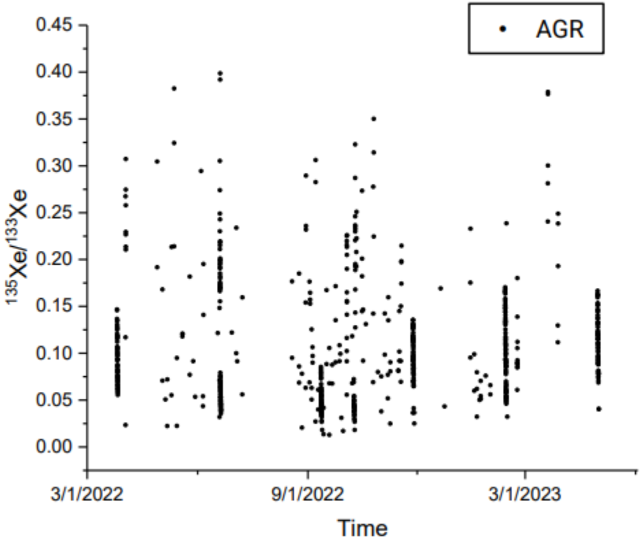
- The $^{135}Xe/^{133}Xe$ isotopic activity ratios get higher for PWR with increased uranium enrichment as shown by Hong et al. (2023) for enrichments ranging from 1.5% to 6.0%.
- It needs to be further studied what the activity of radioxenon isotopes is from HEU (high enriched uranium) fuel.

NPP release measurements

- Only few empirical studies are available on reactor power ramping up or down.
- STAX data for a BWR show rarely $^{135}Xe/^{133}Xe$ activity ratios >1.

(Never for AGR)

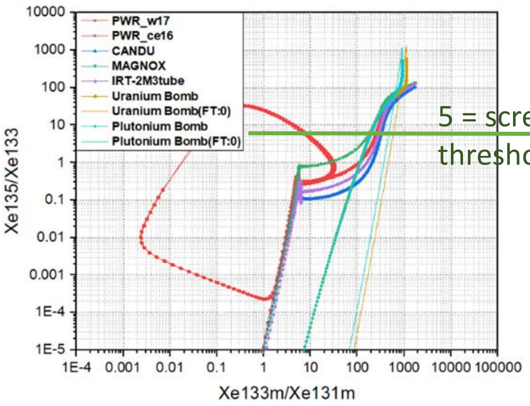




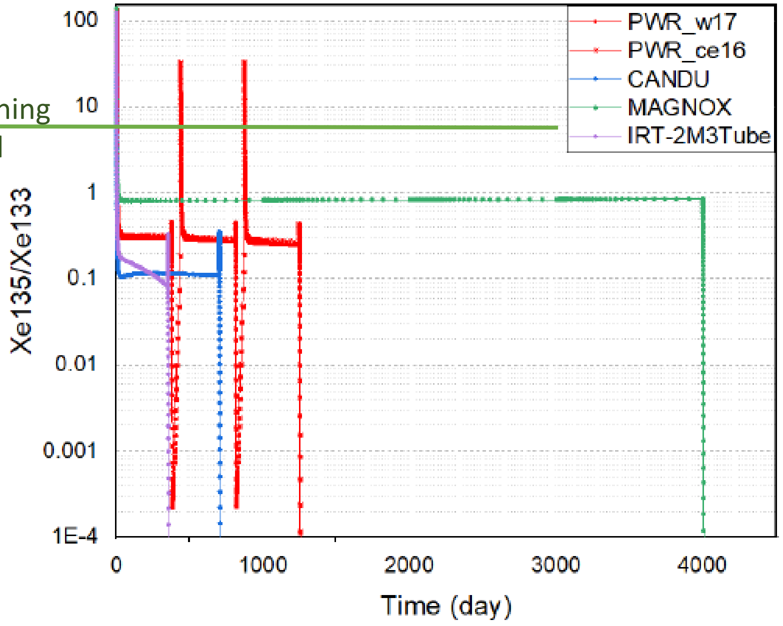
Friese et al. (2023)

High $^{135}Xe/^{133}Xe$ activity ratio at NPP shutdown and startup

- Hong et al. (2023) compare different reactor types.



- Effect only for PWR (neither CANDU, nor MAGNOX, nor IRT)



- After shutdown (end of irradiation), ^{135}Xe increases due to the very large absorption cross section. $0.5 > {}^{135}Xe/^{133}Xe >$ saturation level of ~0.3 for PWR.
- During start-up, the ratio increases (even higher) because buildup of ^{135}Xe is quicker than ^{133}Xe.

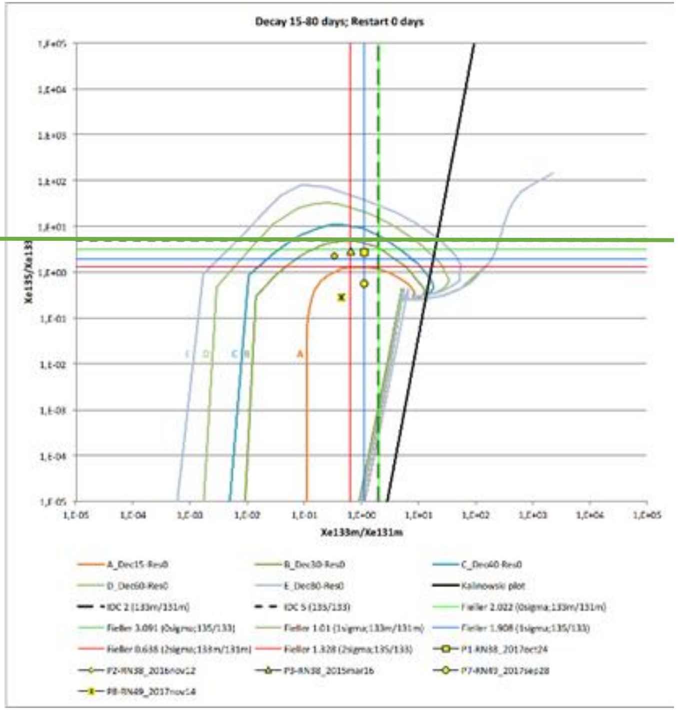
Ser Gi Hong, Geon Hee Park, Sang Woo Kim, Yu Yeon Cho (2023). An extensive characterization of xenon isotopic activity ratios from nuclear explosion and nuclear reactors in neighboring countries of South Korea. Nucl Eng & Techn, NET 2646.

High $^{135}Xe/^{133}Xe$ activity ratio at NPP shutdown and startup

CTBTO PREPARATORY COMMISSION | COMPREHENSIVE NUCLEAR-TEST-BAN TREATY ORGANIZATION

- Ottaviano et al., (2017) did simulations of LWR trajectories with varying duration of shutdown prior to the restart of the reactor.

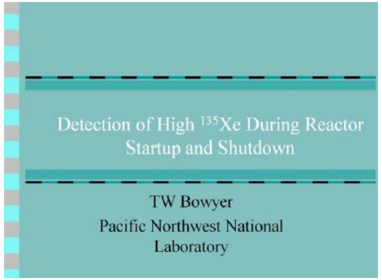
5 = screening threshold



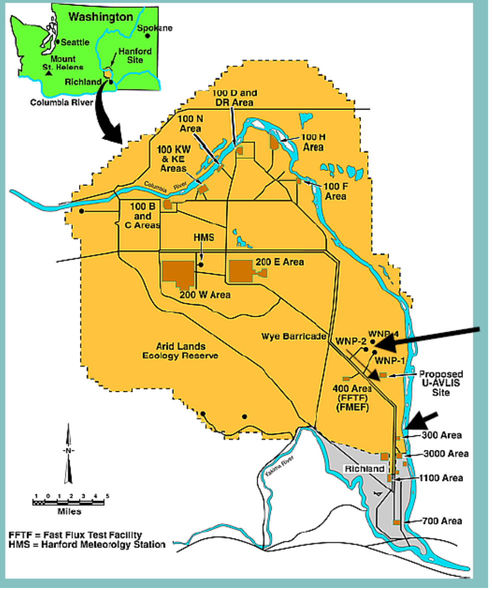
- For 30 days: $^{135}Xe/^{133}Xe$ = 5
- For 80 days: $^{135}Xe/^{133}Xe$ =100

Early hint on NPPs by PNNL

- Elevated levels of the $^{135}Xe/^{133}Xe$ activity ratio are observed during reactor startup and shutdown.
- This was reported first at INGE Workshop 2001 by Ted Bowyer.



- In late July, the Columbia Generating Station WNP-2 BWR nuclear reactor performed a shutdown
- Approximately 7 miles away ^{135}Xe was detected at low levels correlated with the shutdown
- Wind direction toward our sampling location



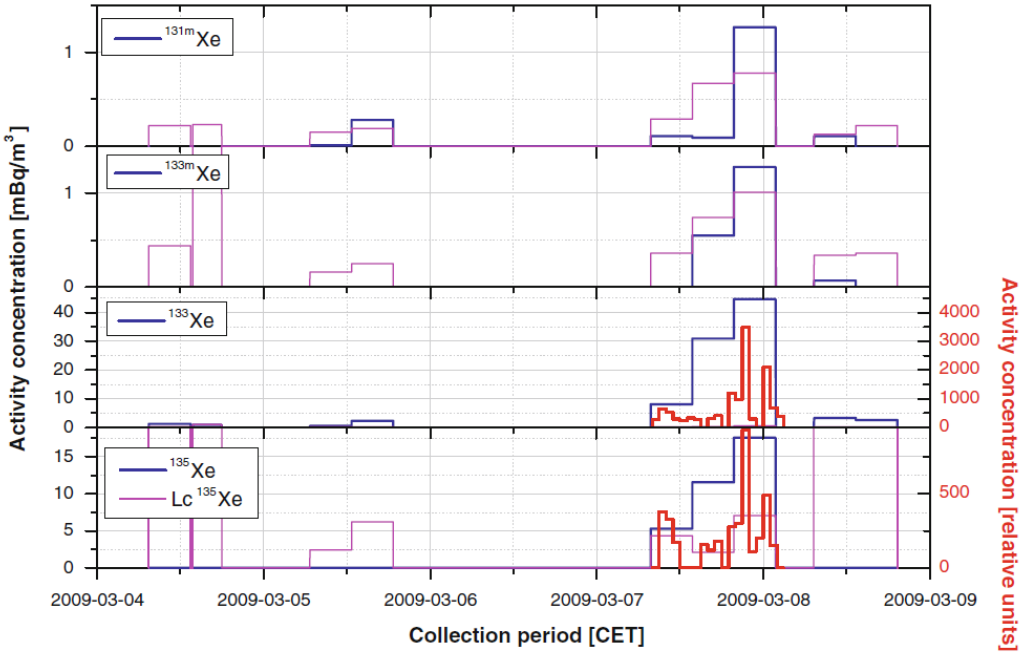
ARSA Measurement
Xe-135 = 3.5 mBq/m³
Xe-133 = 0.6 mBq/m³
Ratio = 5.8

Study by CTBTO, FOI and PNNL

- Saey et al. (2013) show stack measurements of $^{135}Xe/^{133}Xe$ activity ratio during the shut-down and revision of the Isar-I NPP.

- Stack measurements and atmospheric air at 5 km distance.

- Always $^{135}Xe/^{133}Xe < 0.63$



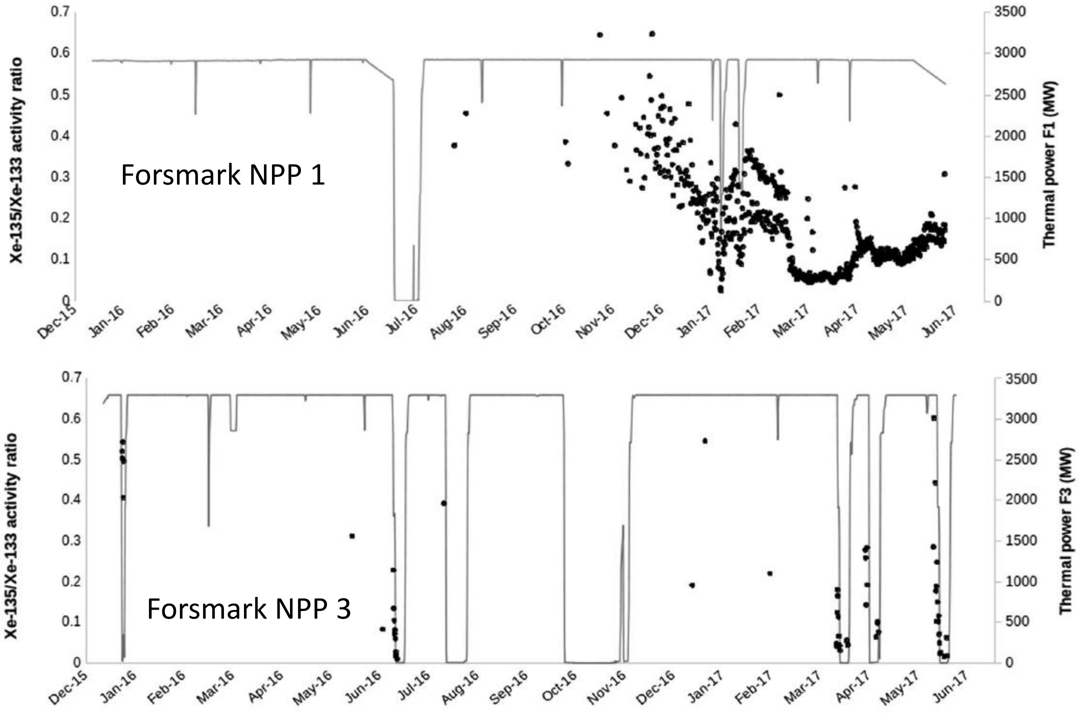
Isar-I NPP

Study by FOI

- Ringbom et al. (2021) show stack measurements of $^{135}Xe/^{133}Xe$ activity ratio during the operation of the Forsmark NPP.

- The shutdown operations caused activity increases; the startup gave no signal.
- The MDA for ^{135}Xe: 0.6 GBq (^{133}Xe: 2 GBq) per 6-h period.

- Always $^{135}Xe/^{133}Xe < 0.7$



Overview of possible sources

Source	Max. $^{135}Xe/^{133}Xe$ act. ratio	High end ^{135}Xe activity per day
Nuclear power plants (NPPs) at steady power	0.3	5 GBq
Medical isotope production facilities, e.g. PT Inuki Others (ANSTO, CNL, IRE)	1 <10 <1	10 GBq 1,000 GBq
Highly pure ^{135}Xe generator	> 200	0.001 GBq
Neutron activation in spallation neutron sources or at NRRs, example HFIR	5.6 1 5.6	0.02 GBq 3 GBq
Spent Fuel reprocessing facilities	1.3	0.28 GBq
Nuclear research reactors (NRRs) using HEU	>1 ?	From 1 Bq to 0.6 GBq
LWRs start-up and shut-down 152	>30 0.7 (Forsmark) 5.8 (WNP)	<0.6 GBq per 6-h period >0.6 GBq per 6-h period

Overview

$^{135}Xe/^{133}Xe$ isotopic activity ratios observed in the atmosphere

- IMS noble gas systems
- Background campaign in Japan

$^{135}Xe/^{133}Xe$ isotopic activity ratios of various sources

- nuclear explosions
- nuclear facilities

Comparison

- Comparison of IMS observations with various sources
- Usefulness of the $^{135}Xe/^{133}Xe$ ratio for nuclear explosion monitoring

Strength of a ^{135}Xe source depends on distance

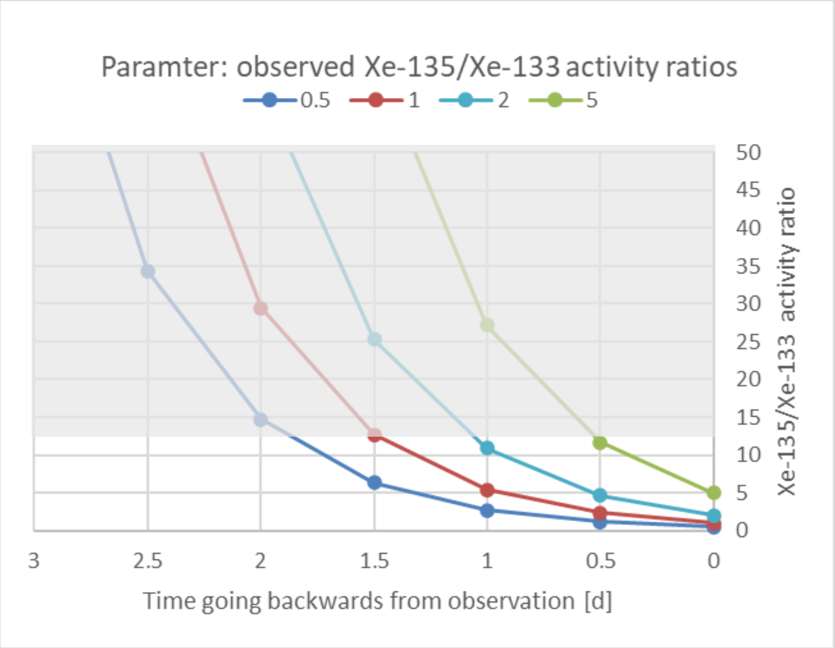
- Looking backwards from time of observation

- Strength of a ^{135}Xe source depends on atmospheric transport time, i.e. distance.
- Minimum release to be seen at closest IMS site: 5 GBq/d (Bowyer et al., 2014)

- Example:

If observation ^{135}Xe/^{133}Xe = 1 and the delay is 1.5 d, the source has ^{135}Xe/^{133}Xe = 13.

➢ ^{135}Xe sources must be in close vicinity of detector (1 to 2 days of atmospheric transport)



High ^{135}Xe/^{133}Xe ratio from Research reactors using HEU?

HEU reactors worldwide:

- Bubble size for enrichment level
- display filter set for release rate estimate of ^{135}Xe > 0.1 GBq/a

CTBTO PREPARATORY COMMISSION

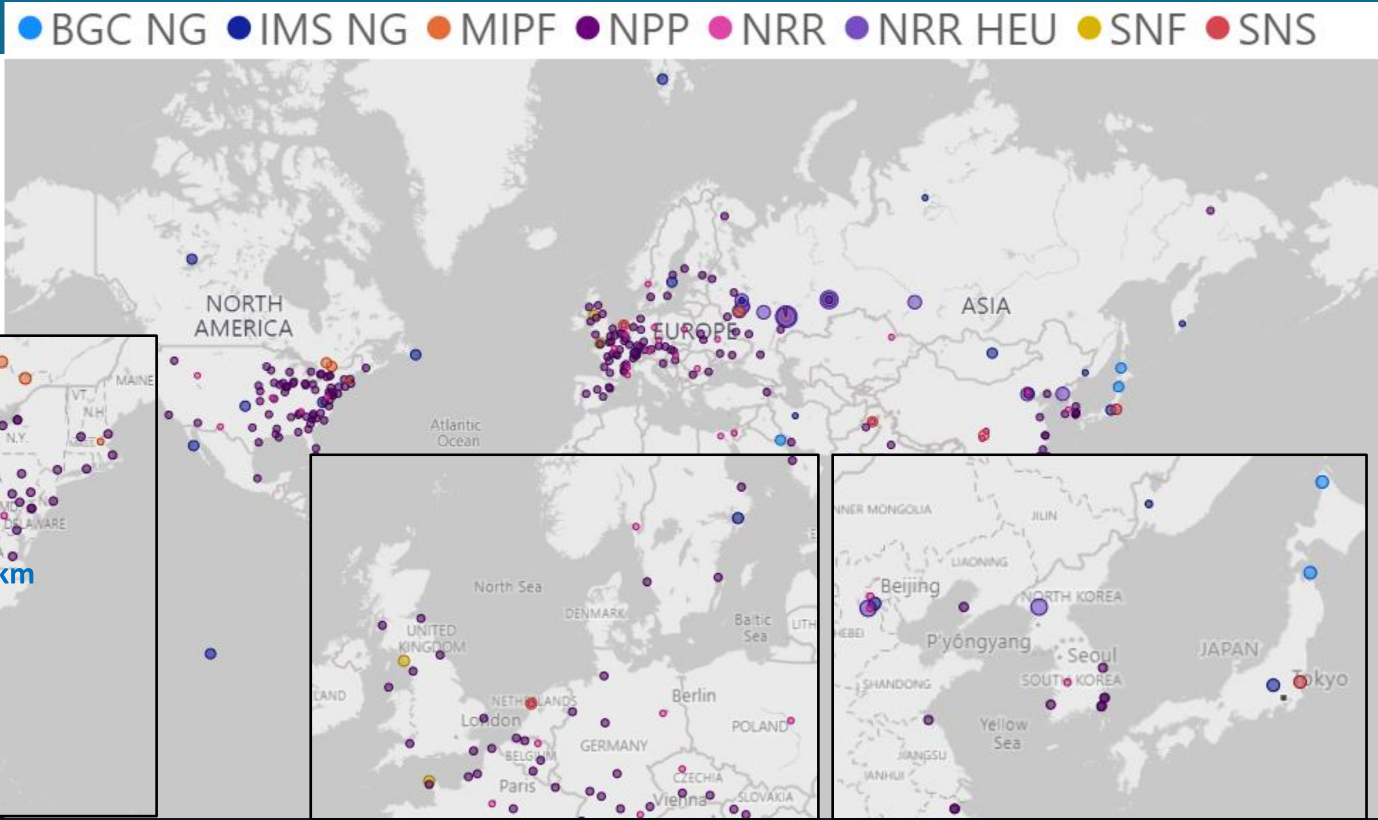
COMPREHENSIVE NUCLEAR-TEST-BAN TREATY ORGANIZATION

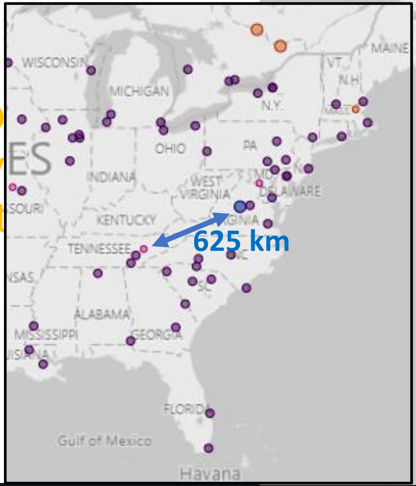
To be updated

Xe-135 sources in the close vicinity of IMS noble gas systems

Possible sources of high $^{135}Xe/^{133}Xe$ ratios

- What are possible sources?
- How far these so can they observat





To be updated

Location of Xe-135 sources (LWRs)

Interactive map

Summary

Research question:

- We study an issue that is open for long: what is a possible source of pure ^{135}Xe and high $^{135}Xe/^{133}Xe$ isotopic activity ratio observations? *Note: it could indicate a nuclear test!*

Method:

- We take a comprehensive look at all IMS NG locations and background campaigns.
- We scrutinize every conceivable source of high $^{135}Xe/^{133}Xe$ ratios.

Conclusions:

- There is not a unique explanation for all occurrences of elevated $^{135}Xe/^{133}Xe$ ratios and for most locations the sources are still not clear.
- In view of the requirement for the source being at close distance (<2 days transport), we need to confess that there are observations of elevated $^{135}Xe/^{133}Xe$ ratios at some locations which are too far away from nuclear installations for any convincing explanation.
- Investigate further, especially LWR start-up and activation sources, possibly HEU reactors.

References

- Bowyer, T. W., Abel, K. H., & Hensley, W. K. (1996). Automatic radioxenon analyzer for CTBT monitoring (No. PNNL-11424). Pacific Northwest National Lab.(PNNL), Richland, WA (United States).
- Bowyer, T. W., Eslinger, P. W., Cameron, I. M., Friese, J. I., Hayes, J. C., Metz, L. A., & Miley, H. S. (2014). Potential impact of releases from a new Molybdenum-99 production facility on regional measurements of airborne xenon isotopes. Journal of environmental radioactivity, 129, 43-47.
- Lars-Erik De Geer (2012): Radionuclide Evidence for Low-Yield Nuclear Testing in North Korea in April/May 2010, Science & Global Security: The Technical Basis for Arms Control, Disarmament, and Nonproliferation Initiatives, 20:1, 1-29
- Eslinger et al.: Determining the source of unusual xenon isotopes in samples. Journal of Environmental Radioactivity (2022) 247:106853.
- Hong, S. G., Park, G. H., Kim, S. W., & Cho, Y. Y. (2023). An extensive characterization of xenon isotopic activity ratios from nuclear explosion and nuclear reactors in neighboring countries of South Korea. Nuclear Engineering and Technology.
- Kalinowski, M. B., & Pistner, C. (2006). Isotopic signature of atmospheric xenon released from light water reactors. Journal of environmental radioactivity, 88(3), 215-235.
- Kalinowski, M.B.: Global emission inventory of 131mXe, 133Xe, 133mXe, and 135Xe from all kinds of nuclear facilities for the reference year 2014. Journal of Environmental Radioactivity (2023) 261:107121
- Klingberg et al.: 127Xe coincidence decay analysis in support of CTBT verification. Journal of Radioanalytical Nuclear Chemistry (2015) 305:225-232.
- Ringbom, A., Axelsson, A., Björnham, O., Brännström, N., Fritioff, T., Grahn, H., ... & Olsson, M. (2021). Radioxenon releases from a nuclear power plant: Stack data and atmospheric measurements. Pure and Applied Geophysics, 178, 2677-2693.
- Saey, P.R.J., Ringbom, A., Bowyer, T.W. et al. Worldwide measurements of radioxenon background near isotope production facilities, a nuclear power plant and at remote sites: the “EU/JA-II” Project. J Radioanal Nucl Chem 296, 1133–1142 (2013). https://doi.org/10.1007/s10967-012-2025-2

CTBTO

PREPARATORY COMMISSION

Vienna International Centre, PO Box 1200
1400 Vienna, Austria
CTBTO.ORG

Overview of possible sources

- Nuclear power plants (NPPs) at steady power
- Medical isotope production facilities (MIPFs)
- Highly pure ^{135}Xe generator with ^{252}Cf or by activating ^{134}Xe or ^{136}Xe
- Neutron activation in spallation neutron sources or NRRs
- Spent Fuel reprocessing facilities
- Nuclear research reactors (NRRs) using HEU
- NPPs start-up and shut-down

Highly pure ^{135}Xe generators

Highly pure ^{135}Xe generator

Source	$^{135}Xe/^{133}Xe$ activity ratios >1	High end ^{135}Xe activity per day
from ^{252}Cf spontaneous fission	9.3 1.4	2.8 kBq 25 kBq
by activating ^{134}Xe or ^{136}Xe	> 200	1 MBq

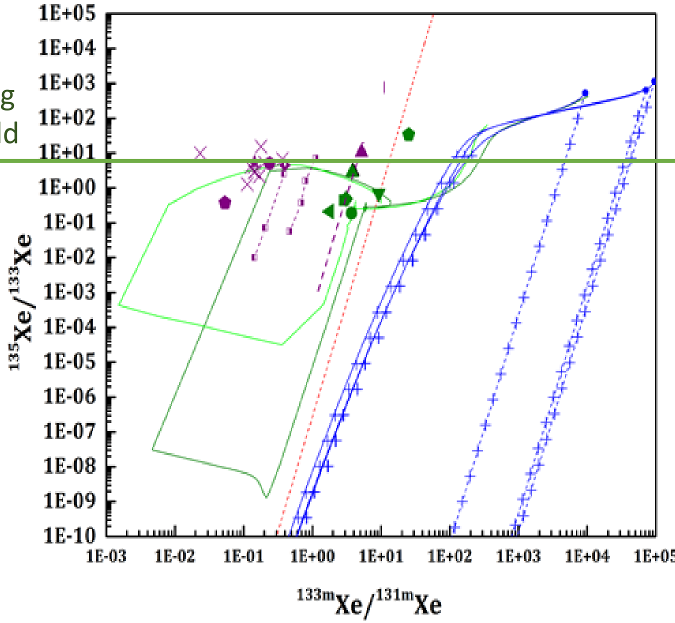
Neutron activation in spallation neutron sources or NRRs

- The isotopic ratio of Xe-135 to Xe-133 from activation is typically very high and unusually high activity ratios may be >5, i.e. above the screening threshold for the nuclear explosion domain (see green horizontal line).

- Strong neutron sources may generate significant amounts of radioxenon by activation of stable xenon.

- A few specific research reactors (e.g. HFIR) and spallation neutron sources are dominated by neutron activation as the production process.

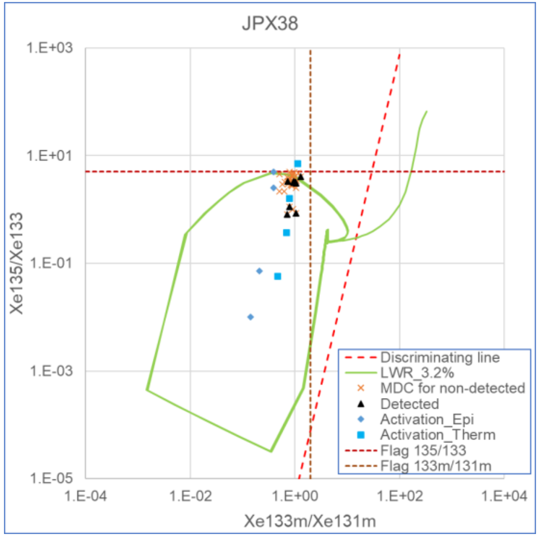
5 = screening threshold

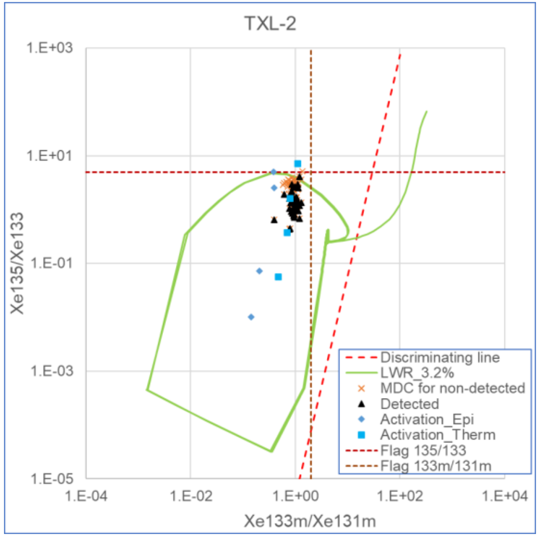


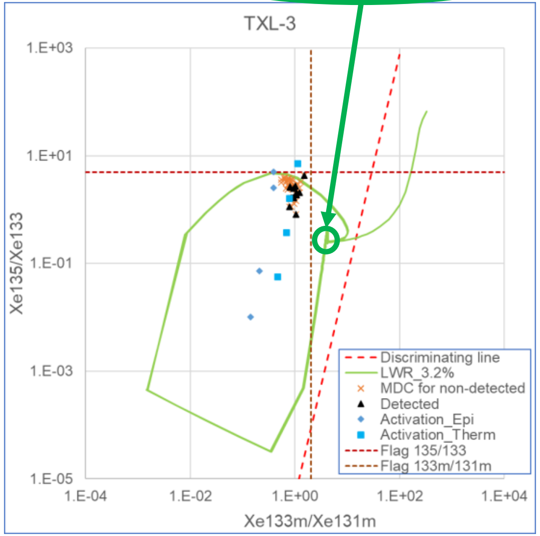
Isotopic ratio analysis for NG systems in Japan

Four xenon relationships

- Plots of $^{133m}Xe/^{131m}Xe$ and $^{135}Xe/^{133}Xe$ for observations at JPX38, TXL-2 and TXL-3.
- MDCs are used for non-detections of Xe-133 and Xe-131m.
- Ratios are close to activation signatures, not in the vicinity of nuclear fission equilibrium.







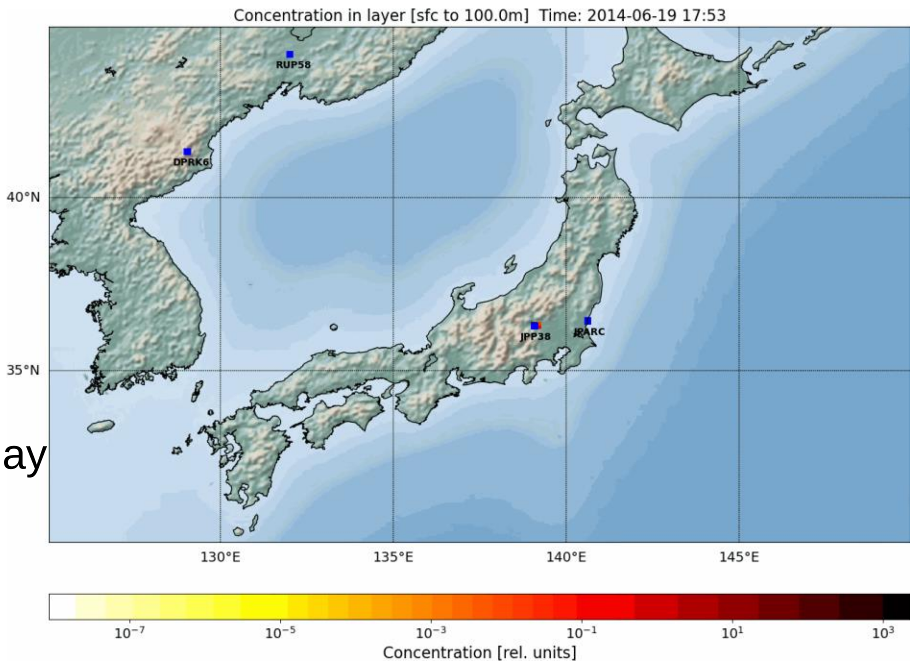
Spallation neutron sources

- The isotopic ratios give evidence that an activation source might be the source.
- A case study shows that the spallation neutron source MLF at J-PARC, Japan, could possibly explain some but not all observations of ^{135}Xe at JPX38 (presentation P2.1-607 at SnT2021).
- Similar evidence was found regarding the spallation neutron source at Oak Ridge, though its contribution cannot be clearly distinguished from the HFIR, another strong activation source (Ely et al, O2.4-138 at SnT2021, Eslinger et al., 2022)
- The possibility of non-traditional radioxenon isotopes from activation being contained in IMS samples at levels that interfere with NG analysis requires further investigation and enhancement of analysis software.

Spallation neutron sources

Estimation of the annual emission of ^{133}Xe and ^{135}Xe

- Based on quarterly reports for ^{41}Ar releases published by J-PARC
- Estimation of radioxenon with ^{41}Ar as proxy, HFIR data for scaling
- The daily release of ^{133}Xe was estimated by Kalinowski (2023) to be around 0.01 GBq.
- The source reconstruction indicated the upper value of hypothetical releases to be ~6 GBq/day for ^{133}Xe, i.e. much higher.

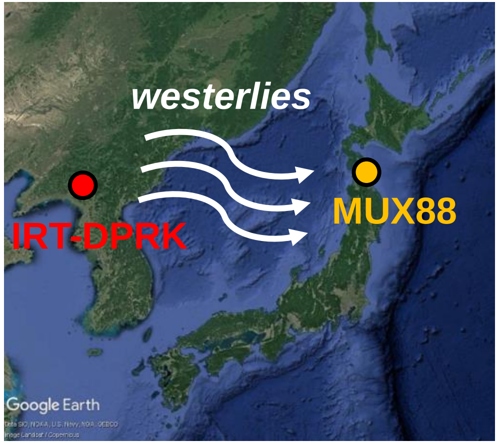


High $^{135}Xe/^{133}Xe$ ratio from Research reactors using HEU?

- There is a nuclear research reactor using high enriched uranium (HEU), IRT-DPRK, in Yongbyon, Democratic People's Republic of Korea (DPRK). As of 2015, IRT-DPRK uses 36% HEU fuel.

Technical specifications of IRT-DPRK as of 2015 *1

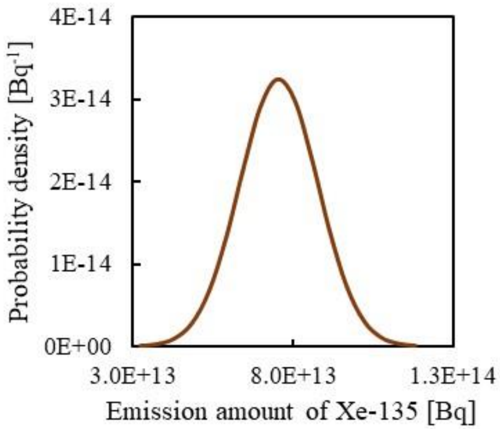
Power level	8 MWt
Thermal neutron flux	10^{13} - 10^{14} n/cm^2/s
Irradiated nuclear fuel	36% HEU



*1 See Lee et al., 2015. Preliminary Analysis on the Management Options of IRT-DPRK Research Reactor. Transactions of the Korean Nuclear Society Spring Meeting, Jeju, Korea, May 7-8, 2015.

High $^{135}Xe/^{133}Xe$ ratio from Research reactors using HEU?

Estimated Xe-135 emission from IPR-DPRK on 16 October 2014



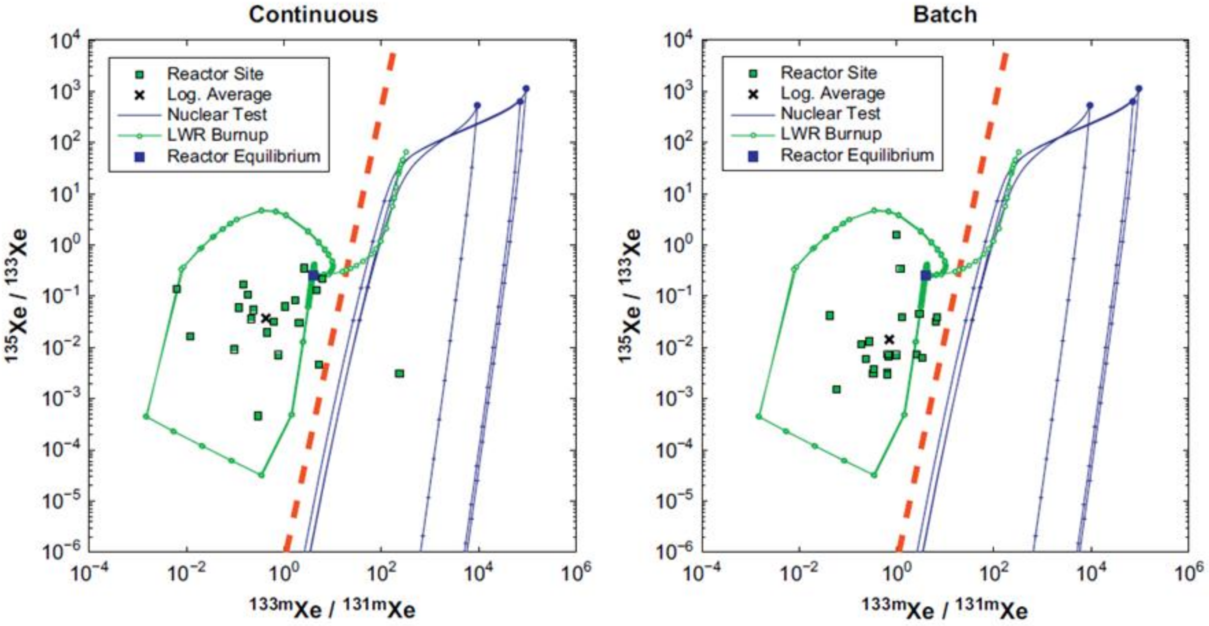
- Highest activity concentration of Xe-135 at MUX88 (in Mutsu, Japan) in 2014 is 1.41 mBq/m^3 at 9am on 19 October 2014.
- Although there is no detailed information on operational status of IRT-DPRK, when we assume that this emission source is IRT-DPRK, the estimated Xe-135 emission amount from IRT-DPRK on 16 October 2014 using ATM is 5.1E13 – 1.0E14 Bq with 95% confidence interval.
- Much higher than estimated release: 0.02 GBq in 24 h.

NPP annual release reports

- In general, batch releases have lower levels of the $^{135}Xe/^{133}Xe$ activity ratio compared to continuous releases.

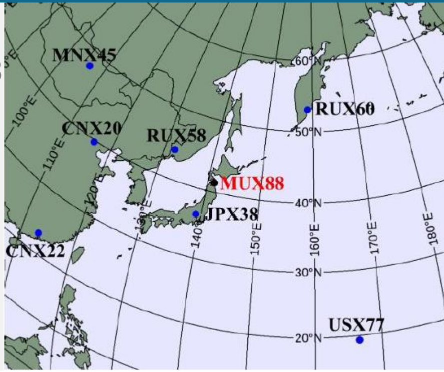
- But the highest ratios are reported from occasional batch releases.

M.B. Kalinowski, M.P. Tuma / Journal of Environmental Radioactivity 100 (2009) 58–70

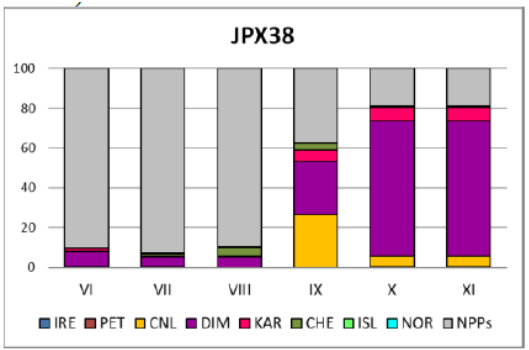


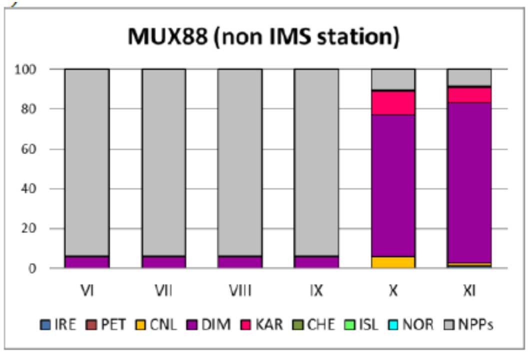
Impact of NPPs on JPX38 and MUX88

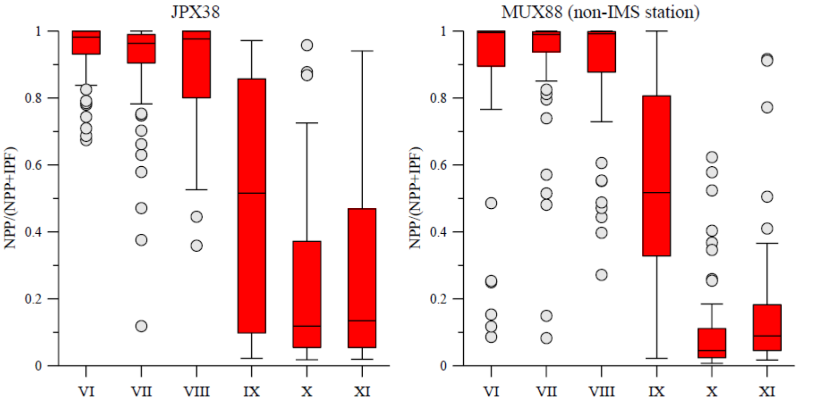
The distance between JPX38 and MUX88 is about 584 km, relatively short in comparison to distances between IMS stations.



Monthly percentage contributions during the study period of 2014 from individual sources as observed at the IMS station at Takasaki, JPX38, and the transportable system in Mutsu, MUX88.







Monthly changes in the ratio between the contribution from NPPs and the joined contribution from all known NPPs and IPFs.

Kuśmierczyk-Michulec, J., Baré, J., Kalinowski, M., Tipka, A. (2022). Characterisation of Xe133 background at the IMS stations in the East Asian region: insights based on known sources and Atmospheric Transport Modelling. Journal of Environmental Radioactivity 255. DOI: 10.1016/j.jenvrad.2022.107033