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Radioxenon isotopic ratios from activation of stable xenon in releases from nuclear facilities in relation to fission sources visualized in multi-isotope-ratio plots

O3.5 - 456

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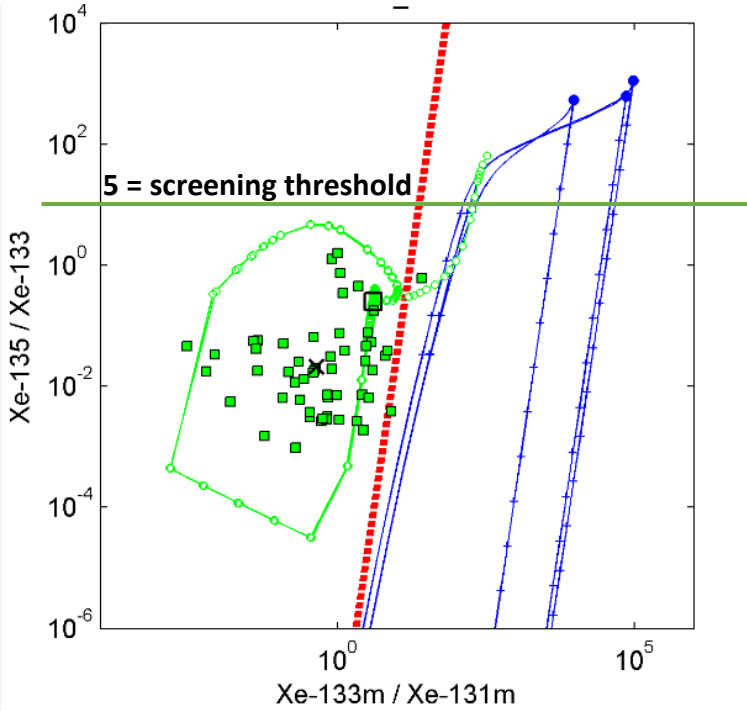
ORAL PRESENTATION

Radioxenon isotopic ratios from activation of stable xenon in releases from nuclear facilities in relation to fission sources visualized in multi-isotope-ratio plots

Motivation

- Radioxenon emission from neutron activation sources have extremely high Xe-135/Xe-133 ratios and may cross the screening threshold.
- If non-traditional radioxenon isotopes appear in IMS samples, they will interfere with the analysis.

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



Current knowledge

Pioneering studies have identified the relevance of non-traditional radioxenon isotopes (from activation of stable xenon) for IMS NG systems. This study takes it a step further.

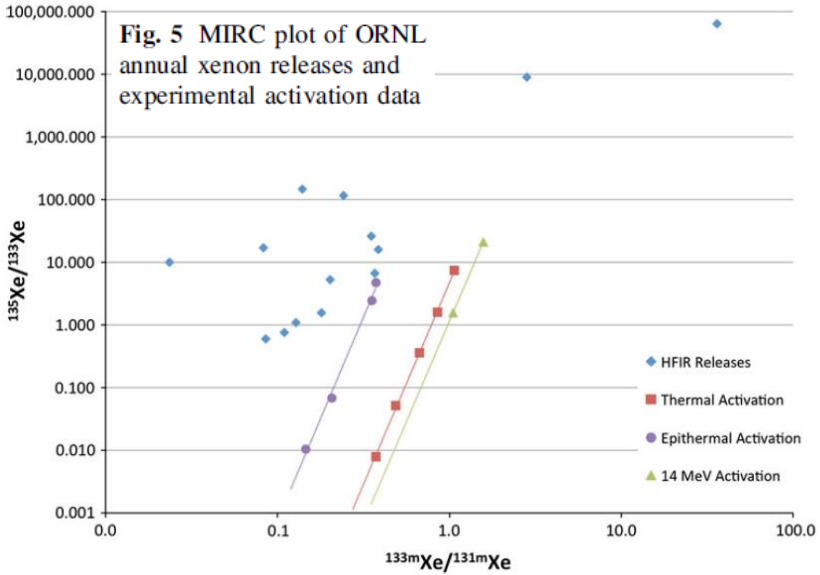


Fig. 5 MIRC plot of ORNL annual xenon releases and experimental activation data

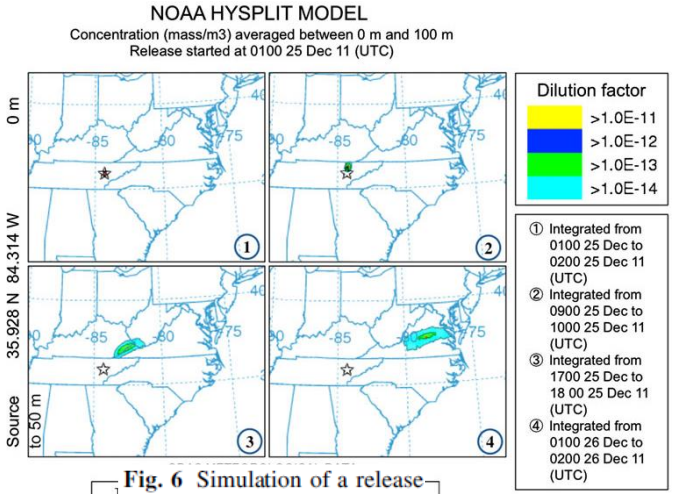


Fig. 6 Simulation of a release from ORNL HFIR and the plume dispersion to an IMS station

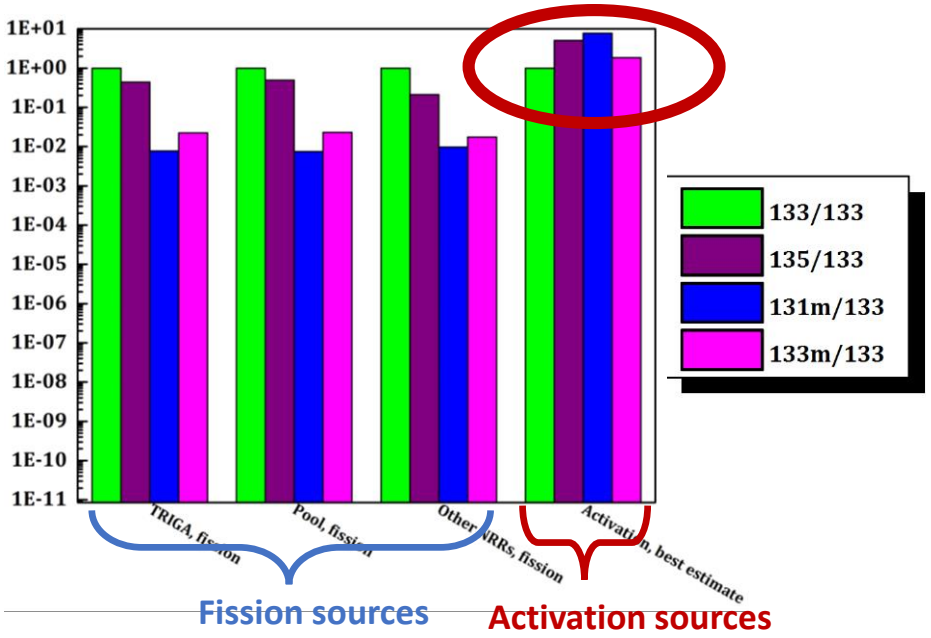
Charlottesville, VA, (USX75)

Figures from F. J. Klingberg, et al. J Radioanal Nucl Chem (2013) 296:117-123

Difference in radioxenon isotopic ratios from fission and from activation

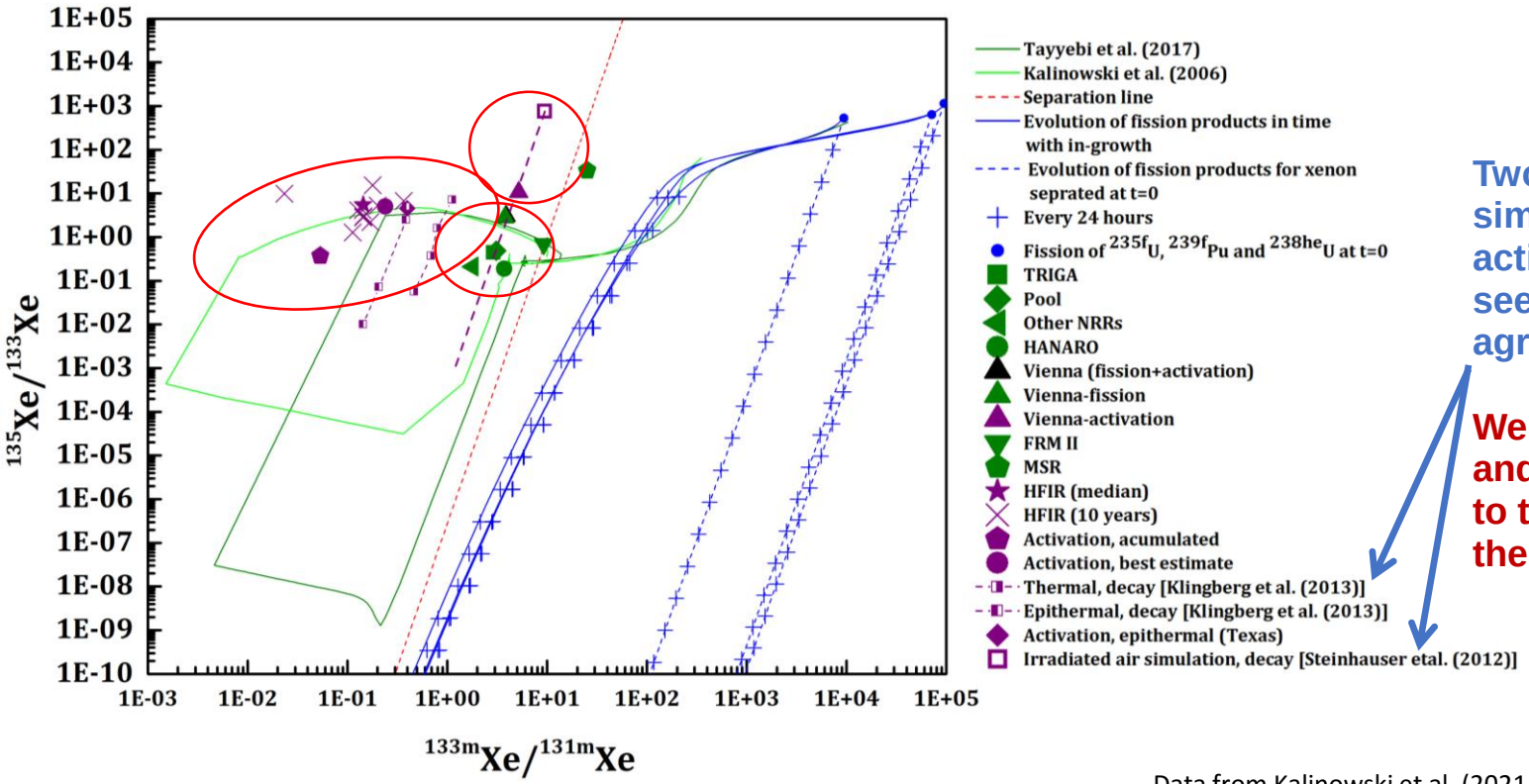
Activation products of stable xenon have also relevance for characterizing the traditional radioxenon isotopes:

- In case of fission: Xe-133 is the lead nuclide
- For activation: Xe-133 has the lowest activity, Xe-131m the highest, followed by Xe-135



Data from Kalinowski et al. (2021)

4-isotope plot shows:
- fission cluster around the equilibrium point
- two distinct activation clusters



Two previous simulations of activation products seem not to be in agreement.

We apply ORIGEN and vary parameters to try to consolidate them.

Data from Kalinowski et al. (2021)

4-isotope plot shows:
- fission cluster around the equilibrium point
- two distinct activation clusters

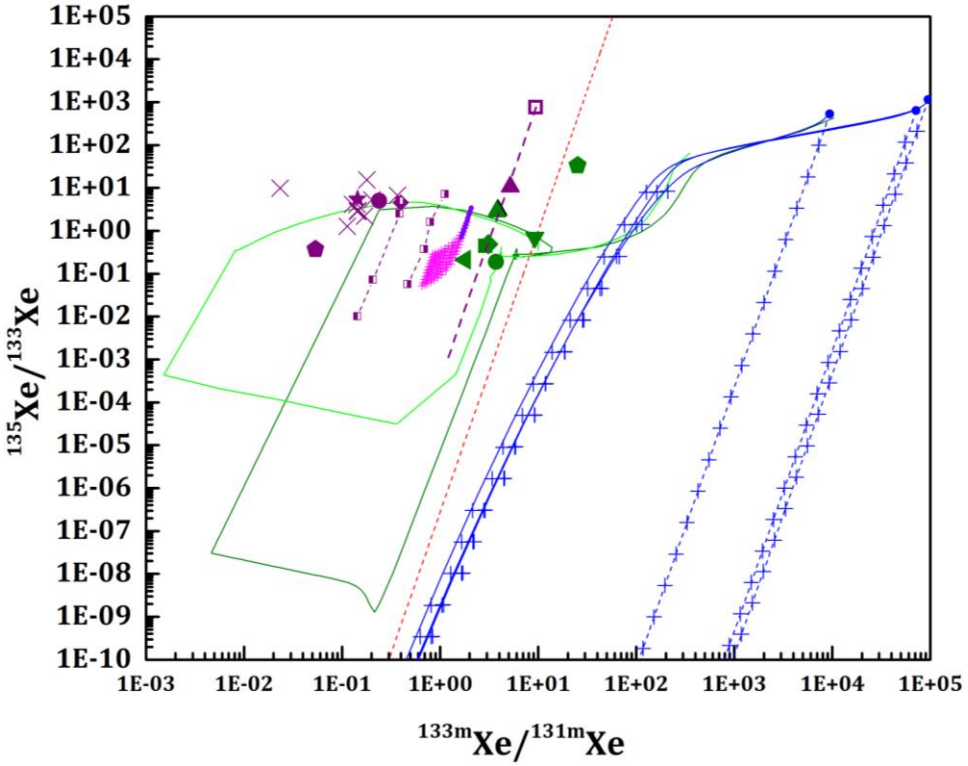
Two previous simulations of activation products apply to the Texas TRIGA and Vienna TRIGA. The table shows their parameters and annual release estimates based on measurements.

We apply ORIGEN and vary parameters to try to shed more light into radioxenon from activation.

	Texas TRIGA Mark II	Vienna TRIGA Mark II
Thermal power (kW)	950	250
Capacity factor (%)	15.4	19.2
Neutron flux ($n/cm^2.s$)	2.70E+13	1.00E+13
^{131m}Xe (Bq/y)	3.10E+07	2.30E+03
^{133}Xe (Bq/y)	1.40E+07	1.81E+05
^{133m}Xe (Bq/y)	1.23E+07	8.93E+03
^{135}Xe (Bq/y)	6.39E+07	5.49E+05

Data from Kalinowski et al. (2021)

4-isotope plot shows:
- fission cluster around the equilibrium point
- two distinct activation clusters



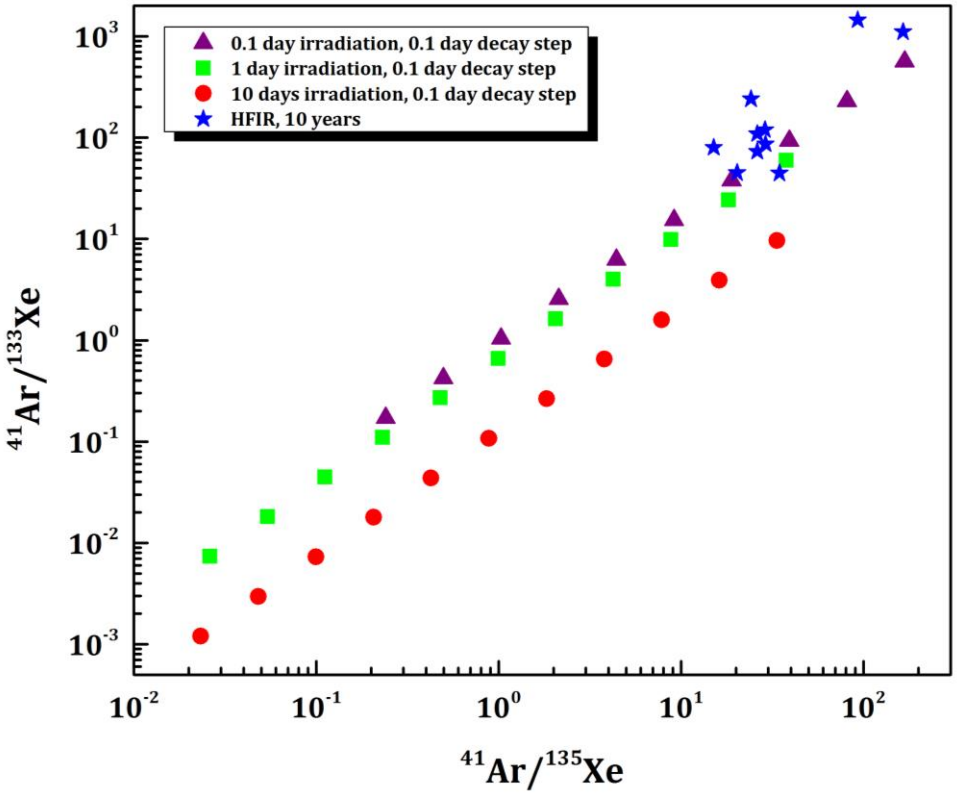
- Tayyebi et al. (2017)
- Kalinowski et al. (2006)
- Separation line
- Evolution of fission products in time with in-growth
- Evolution of fission products for xenon seprated at t=0
- Every 24 hours
- Fission of ^{235f}U, ^{239f}Pu and $^{238he}U$ at t=0
- TRIGA
- Pool
- Other NRRs
- HANARO
- Vienna (fission+activation)
- Vienna-fission
- Vienna-activation
- FRM II
- MSR
- HFIR (median)
- HFIR (10 years)
- Activation, acumulated
- Activation, best estimate
- Thermal, decay [Klingberg et al. (2013)]
- Epithermal, decay [Klingberg et al. (2013)]
- Activation, epithermal (Texas)
- Irradiated air simulation, decay [Steinhauser etal. (2012)]
- ORIGEN, Thermal
- ORIGEN, 0.1 day irradiation, 0.1 day decay step

The ORIGEN results (magenta) use different irradiation and decay times.

The results are found right in the middle between the two previous simulations.

ORIGEN, Thermal:
Irradiation time: between 0.1 to 10 days
Decay time: between 0 and 1 day
ORIGEN 2.1

Comparing ORIGEN results with release reports for the HFIR reactor



The ORIGEN results show best match with HFIR release reports (2007-2016) for

- **Irradiation times between 2.4 and 24 hours)**
- **Decay times of up to two days before release**

Please note that

- The plot shows $^{41}Ar/^{133}Xe$ vs $^{41}Ar/^{135}Xe$.
- Decay has been applied during irradiation.
- The HFIR data are reported annual releases.

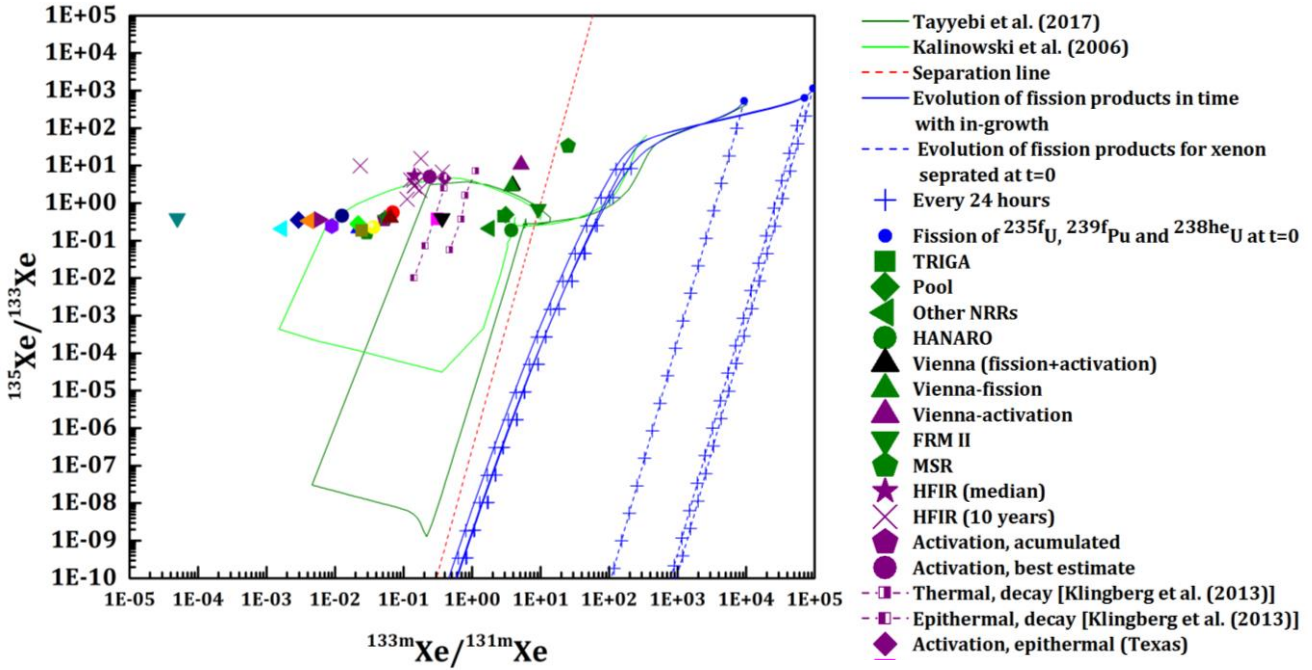
Conclusion:
We get a more robust understanding of radioxenon from activation in research reactors.

Next step:
Apply this knowledge to NPPs.

The logarithmic average for French NPPs (2002-2019)

NPP release reports are found in the activation cluster:

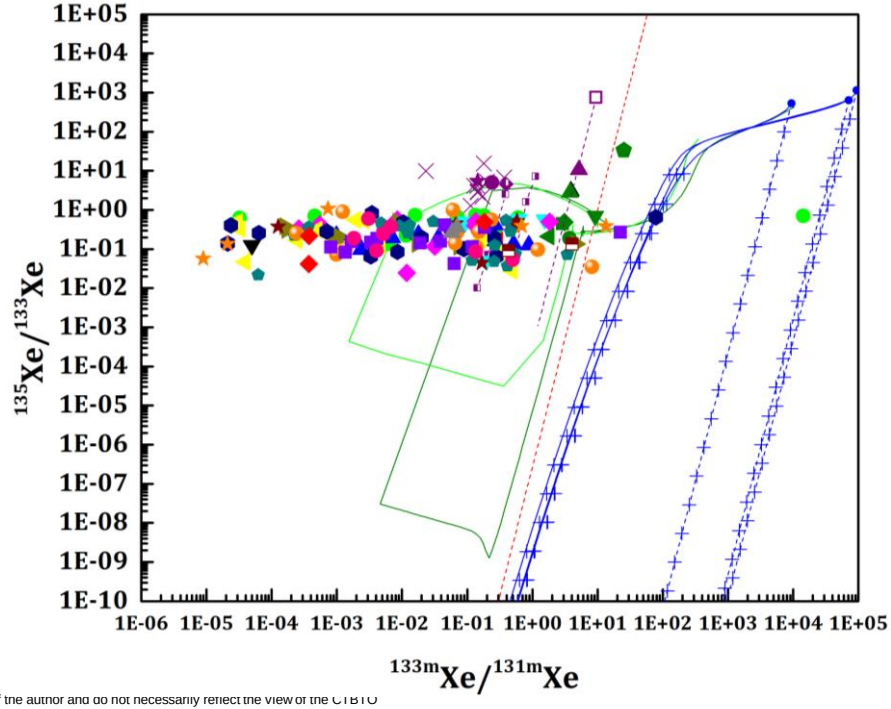
- **Could NPPs release be dominated by activation rather than fission sources?**

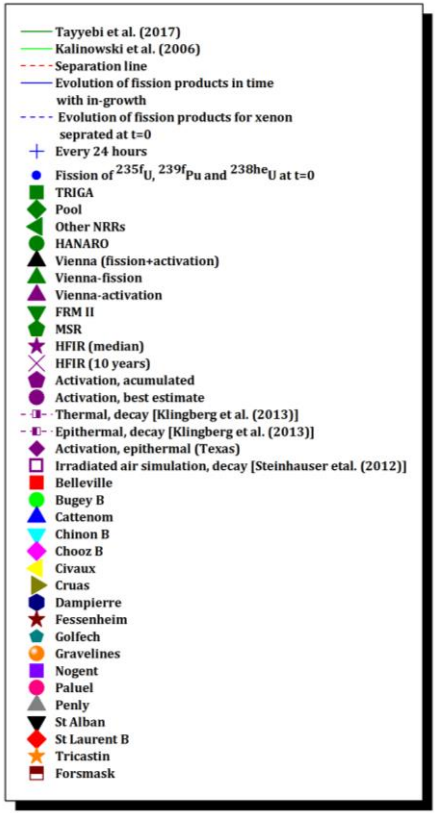


French NPPs (2002-2019) + Forsmask

Many annual French NPP release reports are in the activation cluster:

- **But e.g. Forsmark NPP (Sweden) is not**





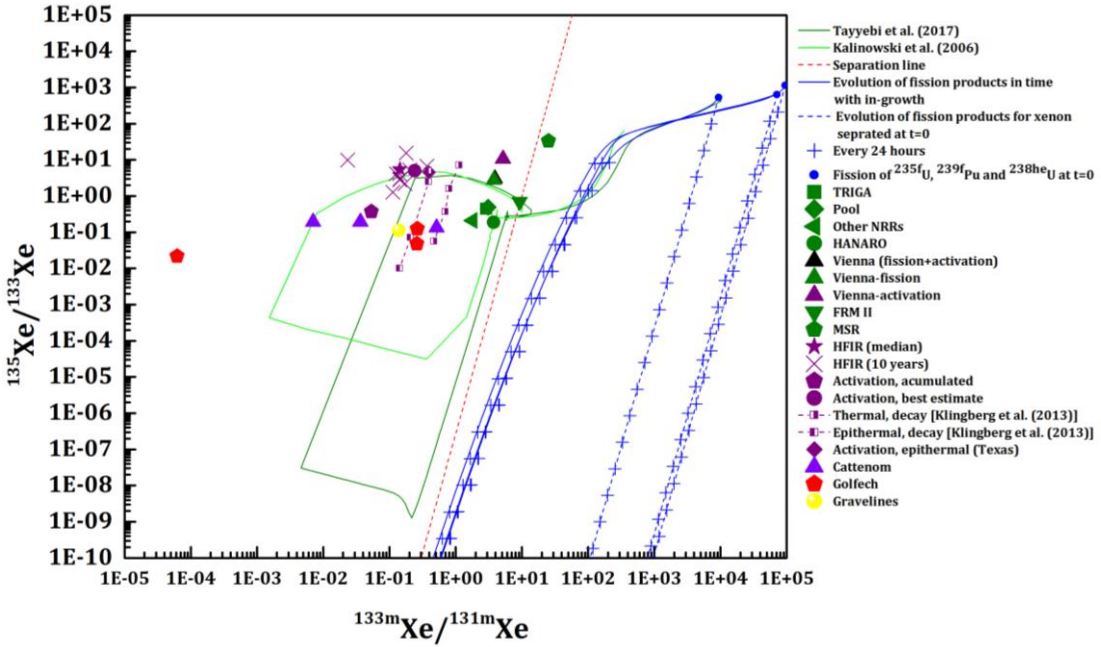
The logarithmic average for French NPPs

NPP release reports are found in the activation cluster:

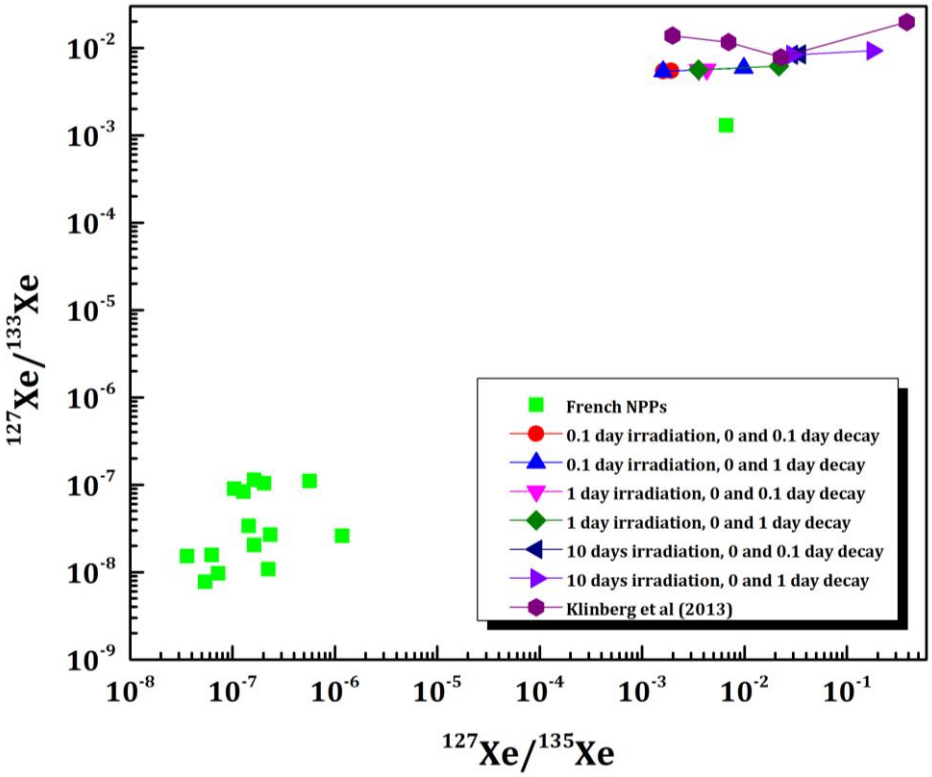
- **Could NPPs release be dominated by activation rather than fission sources?**

Now plotted only data of French NPPs which include the activation product 127Xe in their report.

The Xe-135/Xe-133 ratio is almost the same for all, but the metastable ratio is widely varying.
At least all reports that include Xe-127 are very consistent with the activation data.



French NPPs with ^{127}Xe, a pure activation product



The plot shows French reactors + Klingberg paper + ORIGEN2.1 simulation with thermal neutrons

Klingberg et al. (2013):

- irradiation time: 30 min
- Decay time: zero, 0.7 d, 1.7 d, and 2.7 d after the end of irradiation

Simulations in good agreement with each other.

Except for one outlier, the French reactor data are by several orders of magnitude off from simulations.

Conclusion:
Activation is a negligible contribution to NPP radioxenon releases which are dominated by fission.

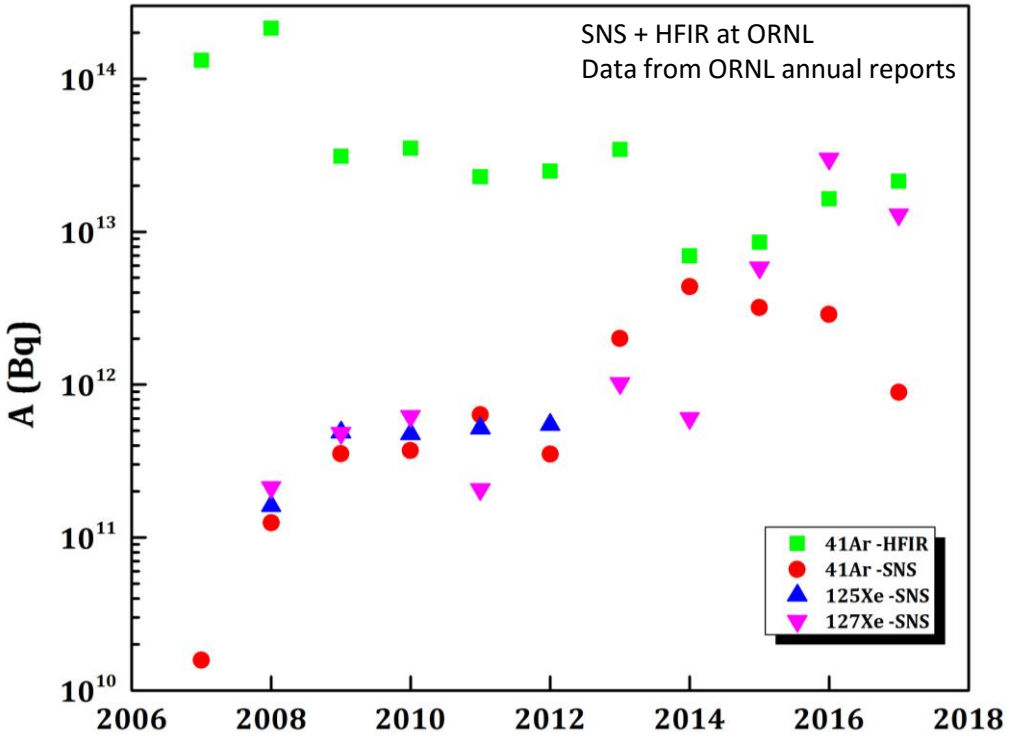
Comparison of SNS and HFIR (both at ORNL)

From 2006 to 2018, the ^{41}Ar releases of the HFIR reactor show a declining trend.

In the same time, ^{41}Ar releases of the Spallation Neutron Source (SNS) increase.

The ^{41}Ar releases of SNS reach almost the level of HFIR.

Conclusion:
Knowing that HFIR is one of the strongest sources of radioxenon from activation, it can be assumed that spallation neutron sources may also be a strong activation source.



Conclusions

- We demonstrate clearly distinct clusters for fission and activation sources in the 4–isotope plot
  - ^{133}Xe leader for fission
  - ^{135}Xe and ^{131m}Xe leader for activation
- More detailed ORIGEN simulations compared to previous studies reveal sensitivity to irradiation and decay time
- NRRs confirmed as possible strong source of radioxenon from activation
- NPPs are very unlikely to be a strong source
- Large Spallation Neutron Sources can be a strong source
- Next step: have a conclusion not only by the simulation, but also with an equation, refer to P3.5-483_Kalinowski

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Thank you for your attention

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