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# New insights into isotopic activity ratios of discharges from nuclear facilities

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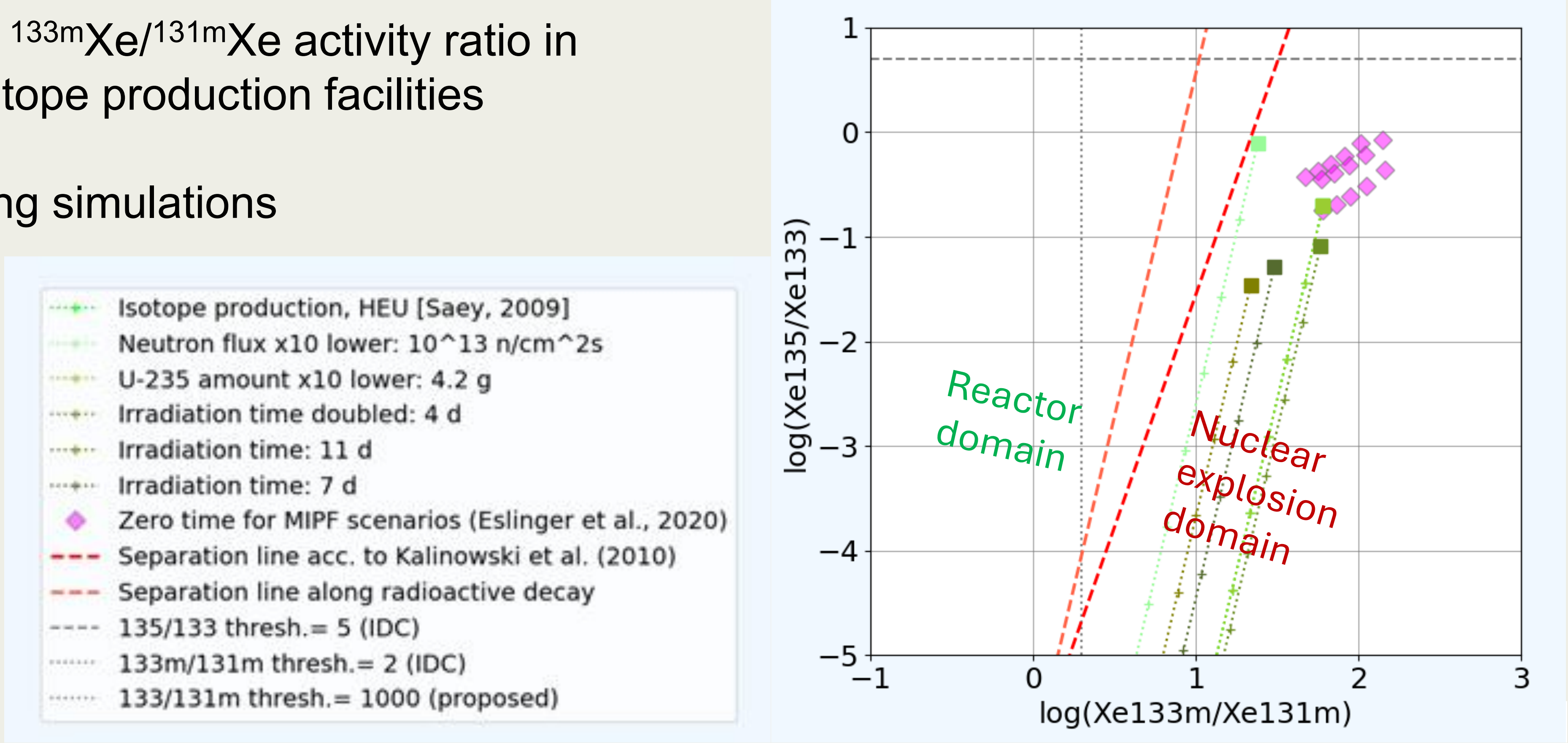
# Discrepancy between Expectations and Observations

Expectations regarding the $^{133m}Xe/^{131m}Xe$ activity ratio in emissions from medical isotope production facilities

Target irradiation and cooling simulations

- ❖ Saey (2009)
- ❖ Eslinger et al. (2020)




Unexpected:
Observations are always found left to the separation line.



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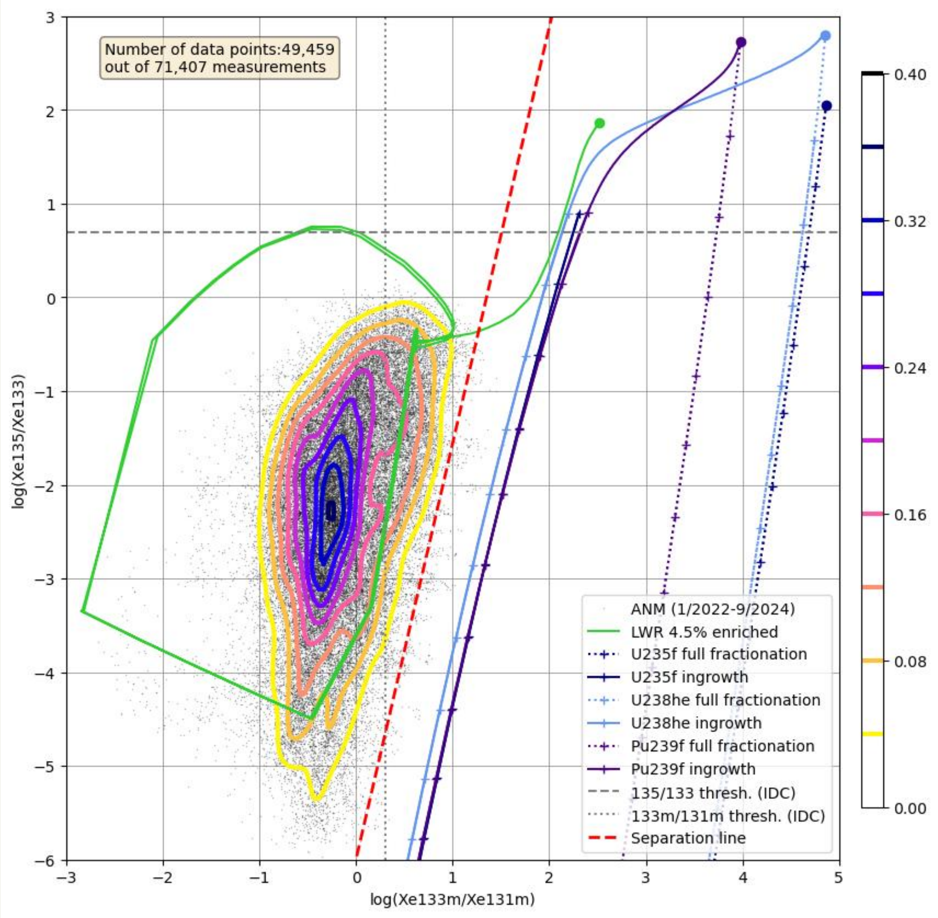
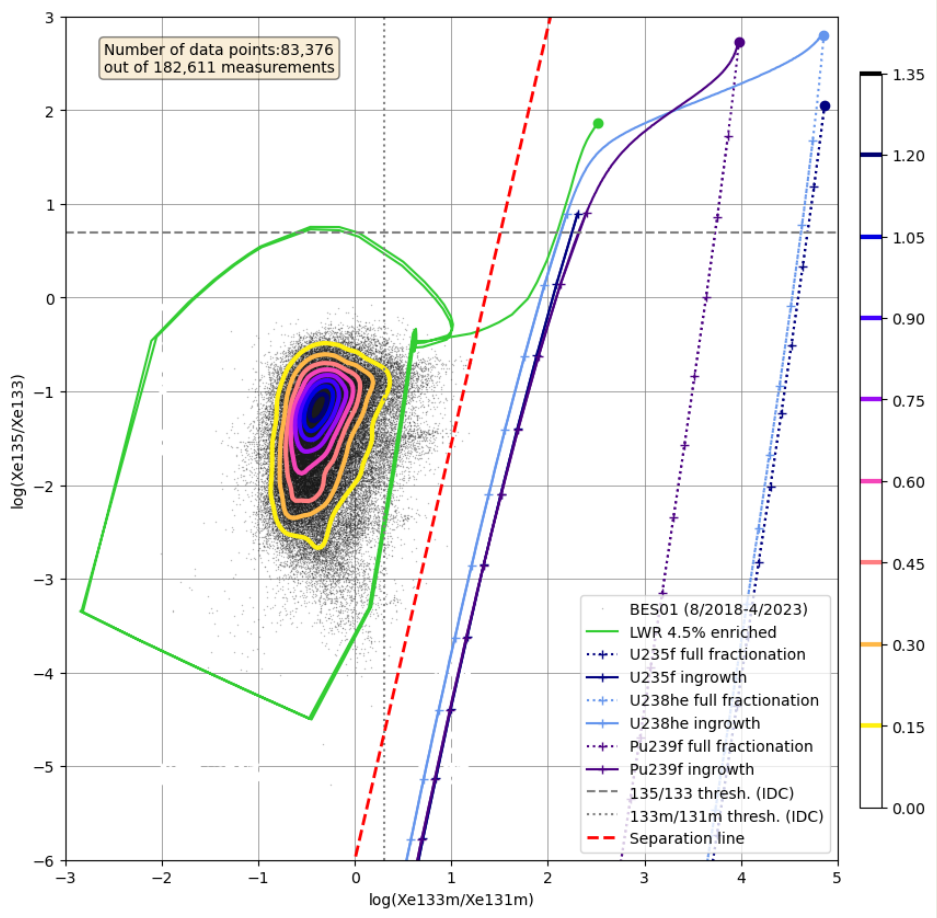
## Overview of Data in Four-Isotope-Ratio Plot

Distribution of STAX measurements at two Medical Isotope Production Facilities



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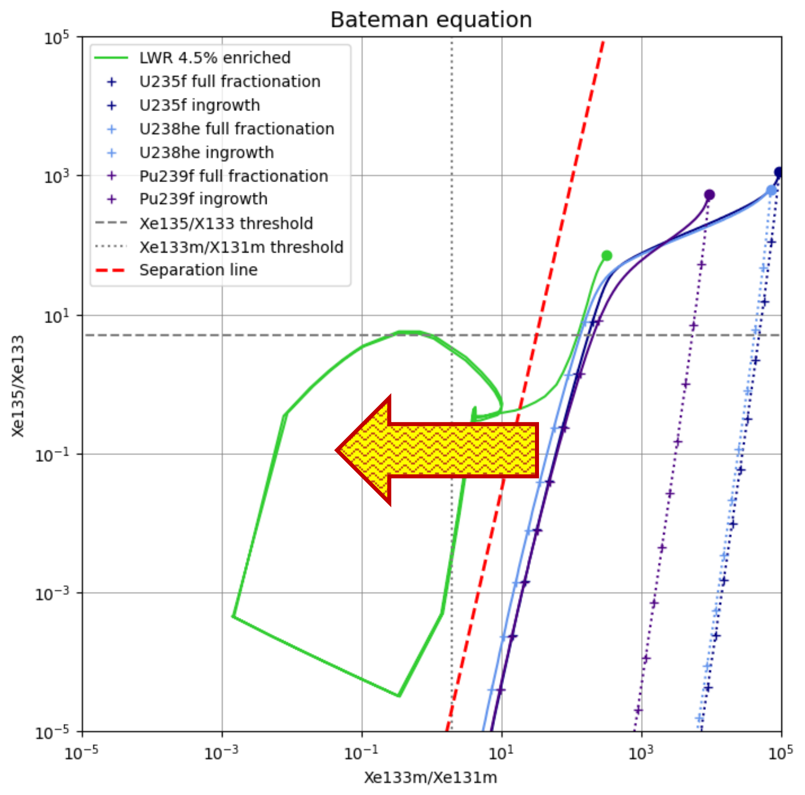
# Overview of Hypothetical Explanations

What is the reason for $^{133m}$Xe/$^{131m}$Xe shifting towards lower values?

1. Radioactive decay of a pure iodine holdup
2. Accumulation of xenon and iodine from successive target dissolutions
3. Accumulation of pure iodine from successive target dissolutions
4. One-time mixing of decayed radioxenon gases with fresh radioxenon
5. One-time mixing of decay products from a stock of pure radioiodine with fresh radioxenon



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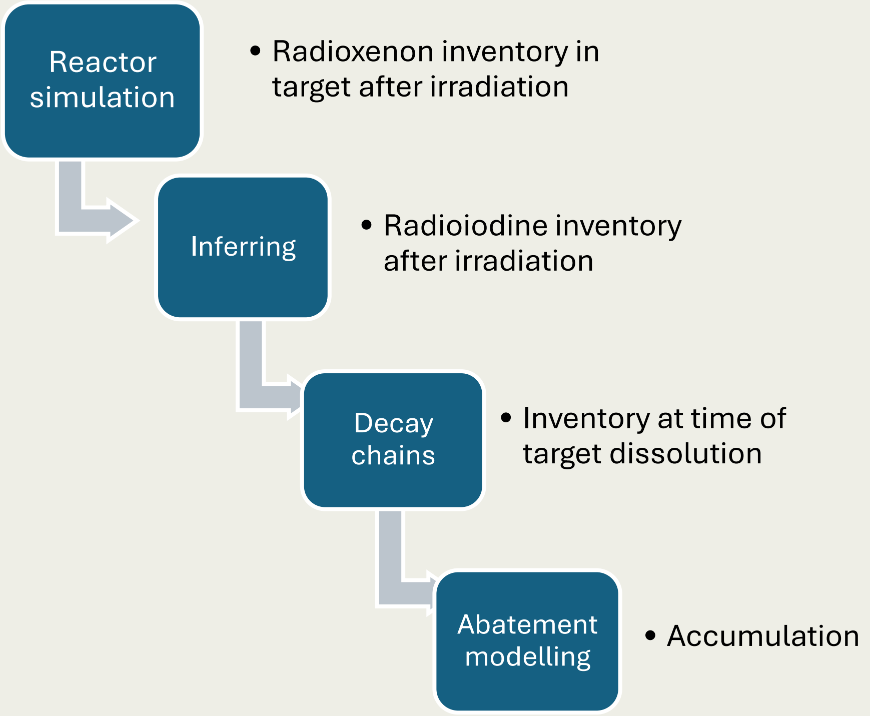
# Simulation Methods for Understanding Ratios at Discharge Time

- Initiation of the radioxenon inventory of the targets before dissolution takes place
- Inferring the radioiodine inventory of the targets from the radioxenon content
- Radioactive decay and in-growth before targets are dissolved
- Further in-growth and decay of the inventory in the abatement system
- Incremental accumulation of inventory in the abatement system



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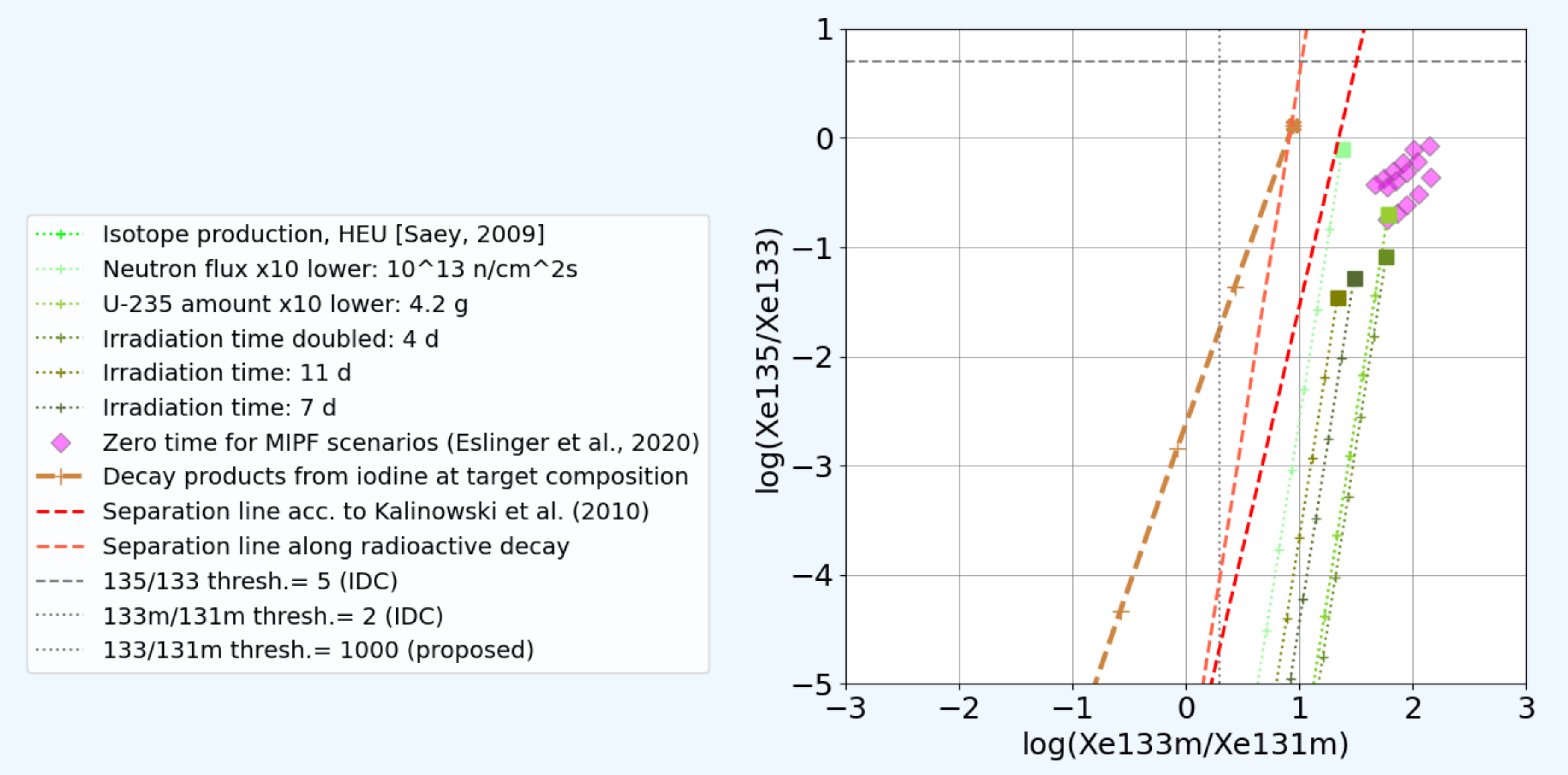
# Radioactive Decay of a Pure Iodine Holdup

On the left side of the separation line.

But not going far enough.



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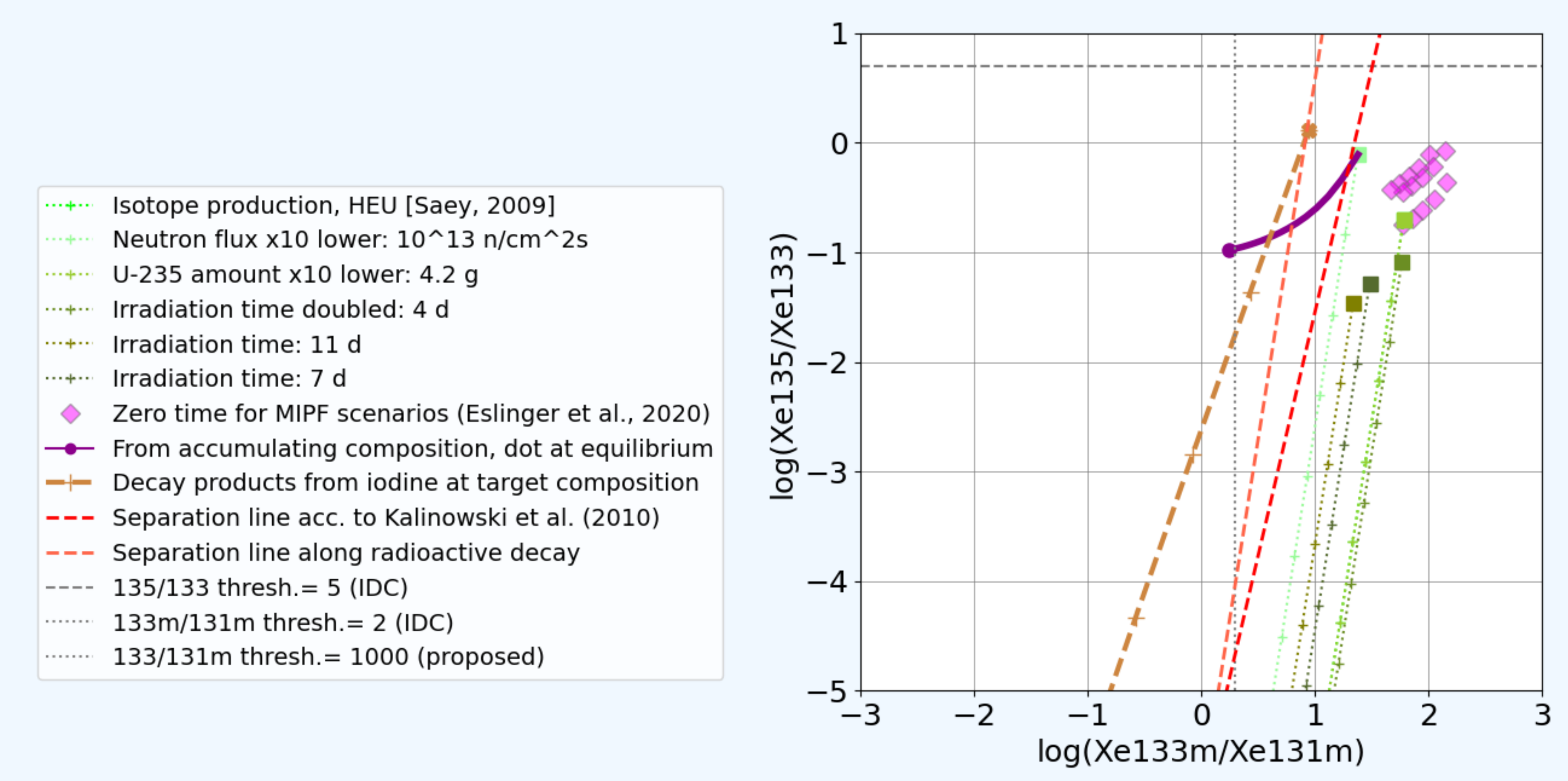
# Accumulation of Xenon and Iodine from Successive Target Dissolutions

On the left side of the separation line along decay.

A little further but too low and not going far enough.



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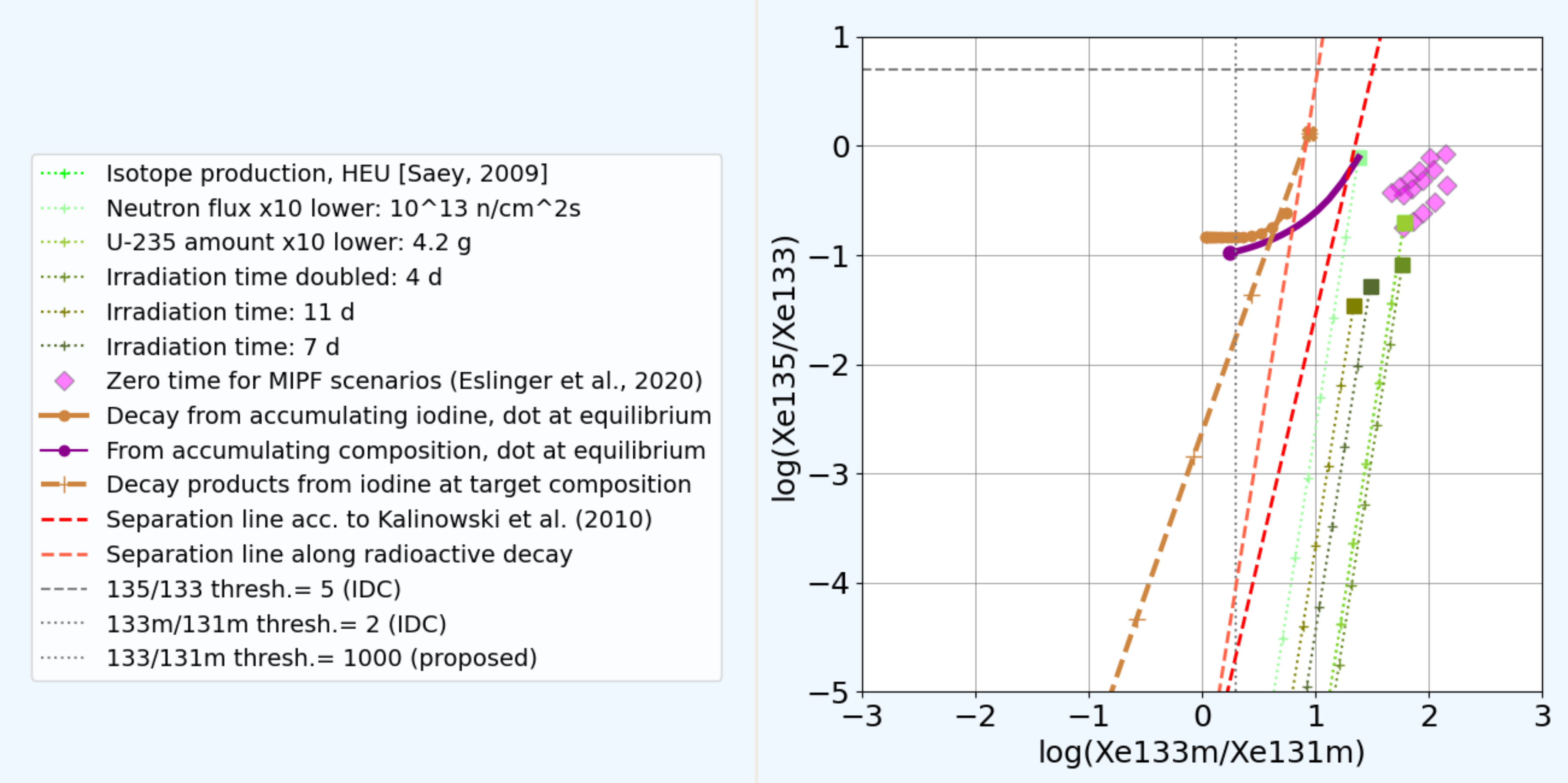
# Accumulation of Pure Iodine from Successive Target Dissolutions

On the left side of the separation line.

A bit further, but still not going far enough.



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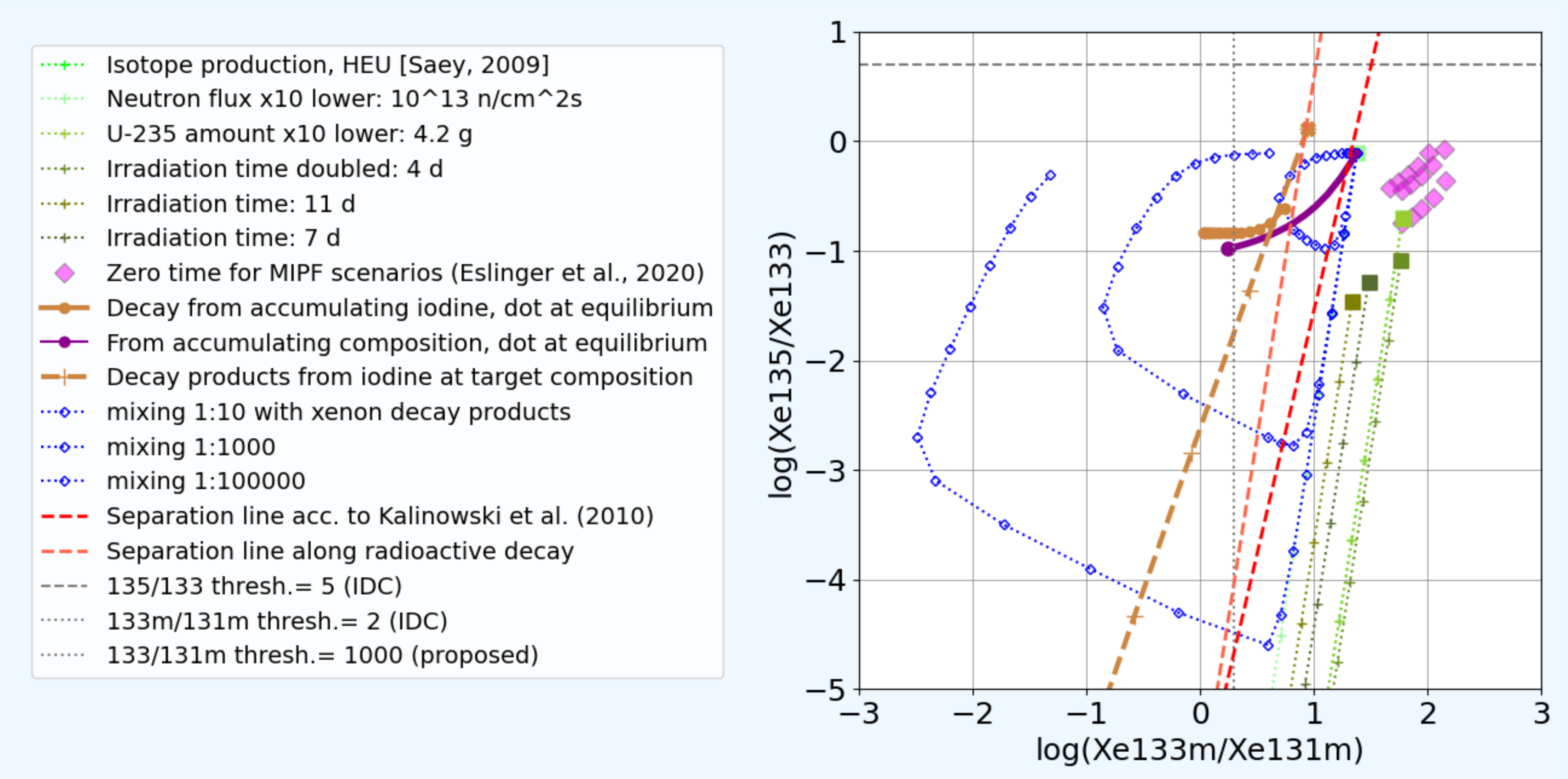
# One-time Mixing of Decayed Radioxenon Gases with Fresh Radioxenon

Mostly on the left side of the separation line.

Filling the whole space.



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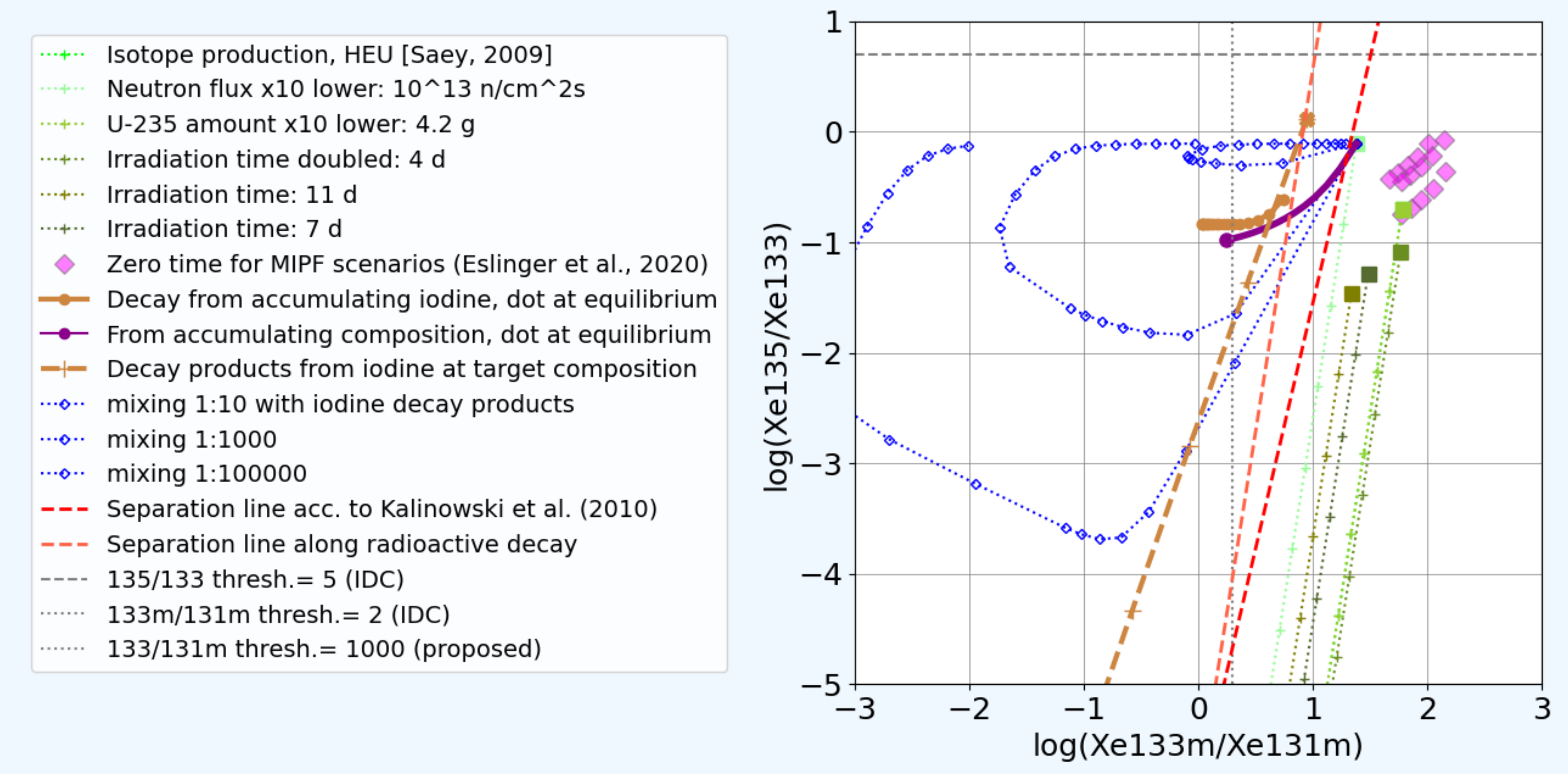
# One-time Mixing of Decay Products from a Stock of Pure Radioiodine with Fresh Radioxenon

On the left side of the separation line along decay.

Filling the whole space even further.



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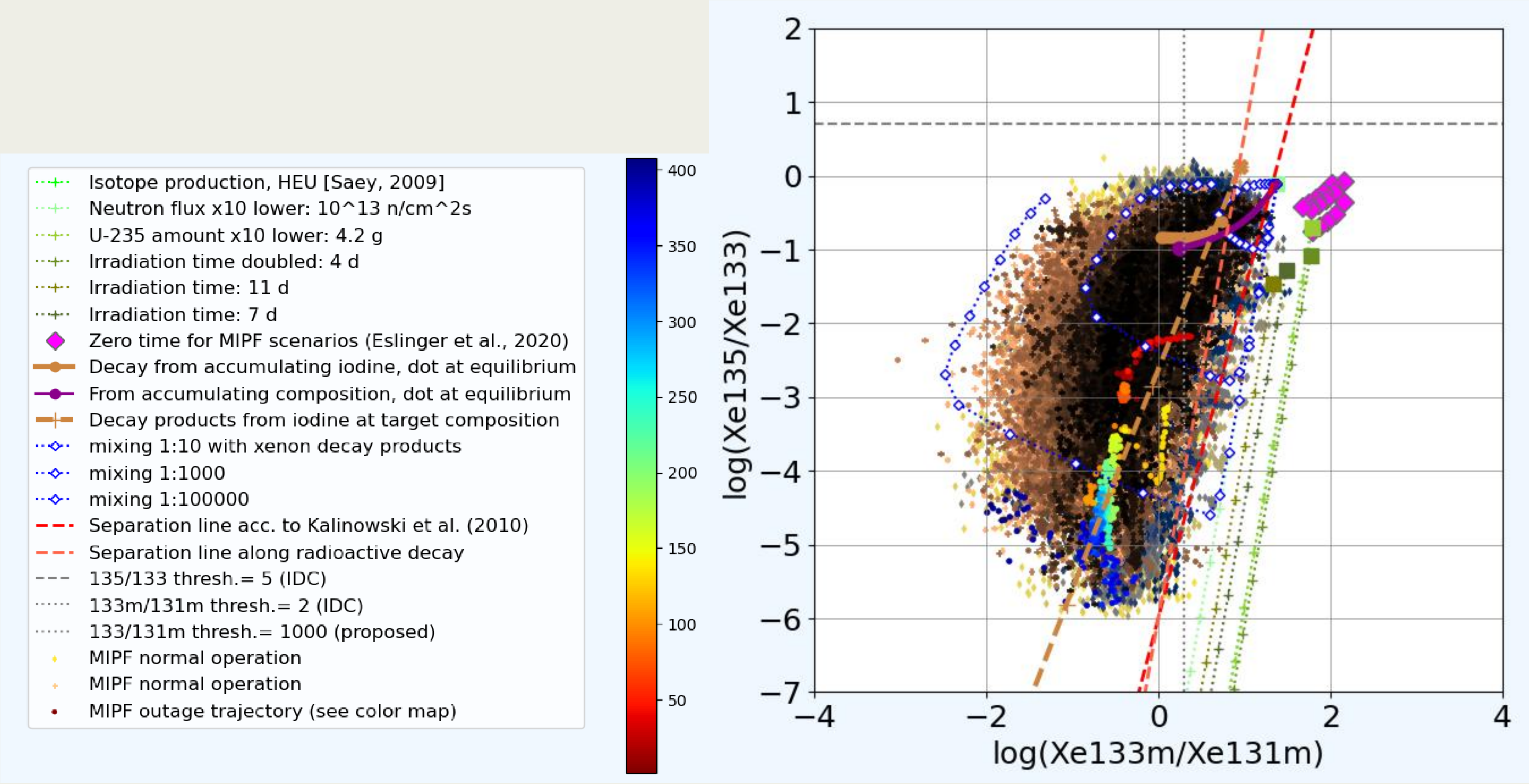
# Comparison of Key Simulations with Time Series of MIPF Discharge Rates

Comparison with STAX data from a MIPF, including a trajectory showing decay in an outage period.




Evidence that mixing is the only physical process that can explain the data.



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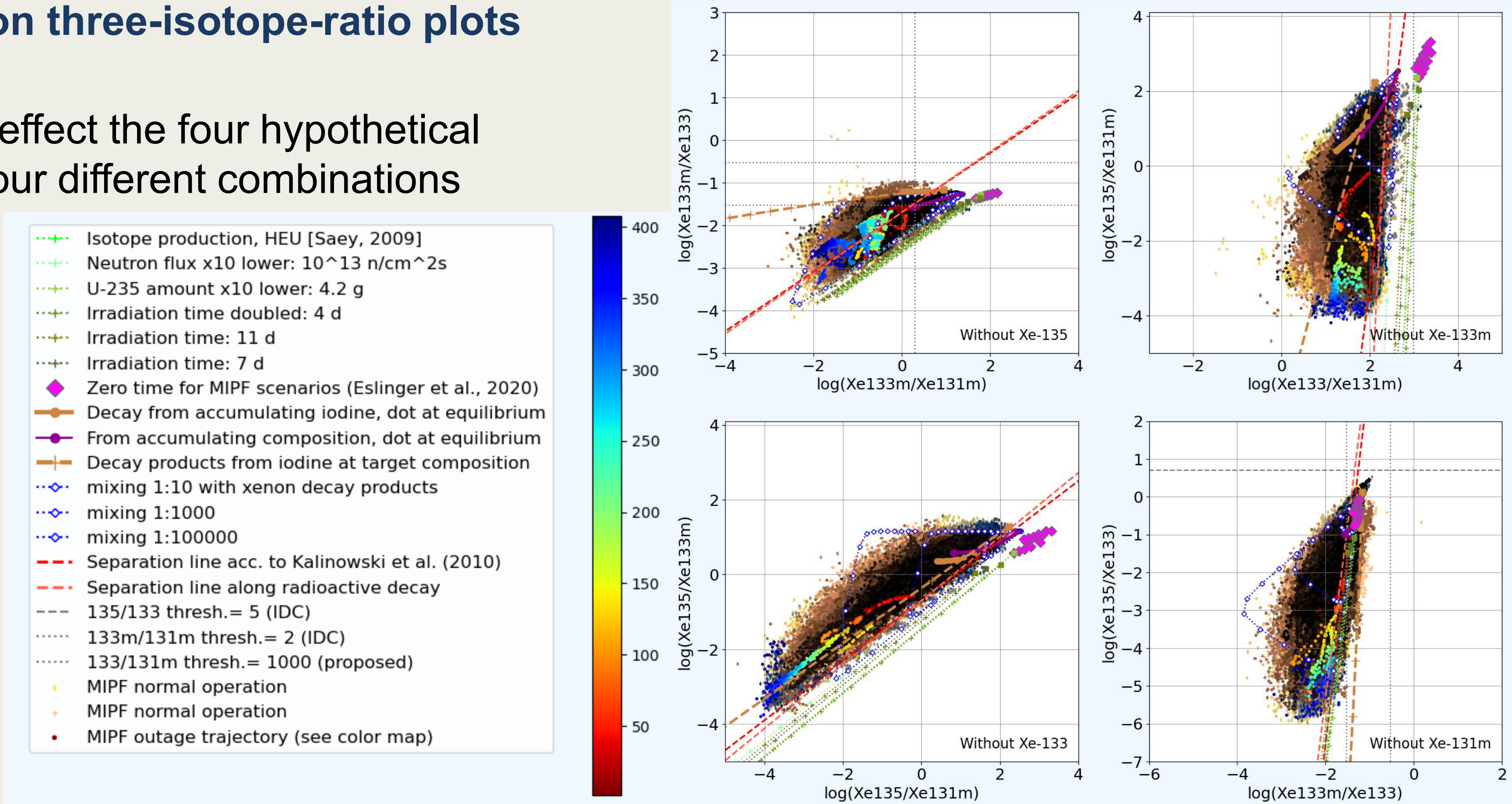
## Results on three-isotope-ratio plots

Demonstration of what effect the four hypothetical explanations have on four different combinations of three isotopes.




Mixing is the only physical process that can explain the full data spread except for a ratio combination not including $^{135}$Xe.

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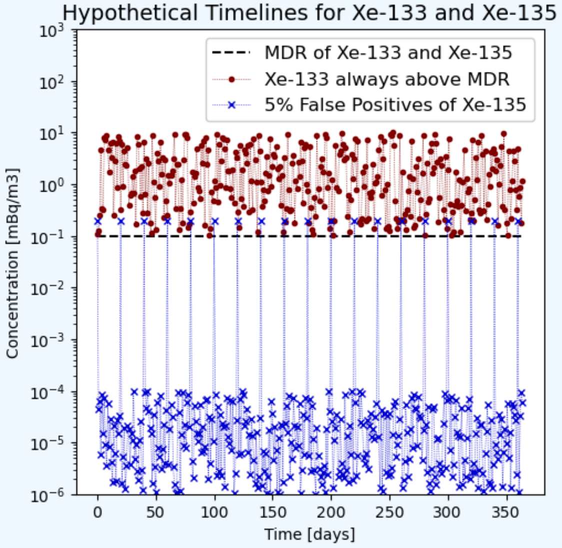
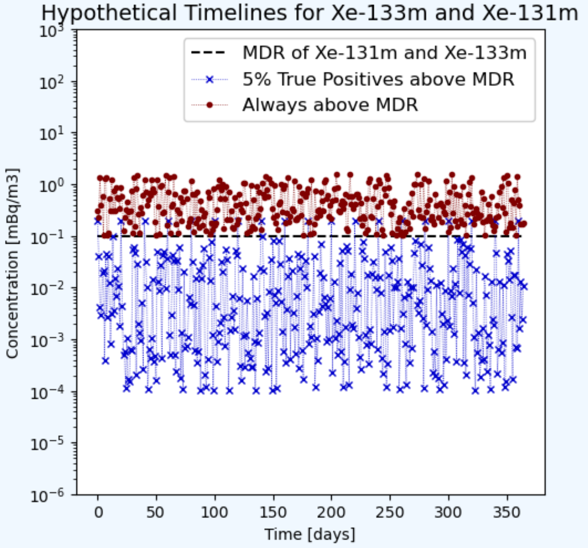
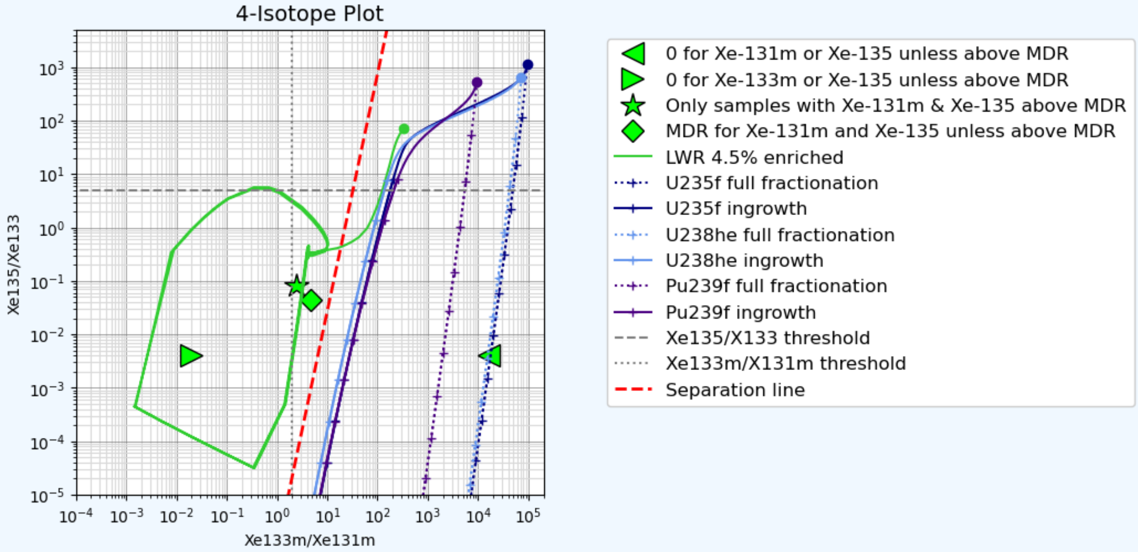
## Effect caused by using non-representative values to substitute for a measurement below MDR




Top: Hypothetical data for a full year to be used to generate *annual total discharges* for each isotope.

Bottom: 3 different ways are used to substitute non-detections when estimating annual total discharges:

1. No substitution
2. Minimum Detectable Rate (MDR)
3. Zero

In the 4-isotope plot, the ratios of annual total discharges show large discrepancies despite same hypothetical data are used. The real value is anywhere between zero and MDR, i.e. the uncertainty is several orders of magnitude.

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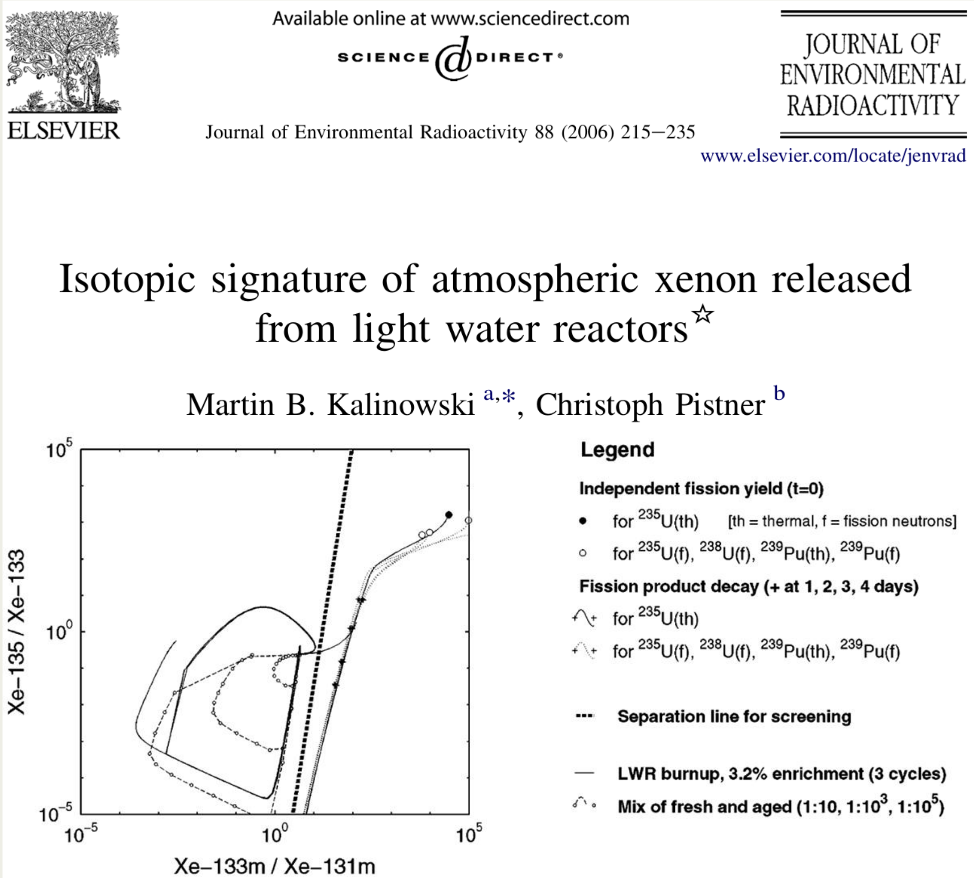
## Summary and Conclusions

- Several hypotheses are tested to explain the shift of the $^{133m}Xe/^{131m}Xe$ isotopic activity ratios towards lower values.
- Decay products from pure radioiodine, accumulation of xenon and iodine from successive target dissolutions can explain a little shift.
- The full spread of the $^{133m}Xe/^{131m}Xe$ activity ratio can only be explained by the one-time mixing of decayed gases with a fresh radioxenon release.
- This was an open question for about 15 years, and the strongest mechanism (mixing) was published in 2006.
- What questions are left to be answered?




Disclaimer: The views expressed here are those of the authors and do not necessarily reflect the opinion of the United States Government, the United States Department of Energy, PNNL, or ISTI.

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