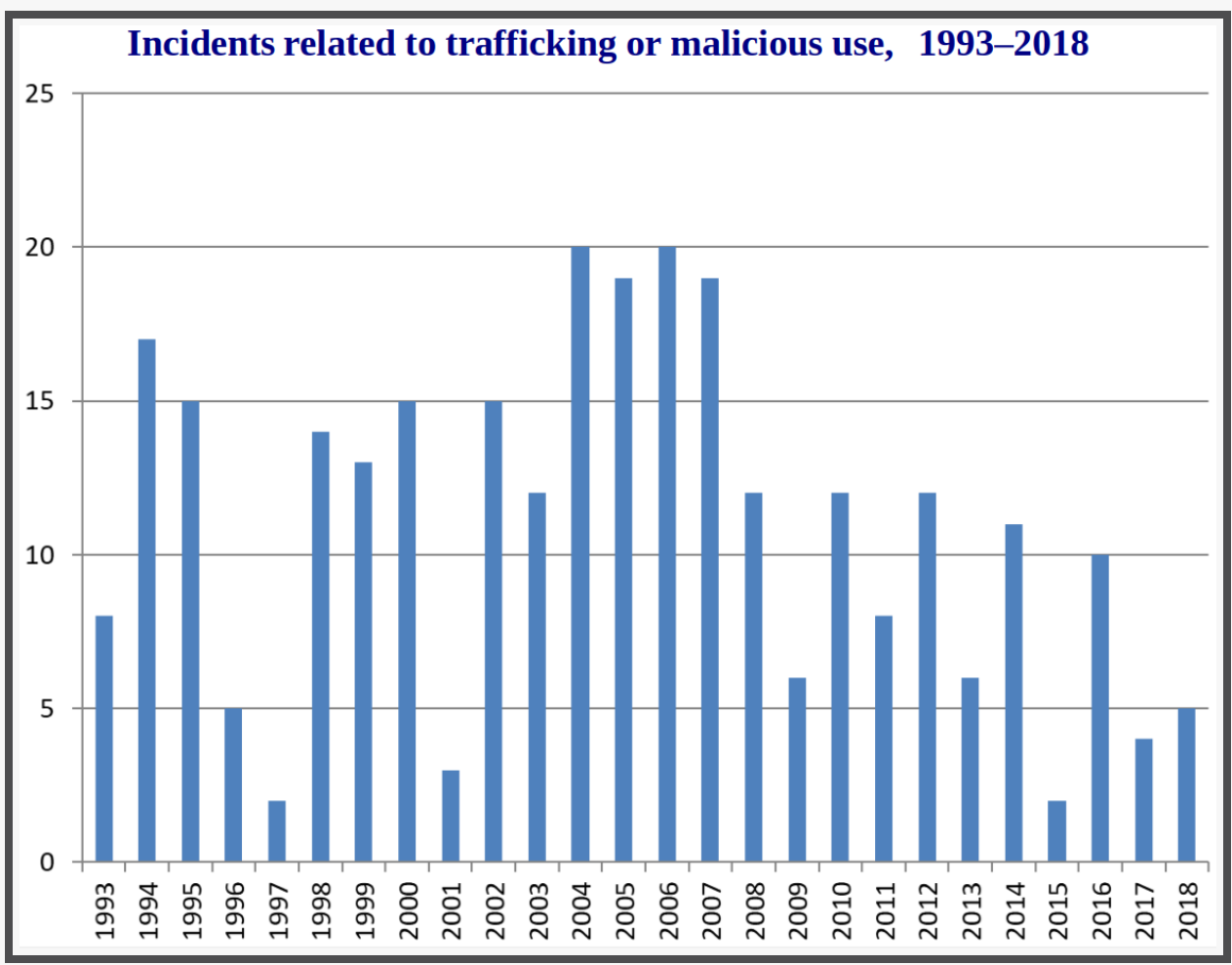


## Motivation: Speeding up nuclear emergency response

After a nuclear weapon is detonated or nuclear material is intercepted, a priority for emergency responders is to determine both where it came from and the radioactive danger to the public. While the latter can be determined quickly, the former often involves lab work that can take days or weeks.

Attribution of nuclear material is a major part of a nuclear forensics investigation. This informs both emergency response and what actions the government takes. A strong nuclear forensics capability both deters governments from engaging in state-sponsored nuclear terrorism and interrupts the pathways being used to create weapons.

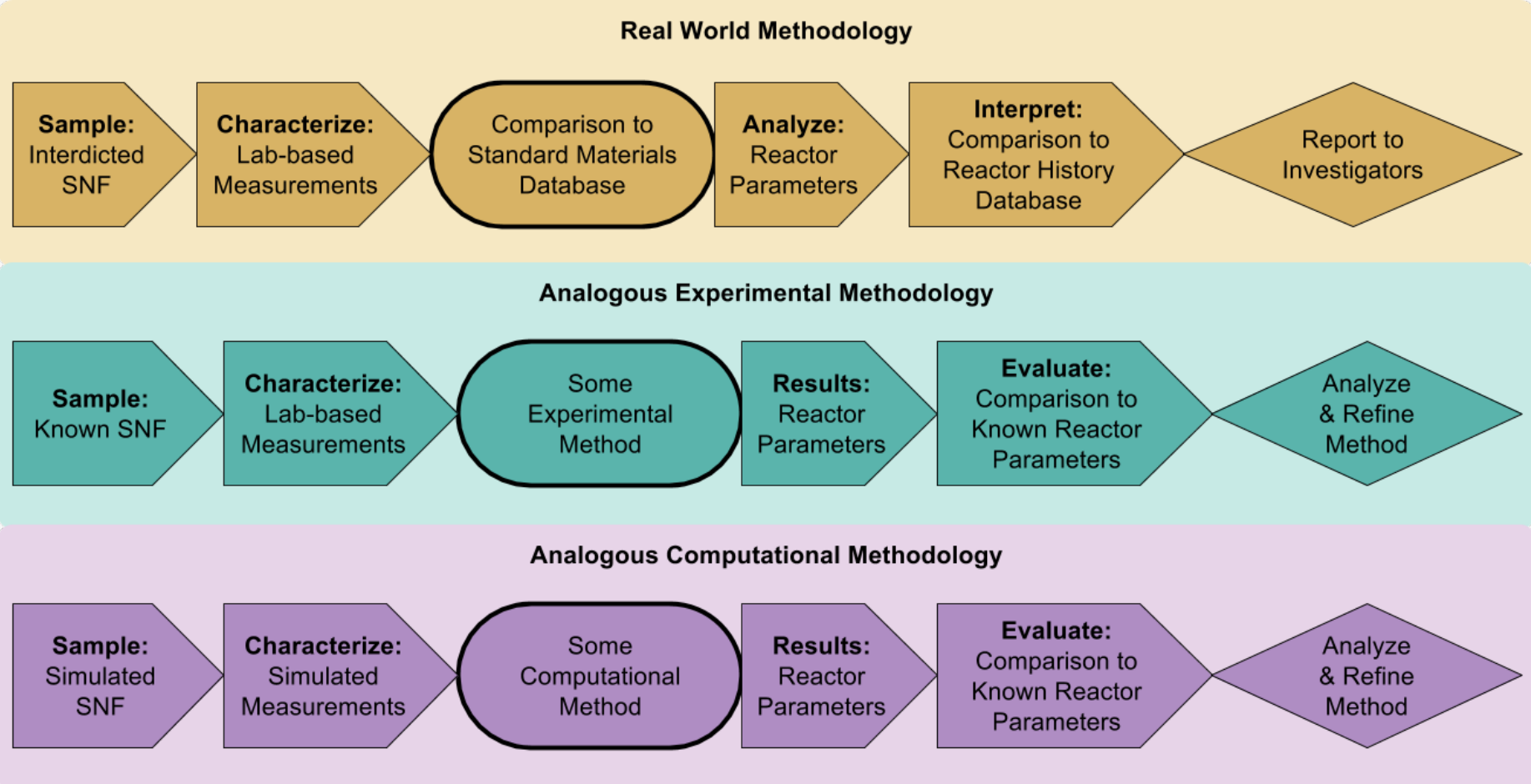


Participating countries (138) report intercepted nuclear materials intended for illicit use to the IAEA.[1]

The figure shows the number of nuclear material incidents tracked in the last 25 years, of which 12 involved highly enriched uranium, 2 plutonium, and 4 plutonium-beryllium neutron sources. Thus, this work focuses on spent nuclear fuel from power reactors outside of regulatory control as a material of interest.

## Background: Using statistical methods to evaluate nuclear forensics signatures

Nuclear forensics research initiatives include characterization of both pre- and post-detonation materials. Measuring isotopic ratios, chemical compounds, and trace elements are signatures used to identify the chain of custody of these materials. Considering spent nuclear fuel, the signatures help determine a set of reactor parameters that generated the material: reactor type, fuel enrichment, burnup, and cooling time. This provides information that can lead to the source (country, exact reactor) of the material in question.



Nuclear forensics research workflows: physical, experimental, and computational

Presented here is a methodology that seeks to rapidly provide investigation-guiding information using measurements taken in the field compared against statistical models. Statistical methods may be able to determine reactor operation parameters faster than the traditionally utilized empirical relationships. Shown in the figure is at the top a typical investigative workflow, followed by an experimental workflow that uses some method instead of direct comparison to a standard materials database. This is extrapolated to a computational method in the bottom panel, which is discussed further in the methodology sections.

## Research Breakdown

How does the ability to determine forensic-relevant spent nuclear fuel attributes using machine learning techniques degrade as less information is available?

**Determine**  
The inverse problem: given end measurements, calculate the model parameters that created them

**Information**  
Nuclide vectors, measurements of isotope ratios

**Forensic-relevant Attributes**  
Reactor type, enrichment, cooling time, burnup

**Machine Learning Techniques**  
Creating statistical models (not physical)

**Degrade**  
Model prediction performance

**Less Information**  
Error in nuclide vectors, fewer measurements, etc

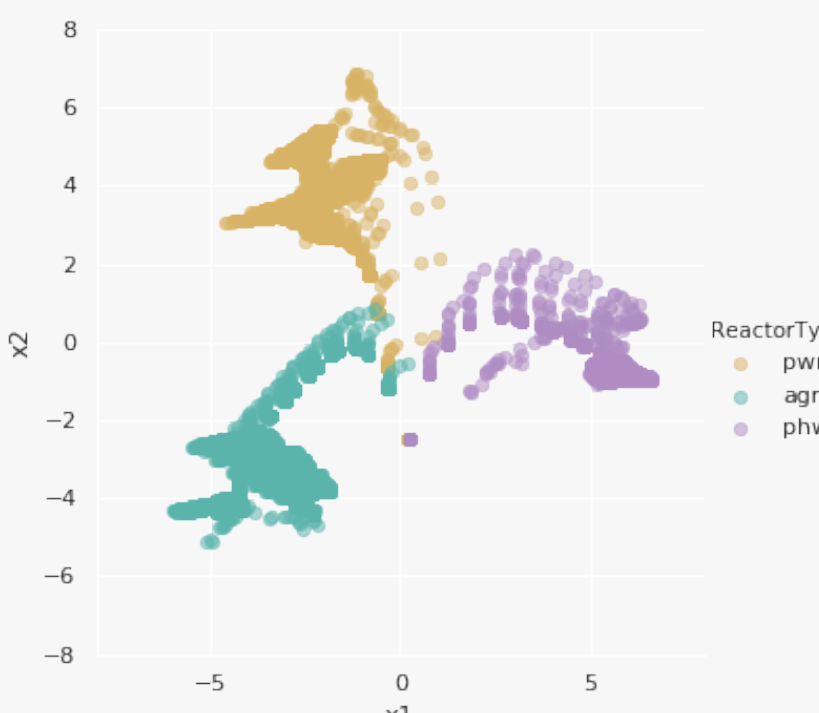
Definition of terms within the main research question

## Methodology: Training set creation

The training set design is based on comparing this workflow to that in previous work [4]. The spent nuclear fuel was simulated using Oak Ridge National Laboratory's ORIGEN [2]. While this database contains only three reactors each at a single enrichment, there will be many reactors at many enrichments added to this database after proof-of-concept is complete.

	PWR	AGR	PHWR
Power Density [MW/MTU]	35	12.5	20
Burnup [MWd/MTU]	{0-8500} in 85 steps		
Moderator Density [g/cc]	0.72	1.65	0.84
Enrichment [% U235]	3.0	4.0	0.711
Cooling Time [days]	{0-5000} in 2500 steps		

Inputs for simulations in training set. The labels include: reactor type, burnup, enrichment, cooling time



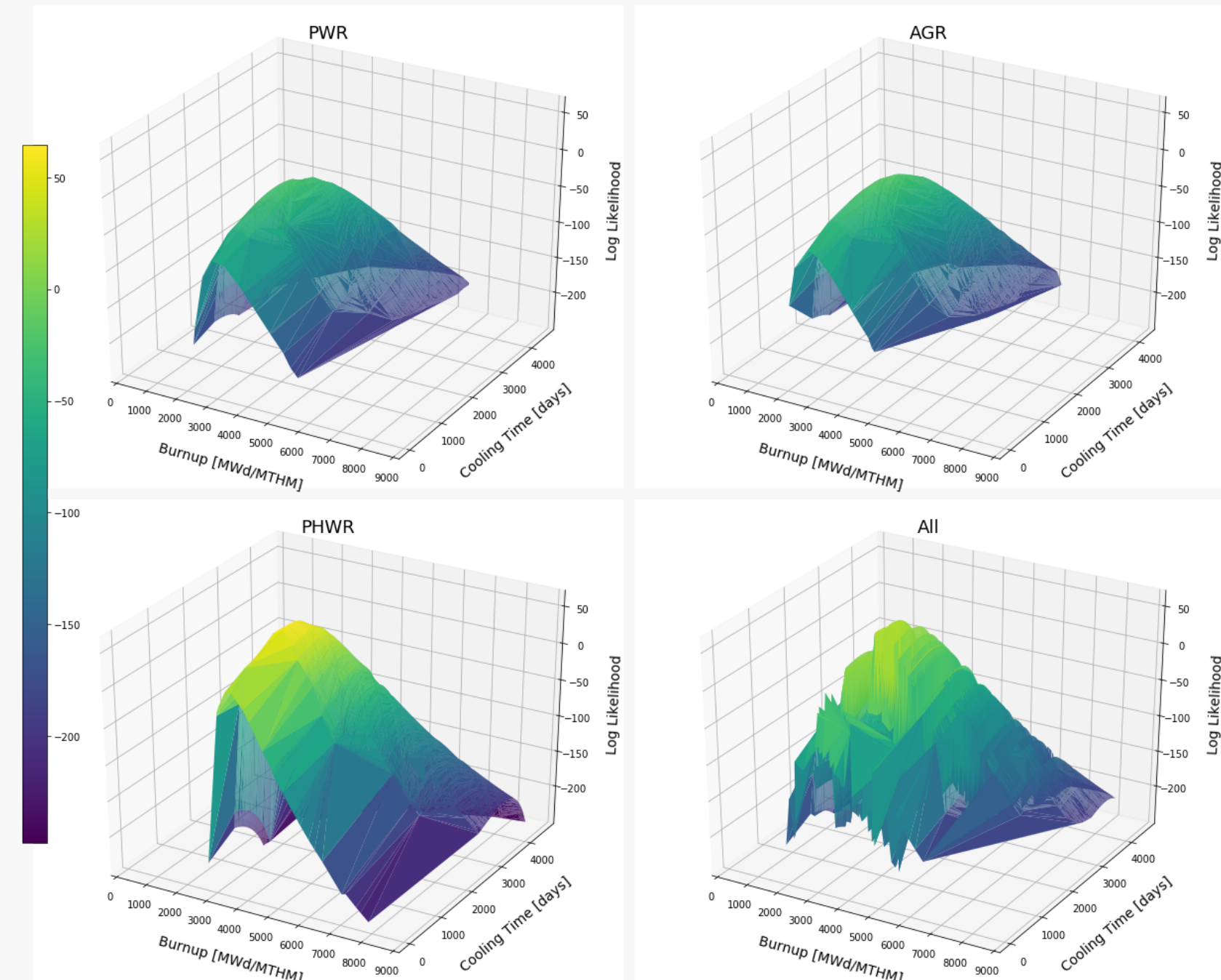
Dimensionality reduction step for visualization: linear discriminant analysis of training set can reduce dimensions from 10-15 to 2.

$\frac{^{137}Cs}{^{133}Cs}$	$\frac{^{134}Cs}{^{137}Cs}$	$\frac{^{135}Cs}{^{137}Cs}$	$\frac{^{136}Ba}{^{138}Ba}$	$\frac{^{150}Sm}{^{149}Sm}$
$\frac{^{152}Sm}{^{149}Sm}$	$\frac{^{154}Eu}{^{153}Eu}$	$\frac{^{240}Pu}{^{239}Pu}$	$\frac{^{241}Pu}{^{239}Pu}$	$\frac{^{242}Pu}{^{239}Pu}$

Features tracked for the training set. This includes 10 isotope ratios known to discriminate the chosen labels well.

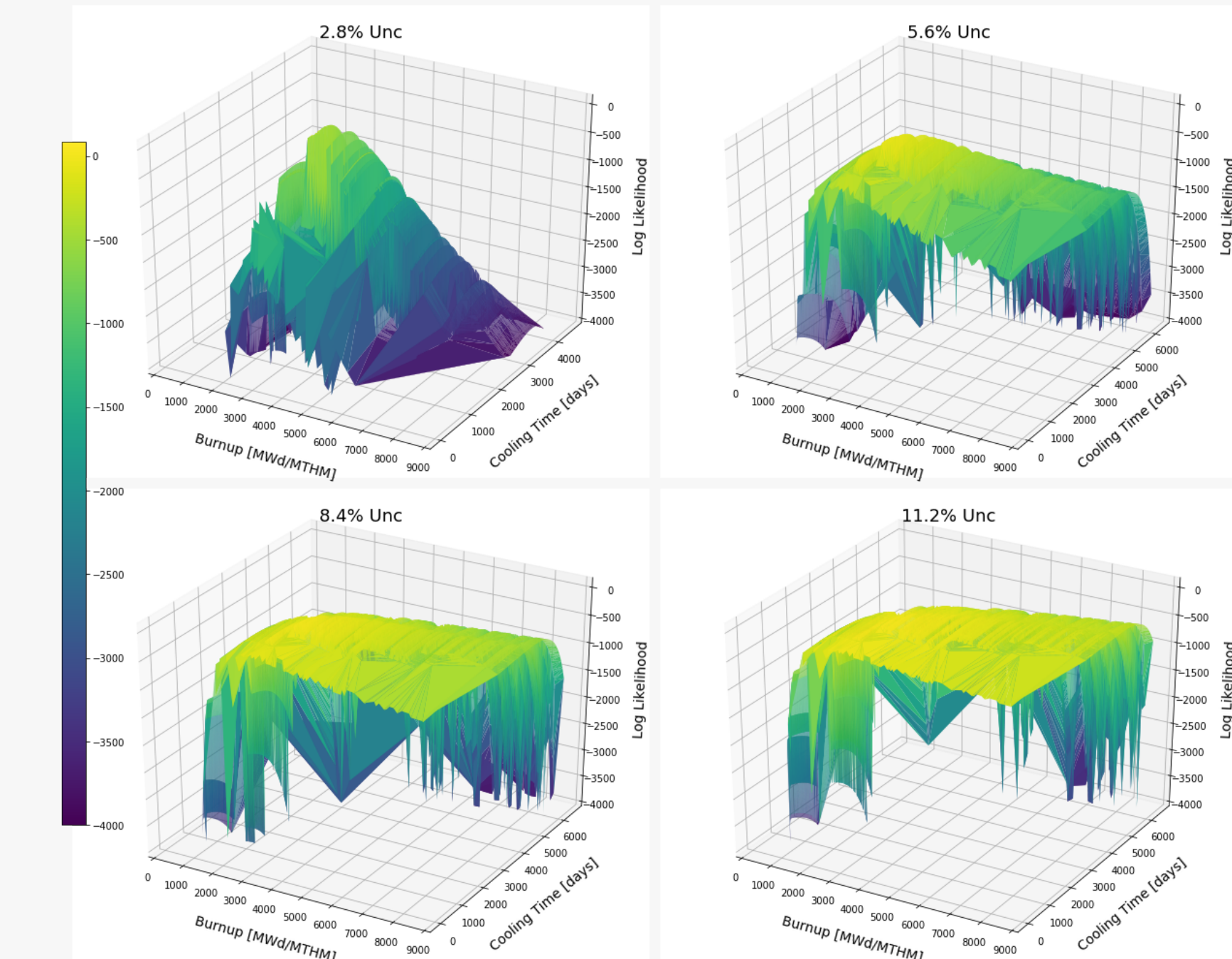
## Results: MLE Method provides more than just predictions

Texty text



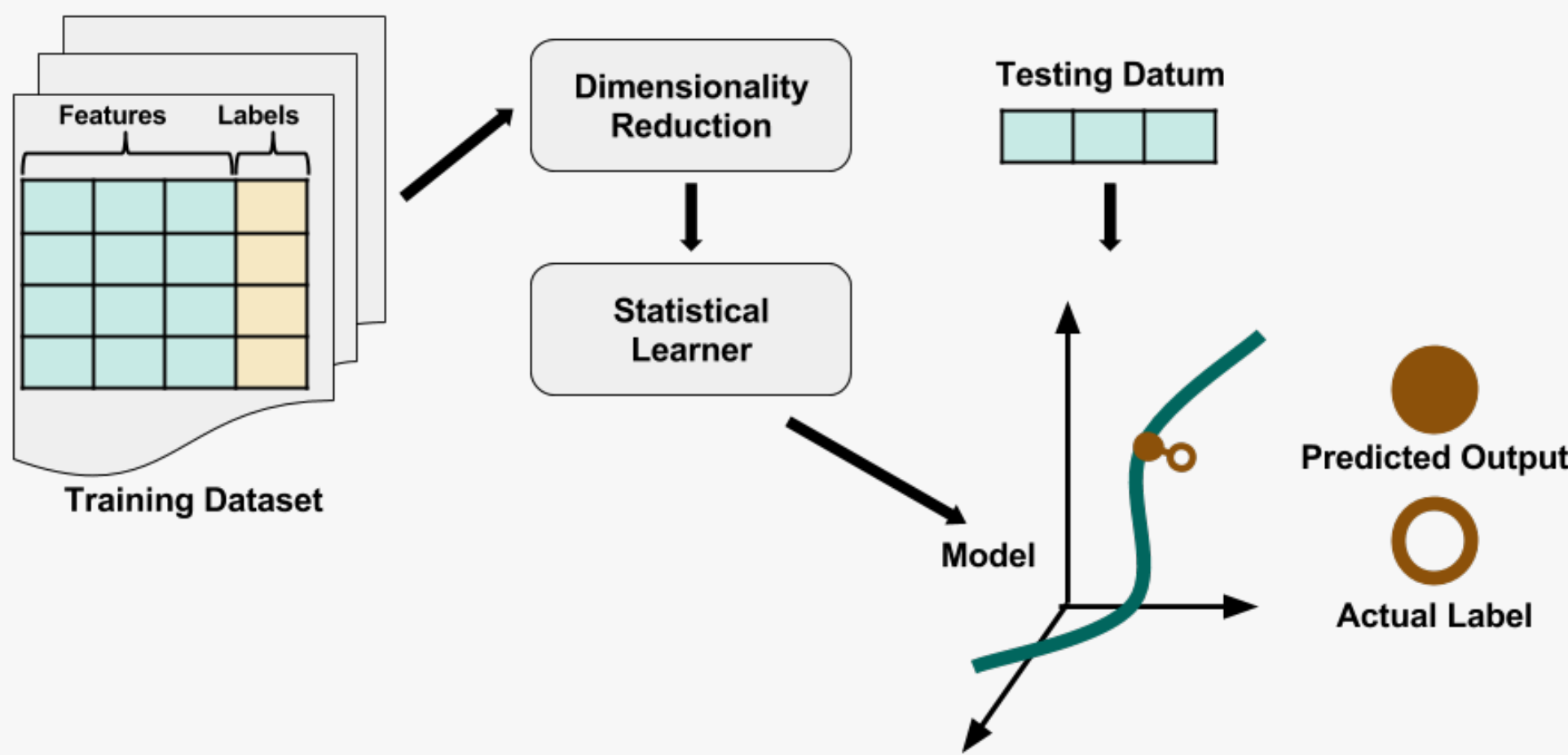
placeholder figure showing filtered results wrt rxtr

More texty text!



placeholder figure showing results wrt increasing uncertainty

## Methodology: Maximum likelihood estimation for prediction



Schematic of a representative training and predicting workflow

A generalized machine learning approach for prediction is shown here. For the dimensionality reduction step, 200+ isotopes from the simulation were reduced to 10 isotope ratios. For the statistical learner, a maximum likelihood estimation (MLE) method was chosen, in part to build upon previous work for this application. [4, 5]

Likelihood calculated is as follows:

$$L(M|r_{meas}) = \prod_i \frac{1}{\sigma_{i,sim} \sqrt{2\pi}} \exp \frac{-(r_{i,meas} - r_{i,sim})^2}{2\sigma_{i,sim}^2}$$

A given unknown sample (i.e., testing datum above) with 10 isotope ratios comprising  $\mathbf{r}_{meas}$  will have a likelihood calculated against the entire training set, each entry containing the ratios  $\mathbf{r}_{sim}$ . The prediction is the reactor type entry with the maximum likelihood. The predicted entry also contains the burnup, cooling time, and fuel enrichment. An uncertainty prediction is given from the simulation uncertainty in the isotope ratios,  $\sigma_{sim}$ .

## Future Work

Information reduction using Gamma Detector and Response and Analysis Software (GADRAS) [6], developed at Sandia National Laboratories, will be used next. This limits the training set features to isotopes that are detectable in realistic scenarios.

Next, a real-world spent fuel database will be used for testing data: the Spent Fuel Composition Database (SFCOMPO) [3]. This will provide a benchmark of how this method performs with measurements from commercial reactors worldwide.

## References

- <sup>1</sup>Incident and Trafficking Database (ITDB) Program, *IAEA Incident and Trafficking Database: 2019 Fact Sheet*, tech. rep. (International Atomic Energy Agency, Division of Nuclear Security, 2019).
- <sup>2</sup>B. Rearden and M. Jessee, "Ch. 5 Depletion, Activation, and Spent Fuel Source Terms", in *SCALE Code System: User Documentation*, Version 6.2.1; ORNL/TM-2005/39 (Apr. 2016), pp. 5-1-5-263.
- <sup>3</sup>F. Michel-Sendis, J. Martinez-González, and I. Gauld, "SFCOMPO 2.0 – a relational database of spent fuel isotopic measurements, reactor operational histories, and design data", *EPJ Web of Conferences* **146**, 06015 (2017).
- <sup>4</sup>J. M. Osborn, E. D. Kitcher, J. D. Burns, C. M. F. III, and S. S. Chirayath, "Nuclear Forensics Methodology for Reactor-Type Attribution of Chemically Separated Plutonium", *Nuclear Technology* **201**, 1-10 (2018).
- <sup>5</sup>E. D. Kitcher, J. M. Osborn, and S. S. Chirayath, "Sensitivity studies on a novel nuclear forensics methodology for source reactor-type discrimination of separated weapons grade plutonium", *Nuclear Engineering and Technology* (2019) 10.1016/j.net.2019.02.019.
- <sup>6</sup>S. M. Horne, G. G. Thoreson, L. A. Theisen, D. J. Mitchell, L. Harding, and W. A. Amai, *Gamma Detector Response and Analysis Software - Detector Response Function (GADRAS-DRF)*, User's Manual, Version 18.5; SAND2014-19465 (Sandia National Laboratories, Albuquerque, New Mexico, USA, Dec. 2014).

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