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Bayesian Optimal Experimental Designs for High-dimensional Physics-based Models

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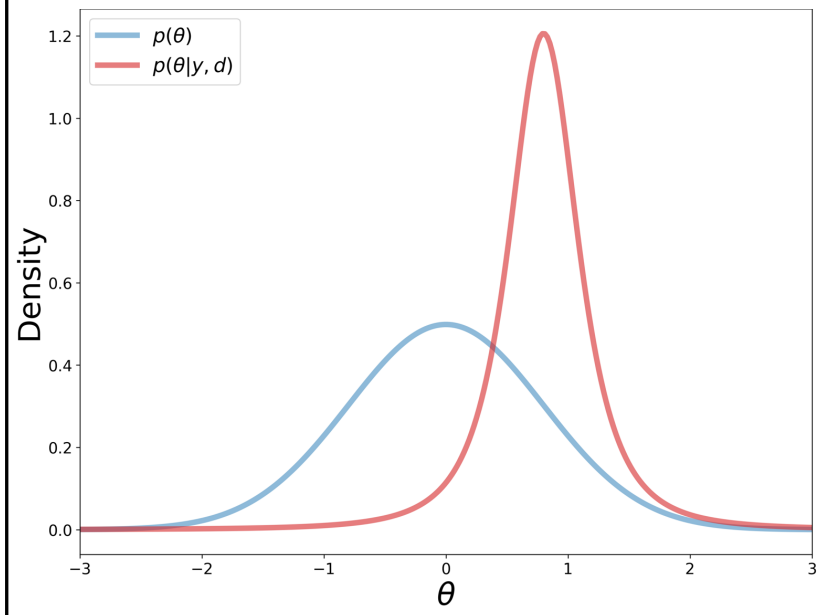
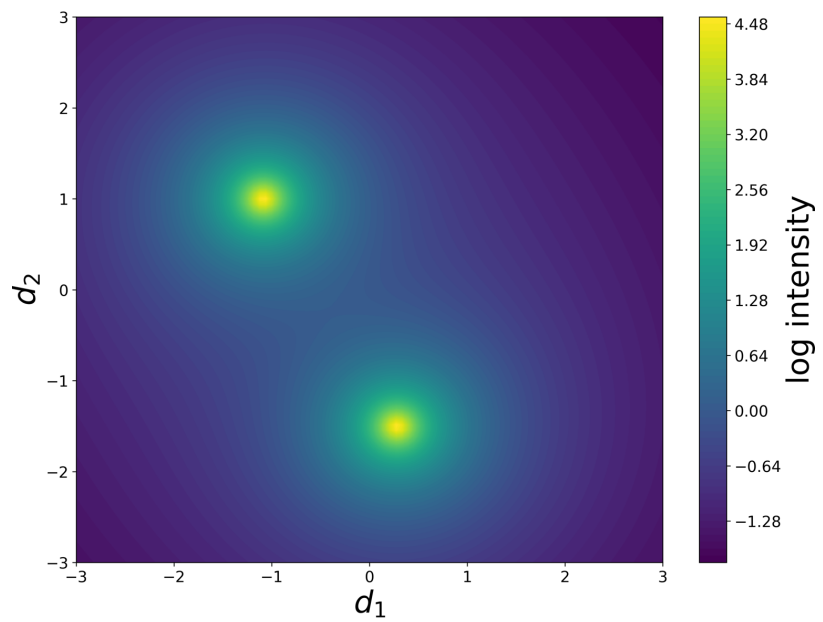
Motivation

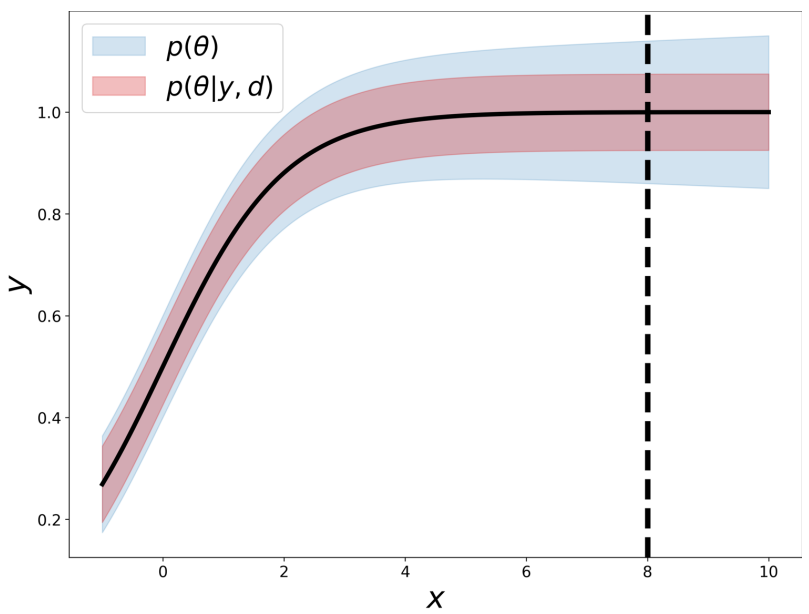
Experimental data helps us improve our understanding of a problem



Learning unknown parameter values



Identifying source locations in a field

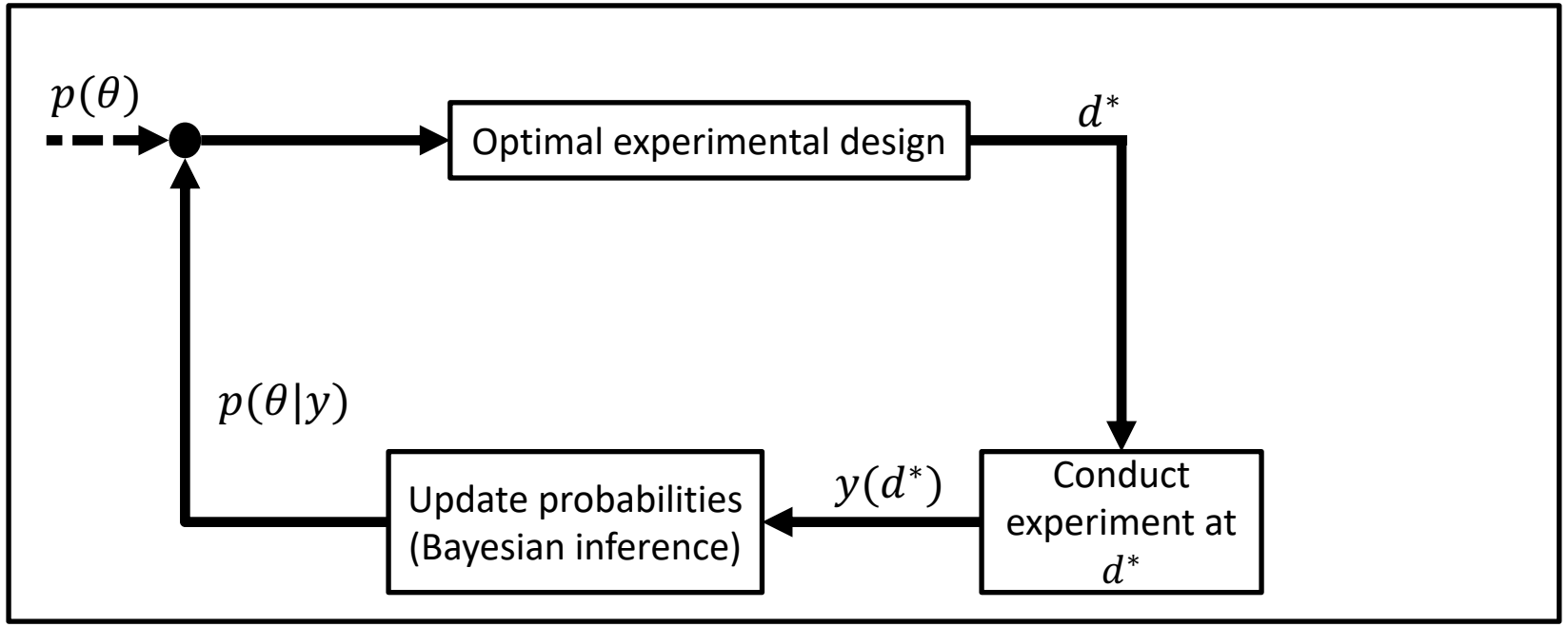


Improving predictivity of a QOI

Decision making problem

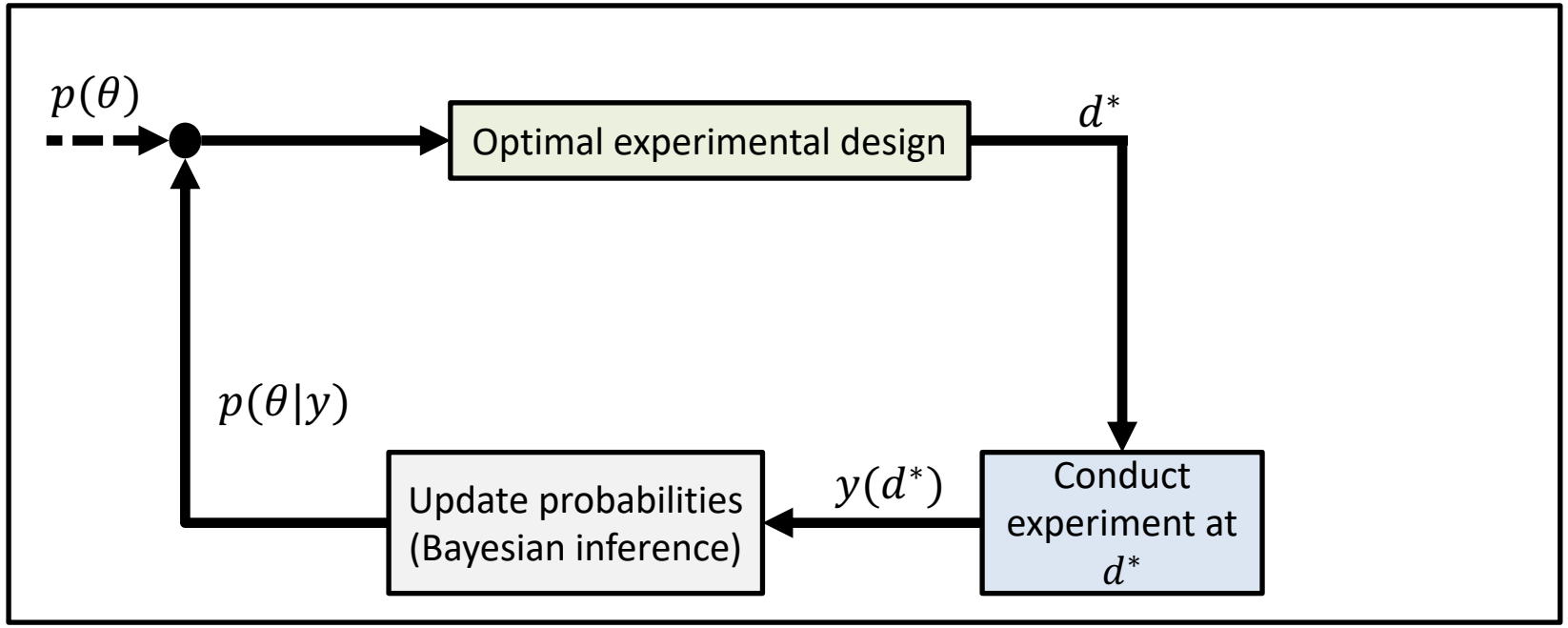
What experiment and/or test should we conduct that is **most** beneficial?

Optimal experimental design loop



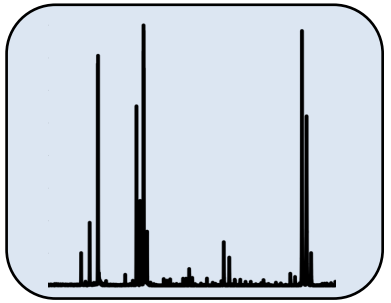
Optimal experimental design loop

$$d^* = \arg\max_{d \in \mathcal{D}} U(d)$$



Bayes' theorem

$$p(\theta|y) = \frac{p(y|\theta)p(\theta)}{p(y)}$$



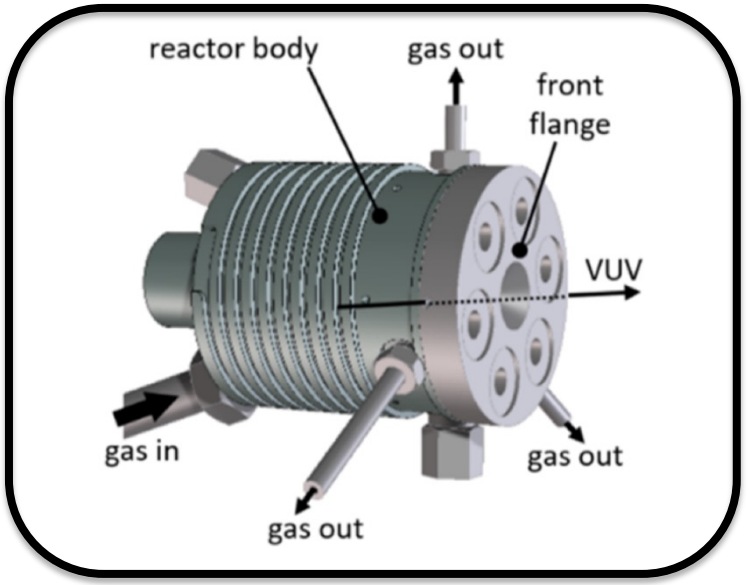
Experimental overview

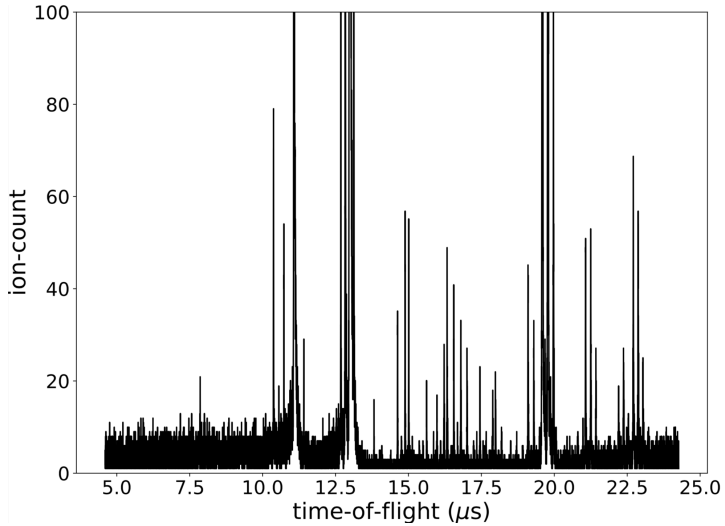
Objective: study the low-temperature oxidation of propane

- Measure the evolution of highly reactive intermediates and products

High-pressure photolysis reactor (HPR) experiment

- Premixed mixture flows into a constant pressure reactor
- Photolysis laser fires instantaneously irradiating the gas mixture
  - Chemical precursor breaks down initiating reactions
- Gas mixture exhausts out, sampled by a synchrotron tunable vacuum-ultraviolet (VUV) photoionization mass spectrometer
  - Measurement of time-of-flight mass spectrum





Time-of-flight mass spectrum at a fixed VUV energy (11.3 eV) and kinetic time (60 ms)

L. Sheps, I. Antonov, K. Au. Sensitive mass spectrometer for time-resolved gas-phase chemistry studies at high pressures. *The Journal of Physical Chemistry A* 123.50 (2019) 10804-10814.

Motivation

- Operation of the real experiment is costly and laborious
  - Potential setup time for the apparatus
  - Calibration of the apparatus and instrumentation
- Limited time to run experiments
  - Advanced Light Source, Lawrence Berkeley National Laboratory

Can we use a computational model to identify which experimental conditions are expected to be most informative?

Modeling the HPR experiment

Data model:

$$y(\boldsymbol{d}, \boldsymbol{x}) = \xi(\boldsymbol{d}, \boldsymbol{x}) + \epsilon(\boldsymbol{d}, \boldsymbol{x})$$

$$y(\boldsymbol{d}, \boldsymbol{x}) = f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) + \delta(\boldsymbol{x}) + \epsilon(\boldsymbol{d}, \boldsymbol{x})$$

$$\boldsymbol{x} = [\tau, t, E], \boldsymbol{\theta} \in \mathbb{R}^{1151}$$

$$\delta(\boldsymbol{x}) \sim GP(\mu_\delta(\boldsymbol{x}), \Sigma_\delta(\boldsymbol{x}, \boldsymbol{x}')), \epsilon(\boldsymbol{x}) \sim \mathcal{N}\left(0, s(\boldsymbol{x})^2\right)$$

$\boldsymbol{d}$: design conditions

$\boldsymbol{\theta}$: model parameters

$\boldsymbol{x}$: spatial/temporal coordinates

$y(\boldsymbol{d}, \boldsymbol{x})$: ion-count data

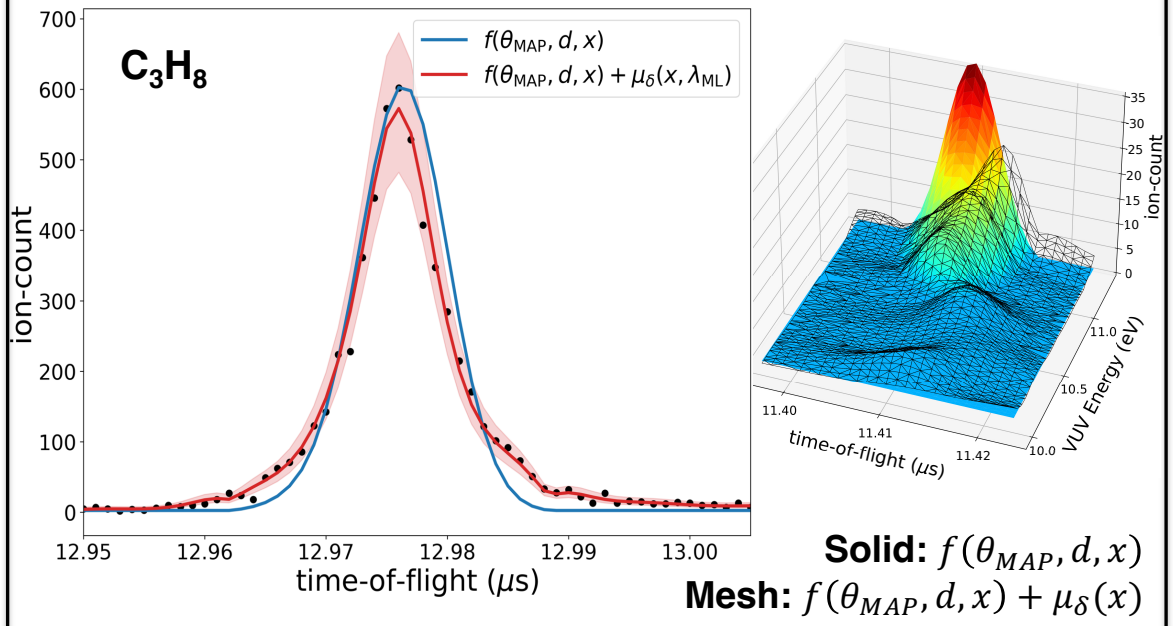
$\xi(\boldsymbol{d}, \boldsymbol{x})$: true physical process

$f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x})$: physics and instrument model

$\delta(\boldsymbol{x})$: model error

$\epsilon(\boldsymbol{d}, \boldsymbol{x})$: observation noise

- **Physics model**
  - Batch reactor model
  - Photolysis laser and ionization model [1]
- **Chemical model**
  - C0 - C3 chemical mechanism
  - 173 species / 1146 reactions [2]
- **Instrument model**
  - Maps concentrations to ion counts
  - Peaks idealized as Gaussian distributions



Solid: $f(\theta_{MAP}, d, x)$

Mesh: $f(\theta_{MAP}, d, x) + \mu_\delta(x)$

Predictions with model error provides additional fidelity to the physics model while being more inline with the data

[1] Oreluk, et al. *Combustion Theory and Modelling*, 2022.

[2] Miller, et al. *Progress in Energy and Combustion Science*, 2021.

Bayesian optimal experimental design

Objective

Find a set of experimental conditions that maximizes the expected utility $U(d)$

- Utility function models and compares the desirability of outcomes
- Target experiments to learn specific chemical rate constant measurements

$$d^* = \arg\max_{d \in \mathcal{D}} U(d)$$

$$\text{where,} \quad U(d) = \int_{y \in \mathcal{Y}} \int_{\theta \in \Theta} u(y, d, \theta) p(\theta, y|d) \mathrm{d}\theta \mathrm{dy}$$

$$= \int_{y \in \mathcal{Y}} \int_{\theta \in \Theta} u(y, d, \theta) p(\theta|y, d) p(y|d) \mathrm{d}\theta \mathrm{dy}$$

Notation

d : design conditions

θ : model parameters

y : data

Expected utility involves a double integral over data and model parameters.
Necessitates a model for the physical system and for the experimental instrument.
Uses Bayesian posterior on model parameters, and Bayesian model evidence on data.

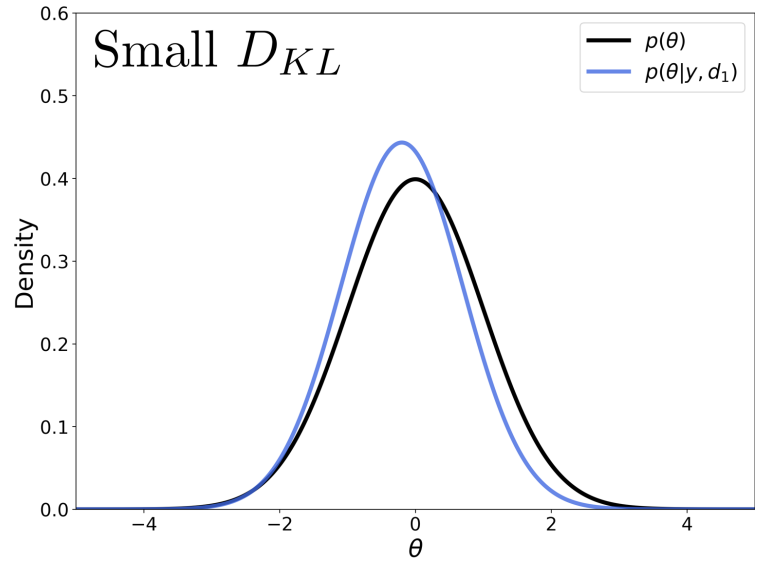
D.V. Lindley, On a measure of the information provided by an experiment, *The Annals of Mathematical Statistics* 27 (1956) 986–1005.
K. Chaloner and I. Verdinelli, Bayesian experimental design: A review. *Statistical Science* (1995) 273-304.

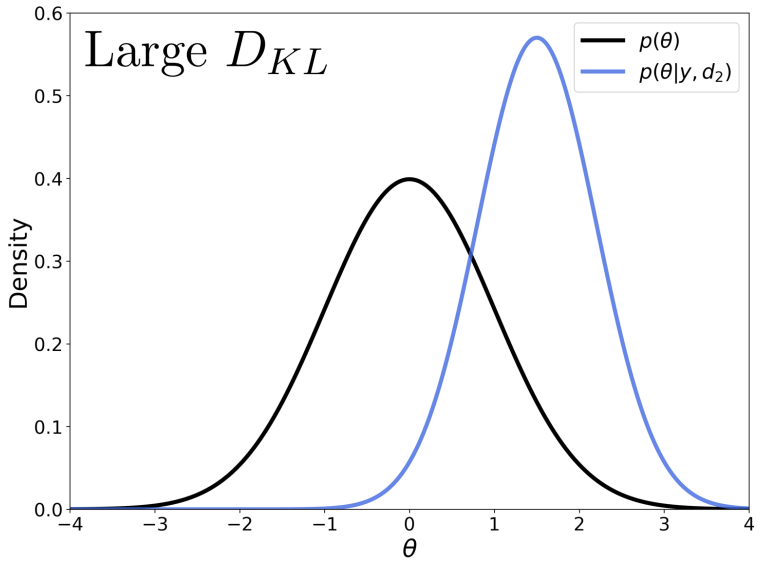
Selecting a utility function, $u(y, d)$

Utility function should *reflect the goal or purpose* of the experiment

1) Improve the estimate of a unknown model parameter
   - Information gain of an experiment is closely related to minimizing the parameter uncertainty

$$u(y,d) = D_{\mathrm{KL}}(p(\theta|y,d)||p(\theta)) = \int p(\theta|y,d) \log \left[\frac{p(\theta|y,d)}{p(\theta)} \right] \mathrm{d}\theta$$





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2) Improve the predictivity of a quantity of interest, q
   - Measure divergence between prior predictive and posterior predictive distributions

$$p(q) = \int_{\theta\in\Theta} p(q|\theta)p(\theta)\mathrm{d}\theta$$

$$p(q|y,d) = \int_{\theta\in\Theta} p(q|\theta)p(\theta|y,d)\mathrm{d}\theta$$

$$\begin{aligned} u(y,d) &= D_{\mathrm{KL}}(p(q|y,d)||p(q)) \\ &= \int p(q|y,d)\log\left[\frac{p(q|y,d)}{p(q)}\right]\mathrm{d}\theta \end{aligned}$$

Selecting a utility function, $u(y, d)$

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$$U(d) = \int_{\mathcal{Y}} \left(\int_{\Theta} p(\theta|y,d) \log\left[\frac{p(\theta|y,d)}{p(\theta)}\right] d\theta \right) p(y|d) dy$$

$$U(d) = \int_{\mathcal{Y}} \int_{\Theta} \left[\log p(y|\theta,d) - \log p(y|d)\right] p(y|\theta,d) p(\theta) d\theta dy$$

Approximating the expected utility

Numerical approximation:

$$U(d) \approx \frac{1}{N}\sum_{i=0}^{N}\left[\log p(y^{(i)}|\theta^{(i)},d) - \log \underbrace{p(y^{(i)}|d)}_{???}\right] \quad \text{where, } \theta^{(i)} \sim p(\theta),\ y^{(i)} \sim p(y|\theta^{(i)},d)$$

Nested Monte Carlo

$$U(d) \approx \frac{1}{N}\sum_{i=0}^{N}\left[\log p(y^{(i)}|\theta^{(i)},d) - \log\left(\frac{1}{M}\sum_{j=0}^{M} p(y^{(i)}|\theta^{(j)},d)\right)\right]$$

$$d^* = \arg\max_{d\in\mathcal{D}} U(d)$$

- *Costly to evaluate* $U(d)$
- $\frac{\mathrm{d}U}{\mathrm{d}d} = \ ?$

T. Rainforth et al., On nesting Monte Carlo estimators, *International Conference on Machine Learning*. PMLR, 2018.
K.J. Ryan, Estimating expected information gains for experimental designs with application to the random fatigue-limit model, *Journal of Computational and Graphical Statistics* 12 (2003) 585–603.

Maximizing the expected utility, $U(d)$

$$d^* = \arg\max_{d \in \mathcal{D}} U(d)$$

Bayesian Optimization

- Construct a GP of the unknown objective function

$$U(d) \sim \mathcal{N}\left(\mu\left(d\right), K\left(d, d'\right)\right)$$

- Use *acquisition function* $\alpha(d)$ to select new samples
  - Gaussian Process upper confidence bound (UCB)

$$\alpha_t\left(d\right) = \mu_{t-1}\left(d\right) + \sqrt{\beta}_t \sigma_{t-1}\left(d\right)$$

$$\sigma_{t-1}\left(d\right) = \sqrt{K\left(d, d\right)}$$

  - Exploits regions with a high mean and explores regions of high uncertainty

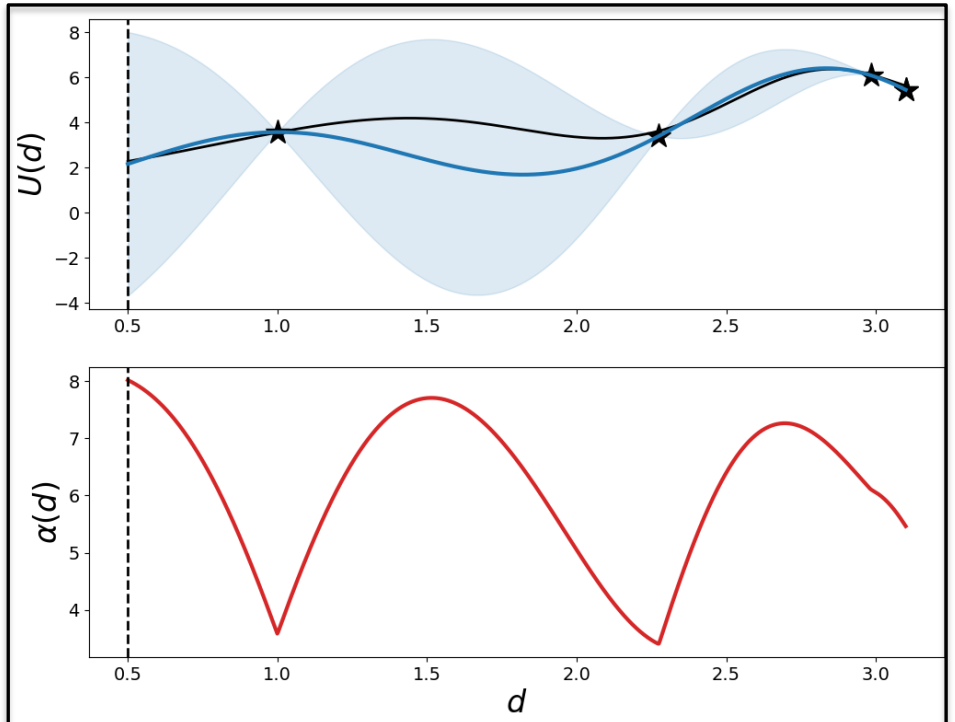

Illustration of an iteration of Bayesian optimization, unknown objective function (top), acquisition function (bottom)

- Select next optimization iteration at: $d_t = \arg\max_{d \in \mathcal{D}} \alpha_t\left(d\right)$
- Evaluate utility function at $U(d_t)$

N. Srinivas, A. Krause, S. Kakade, & M. Seeger. Gaussian process optimization in the bandit setting: No regret and experimental design. *arXiv:0912.3995* (2009).

Challenges

High-dimensional model output

- High-fidelity physics-based simulations are be expensive to evaluate
- Evaluating $U(d) \sim \mathcal{O}(NM)$ forward solves

$$U(d) \approx \frac{1}{N} \sum_{i=0}^{N} \left[\log p(y^{(i)}|\theta^{(i)}, d) - \log \left(\frac{1}{M} \sum_{j=0}^{M} p(y^{(i)}|\theta^{(j)}, d) \right) \right]$$

- Constructing a surrogate model addresses the costly run-time

$$f(\boldsymbol{\theta}, \boldsymbol{d}, x_i) \approx g(\boldsymbol{\theta}, \boldsymbol{d}, x_i) = \sum_{p=1}^{P} c_p \Psi_p(\xi)$$

  - Total **number of outputs** remains problematic $y(\boldsymbol{d}, \boldsymbol{x}) \in \mathbb{R}^{510{,}000{,}000}$

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Can we find *low-dimensional* representations of the high-dimensional output?

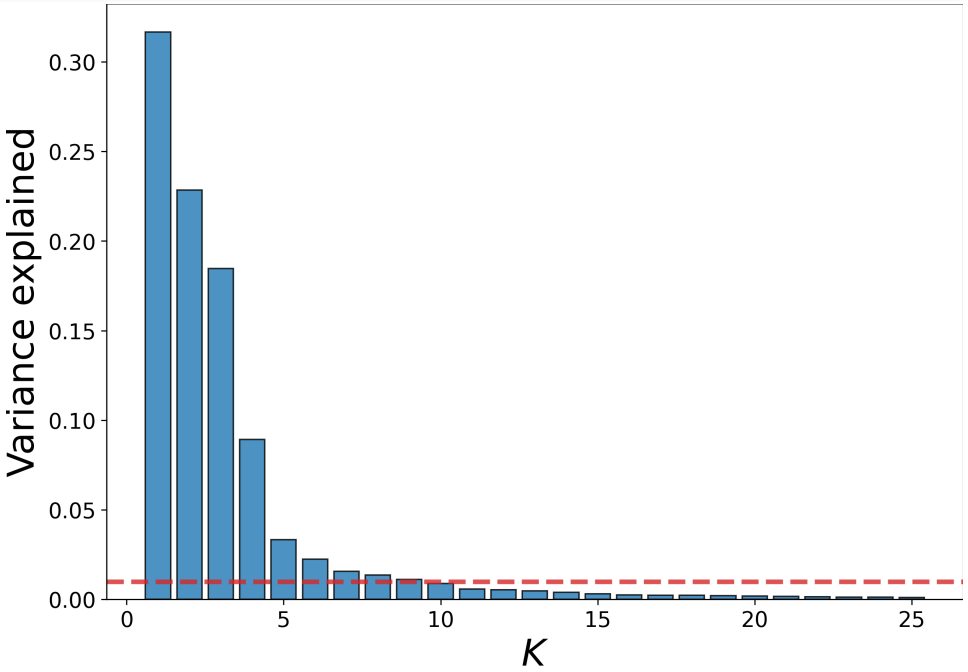
Reducing output dimensionality

Goal: Map high-dimensional model output to a lower-dimensional space while minimizing loss of information

Truncated SVD

At a fixed design d,

- Draw n samples, where $\boldsymbol{\theta}^{(i)} \sim p(\boldsymbol{\theta})$
- Evaluate the model $f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) + \delta(\boldsymbol{\lambda}_{\mathrm{ML}}, \boldsymbol{x}) + \epsilon(\boldsymbol{x})$
- Construct output matrix $\boldsymbol{Y} = \boldsymbol{U}\boldsymbol{S}\boldsymbol{V}^T$, where $\boldsymbol{Y} \in \mathbb{R}^{n \times J}$, $J = 5.1 \times 10^8$
- Retain only top K singular values of $\boldsymbol{S}$
- Low-rank approximation: $\boldsymbol{Y}_K = \boldsymbol{U}_K \boldsymbol{S}_K \boldsymbol{V}_K^T$



93% of the total variance is explained by a small number ($K = 10$) components at 620 K, 2 bar, $n_{\mathrm{C3H8}} = 2\mathrm{e}14, n_{\mathrm{O2}} = 8\mathrm{e}18,\ n_{\mathrm{Cl}} = 1\mathrm{e}13$.

Transformation:

$$q(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) = y(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x})\, \boldsymbol{V}_K$$

$$\underbrace{q(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x})}_{(1 \times K)} = \underbrace{[f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) + \delta(\boldsymbol{\lambda}_{\mathrm{ML}}, \boldsymbol{x}) + \epsilon(\boldsymbol{x})]}_{(1 \times J)} \underbrace{\boldsymbol{V}_K}_{(J \times K)}$$

Challenges: Target *specific* model parameters

Expected utility measures difference between $p(\theta)$ and $p(\theta|y)$, in other words, difference in ***all uncertain*** *parameters*

- Only interested in designs that improve *particular chemical rates*
  - chemical model contains 1146 uncertain chemical rates

Set of targeted reaction rates

Reaction ID	
	n-R + O2 ⟶ QOOH3
1040	O2 + CH3CH2CH2 ⟶ CH3CHCH2OOH
	n-R + O2 ⟶ QOOH1
1041	O2 + CH3CH2CH2 ⟶ CH2CH2CH2OOH
1042	O2 + CH3CH2CH2 ⟶ CH2CH2CH2OOH
	n-R + O2 ⟶ propene
1043	O2 + CH3CH2CH2 ⟶ HO2 + CH3CHCH2
	n-R + O2 ⟶ propylene oxide
1044	O2 + CH3CH2CH2 ⟶ OH + C-CH2OCH(CH3)
	n-RO2 ⟶ products
1045	CH3CH2CH2OO ⟶ CH3CHCH2OOH
1046	CH3CH2CH2OO ⟶ CH2CH2CH2OOH
1047	CH3CH2CH2OO ⟶ CH2CH2CH2OOH
1048	CH3CH2CH2OO ⟶ HO2 + CH3CHCH2
1049	CH3CH2CH2OO ⟶ OH + C-CH2OCH(CH3)
	QOOH3 ⟶ products
1051	CH3CHCH2OOH ⟶ HO2 + CH3CHCH2
1052	CH3CHCH2OOH ⟶ OH + C-CH2OCH(CH3)
	QOOH1 ⟶ propene
1053	CH2CH2CH2OOH ⟶ HO2 + CH3CHCH2
	i-R+O2 ⟶ products
1056	O2 + CH3CHCH3 ⟶ CH3CH(OOH)CH2
1057	O2 + CH3CHCH3 ⟶ HO2 + CH3CHCH2
1058	O2 + CH3CHCH3 ⟶ OH + C-CH2OCH(CH3)
	i-RO2 ⟶ products
1059	CH3CH(OO)CH3 ⟶ CH3CH(OOH)CH2
1060	CH3CH(OO)CH3 ⟶ HO2 + CH3CHCH2
1061	CH3CH(OO)CH3 ⟶ OH + C-CH2OCH(CH3)
	QOOH2 ⟶ products
1062	CH3CH(OOH)CH2 ⟶ HO2 + CH3CHCH2
1063	CH3CH(OOH)CH2 ⟶ OH + C-CH2OCH(CH3)
	O2 + QOOH1 ⟶ products
1065	O2 + CH2CH2CH2OOH ⟶ OHOCH2CH2CH2OO
	O2 + QOOH3 ⟶ products
1069	O2 + CH3CHCH2OOH ⟶ CH3CH(OO)CH2OOH
1071	O2 + CH3CHCH2OOH ⟶ CH2CH(OOH)CH2OOH
	O2 + QOOH2 ⟶ products
1080	O2 + CH3CH(OOH)CH2 ⟶ CH3CH(OOH)CH2OO
1081	O2 + CH3CH(OOH)CH2 ⟶ CH2CH(OOH)CH2OOH
	3−hydroperoxy−propylperoxyradical ⟶ KHP
1088	OHOCH2CH2CH2OO ⟶ OH + OCHCH2CH2OOH
	KHP ⟶ products
1112	OCHCH2CH2OOH ⟶ OCHCH2CH2O + OH

Challenges: Target *specific* model parameters

Expected utility measures difference between $p(\theta)$ and $p(\theta|y)$, in other words, difference in ***all uncertain*** *parameters*

- Only interested in designs that improve *particular chemical rates*
  - chemical model contains 1146 uncertain chemical rates

Solution: Reformulate utility to marginalize over nuisance parameters

Let θ be the parameters of interest and η be the nuisance parameters

Nested Monte Carlo estimator becomes:

$$U(d) \approx \frac{1}{N}\sum_{i=1}^{N}\left[\log\left(\frac{1}{K}\sum_{k=1}^{K} p\left(y^{(i)}|\theta^{(i)},\eta^{(k)},d\right)\right) - \log\left(\frac{1}{M}\sum_{j=1}^{M} p\left(y^{(i)}|\theta^{(j)},\eta^{(j)},d\right)\right)\right]$$

Additional loop in likelihood to marginalize over nuisance parameters

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	O2 + QOOH3 ⟶ products
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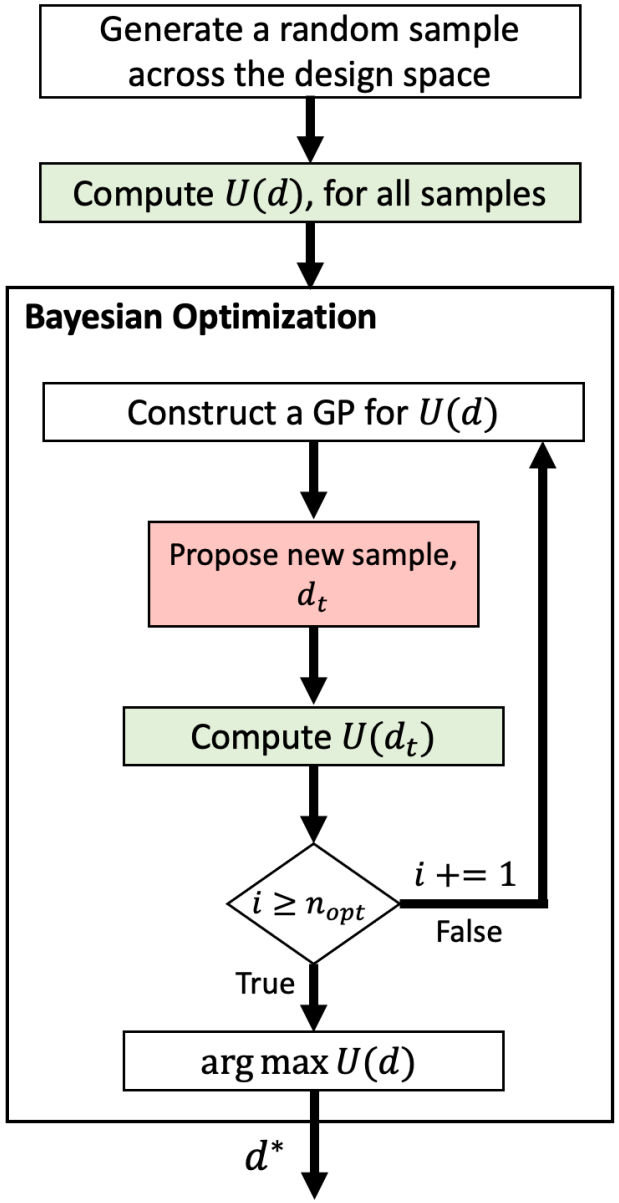
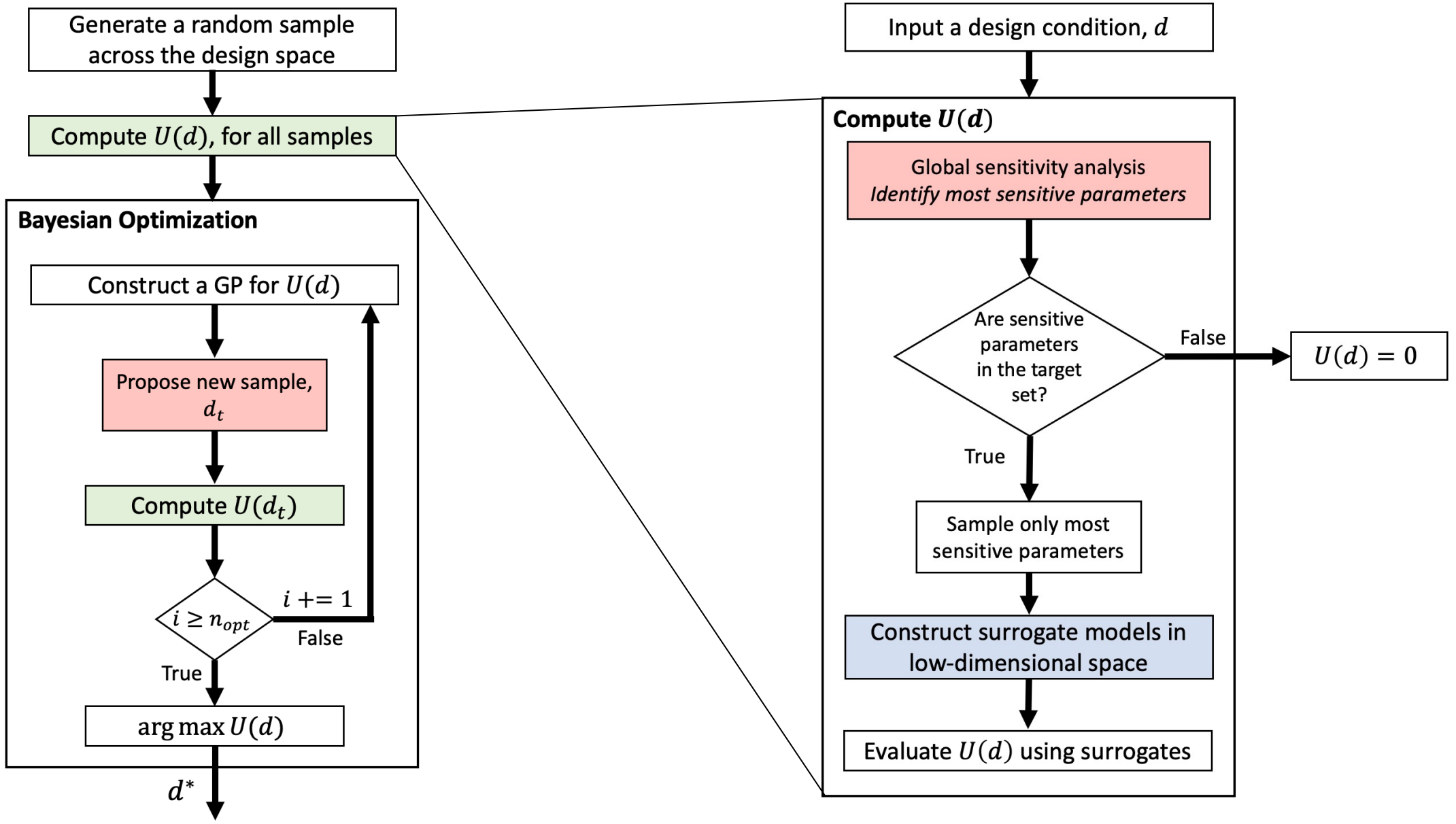
Feng, Chi, and Youssef M. Marzouk. "A layered multiple importance sampling scheme for focused optimal Bayesian experimental design." *arXiv preprint arXiv:1903.11187* (2019).

Targeted OED workflow



Targeted OED workflow



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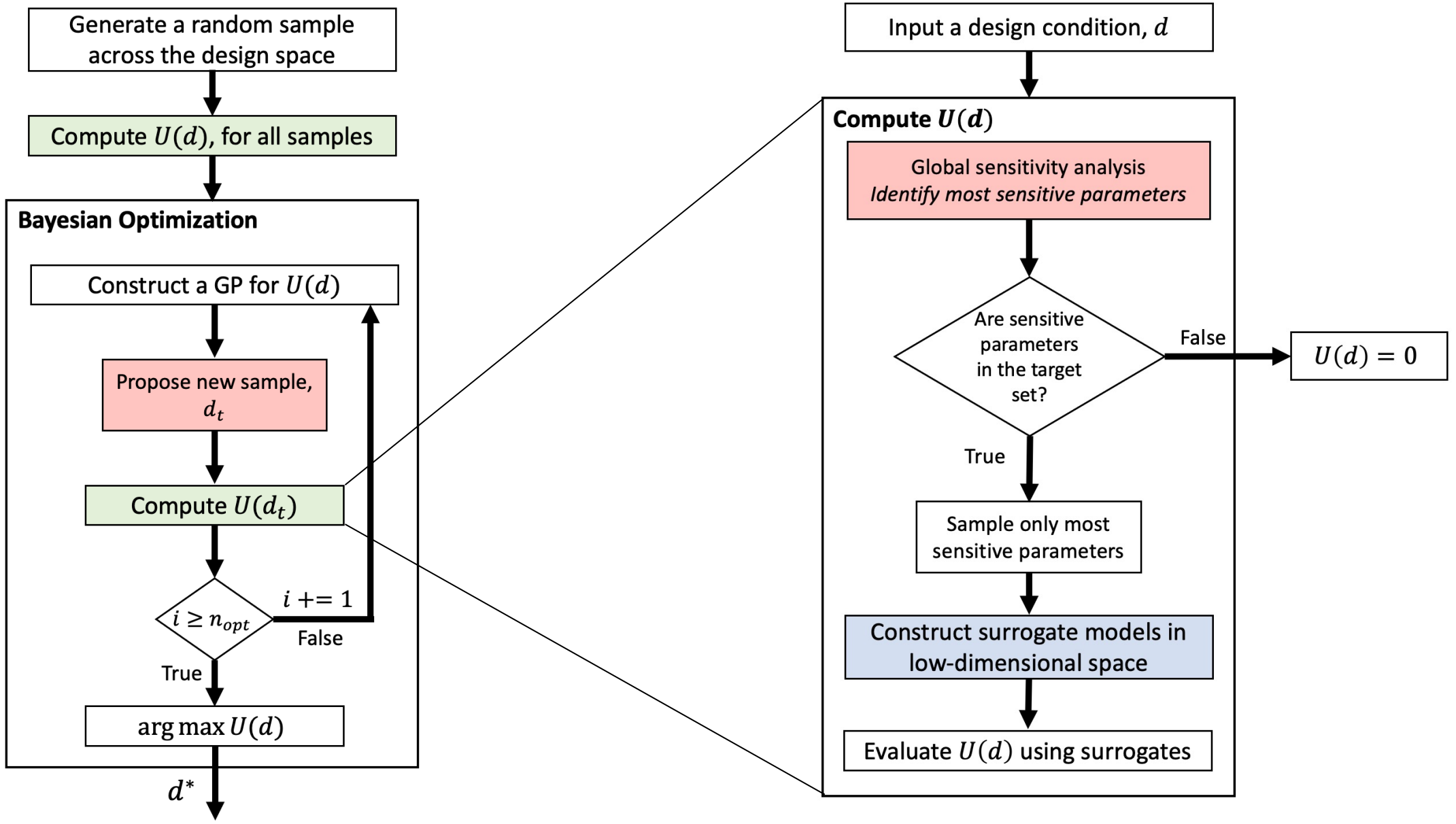

Results

Original data model,

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$$y(\boldsymbol{d}, \boldsymbol{x}) = f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) + \delta(\boldsymbol{x}) + \epsilon(\boldsymbol{d}, \boldsymbol{x})$$
$$\boldsymbol{x} = [\tau, t, E], \boldsymbol{\theta} \in \mathbb{R}^{1151}$$
$$\delta(\boldsymbol{x}) \sim GP(\mu_\delta(\boldsymbol{x}), \Sigma_\delta(\boldsymbol{x}, \boldsymbol{x}')), \epsilon(\boldsymbol{x}) \sim \mathcal{N}\left(0, s(\boldsymbol{x})^2\right)$$

Design space: $\boldsymbol{d} = [T, p, n_{Cl}, n_{O2}]$

$$\Sigma_\delta = \begin{bmatrix} \Sigma_1 & & & & & \\ & \ddots & & & & \\ & & \Sigma_D & & & \\ & & & \ddots & & \\ & & & & \Sigma_{Dt_e+1} & \\ & & & & & \ddots \\ & & & & & & \Sigma_{D(t_e+1)} \end{bmatrix}$$

Assumptions:

- **Model error $\boldsymbol{\delta(x)}$ independent of $\boldsymbol{d}$**
- Local GPs constructed independently for each kinetic time
- Considering all 1151 model parameters as uncertain
  - Designs are constructed to target specific sets of the model parameters
- Prior parameter uncertainty is specified by literature

Results

Original data model,

$$y(\boldsymbol{d}, \boldsymbol{x}) = \xi(\boldsymbol{d}, \boldsymbol{x}) + \epsilon(\boldsymbol{d}, \boldsymbol{x})$$

$$y(\boldsymbol{d}, \boldsymbol{x}) = f(\boldsymbol{\theta}, \boldsymbol{d}, \boldsymbol{x}) + \delta(\boldsymbol{x}) + \epsilon(\boldsymbol{d}, \boldsymbol{x})$$

$$\boldsymbol{x} = [\tau, t, E], \boldsymbol{\theta} \in \mathbb{R}^{1151}$$

$$\delta(\boldsymbol{x}) \sim GP(\mu_\delta(\boldsymbol{x}), \Sigma_\delta(\boldsymbol{x}, \boldsymbol{x}')), \epsilon(\boldsymbol{x}) \sim \mathcal{N}\left(0, s(\boldsymbol{x})^2\right)$$

Design space: $\boldsymbol{d} = [T, p, n_{Cl}, n_{O2}]$

$$\Sigma_\delta = \begin{bmatrix} \Sigma_1 & & & & \\ & \ddots & & & \\ & & \Sigma_D & & \\ & & & \ddots & \\ & & & & \Sigma_{Dt_e+1} & & \\ & & & & & \ddots & \\ & & & & & & \Sigma_{D(t_e+1)} \end{bmatrix}$$

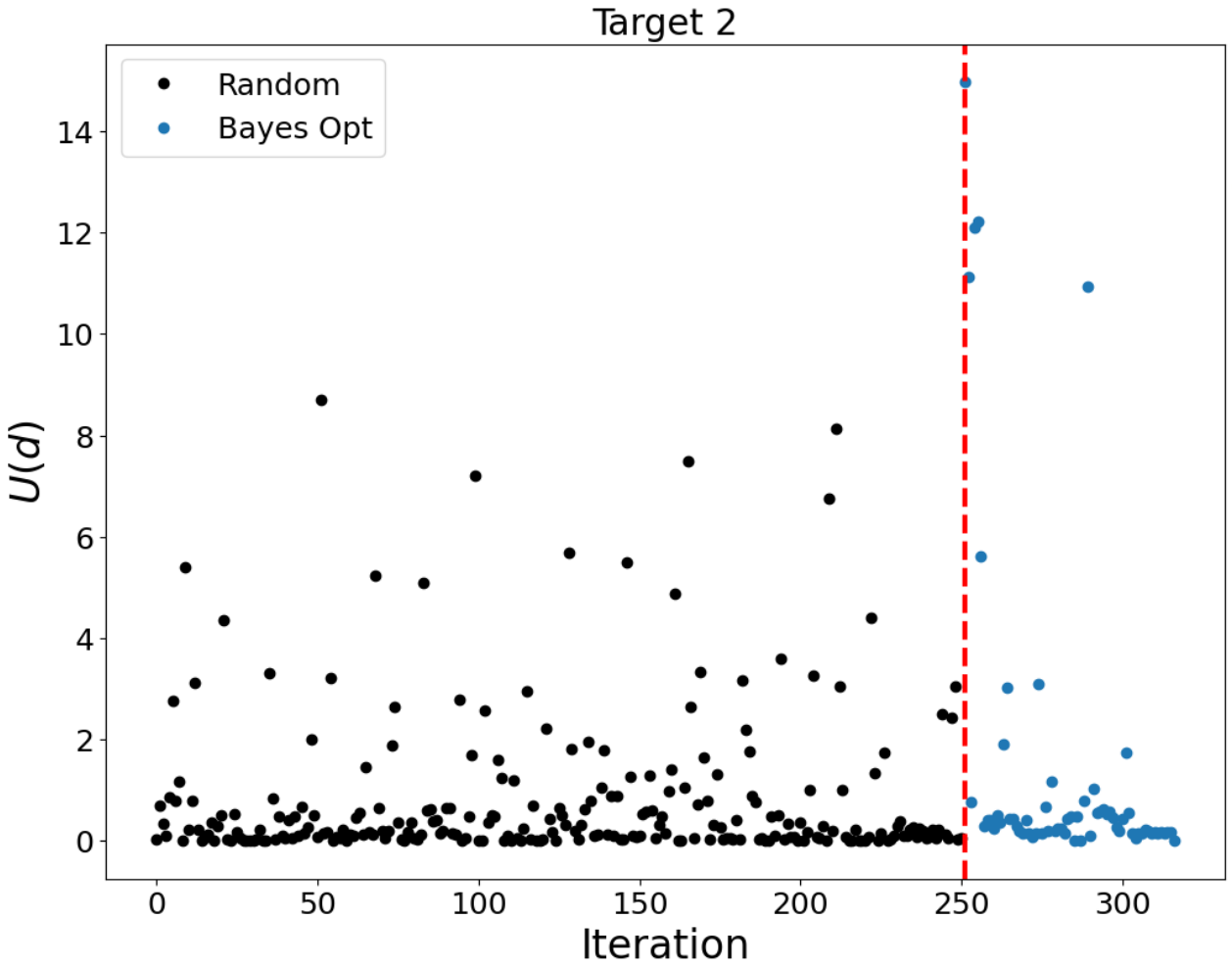
$t = 0$

$t = t_e$

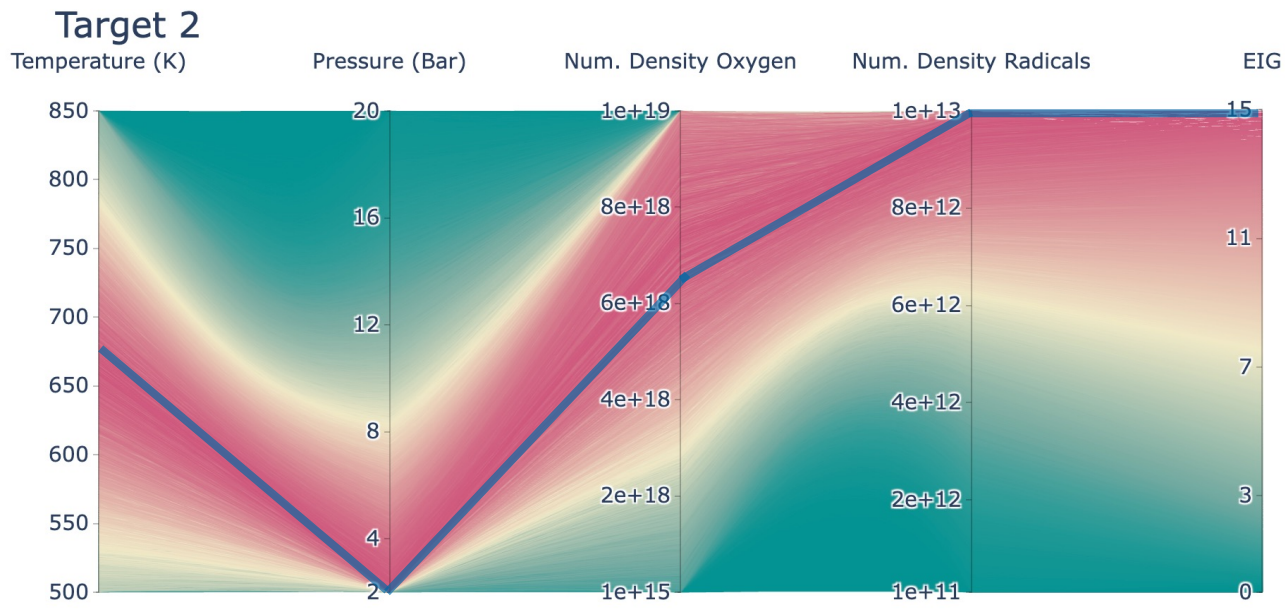
Assumptions:

- Model error $\delta(x)$ independent of d
- **Local GPs constructed independently for each kinetic time**
- Considering all 1151 model parameters as uncertain
  - Designs are constructed to target specific sets of the model parameters
- Prior parameter uncertainty is specified by literature

Results

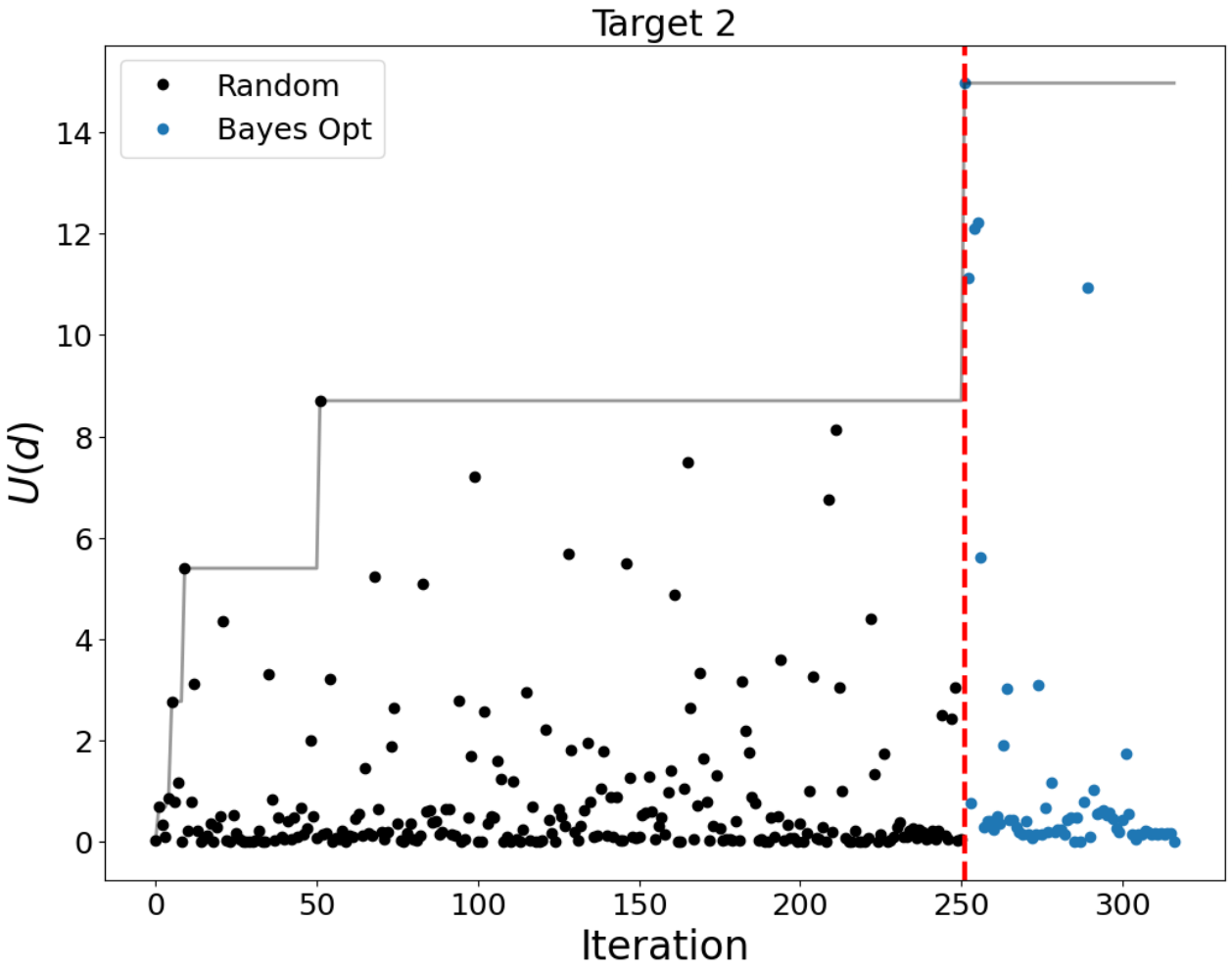


Optimization iterations for maximizing the expected utility. A number of random samples (black points) are evaluated in parallel prior to Bayesian Optimization (blue points) , which quickly improves on the previous maxima.



Parallel coordinate plot shows the range of design variables associated with high $U(d)$ in red. Trajectory of optimal design is highlighted in blue.

Results



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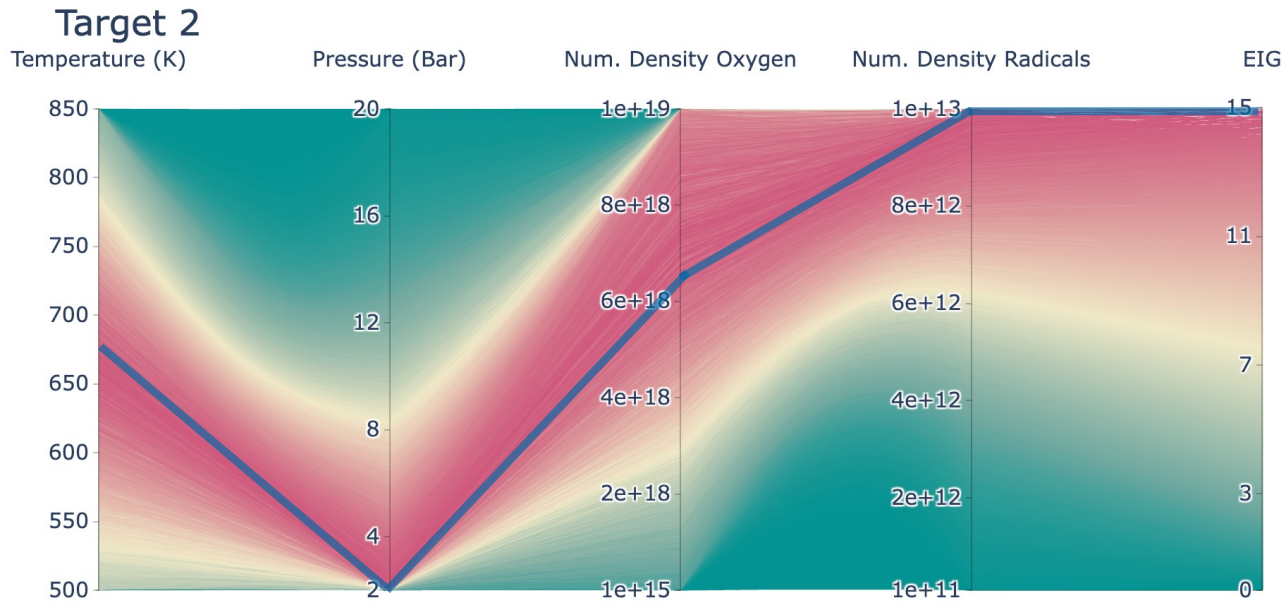

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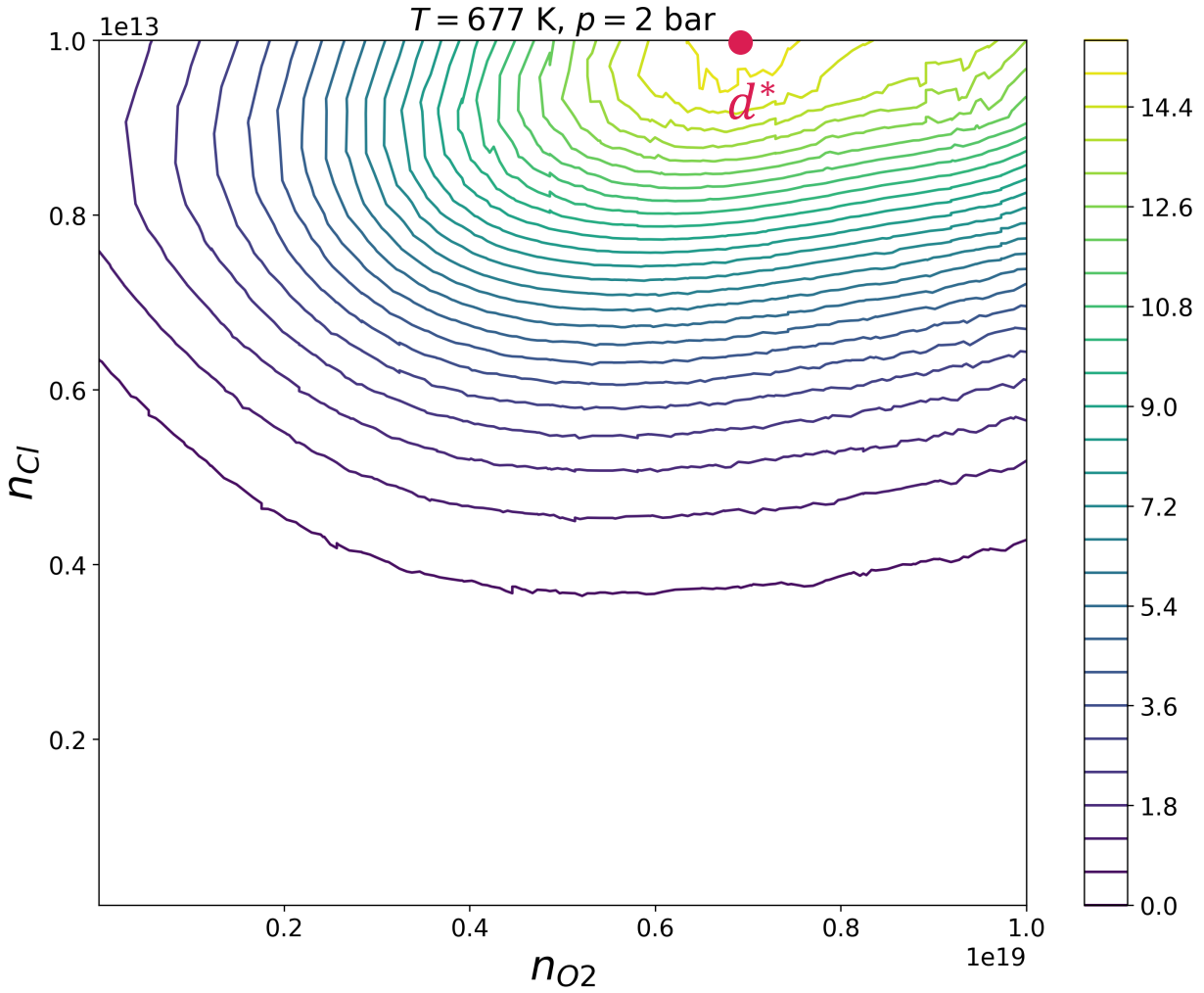
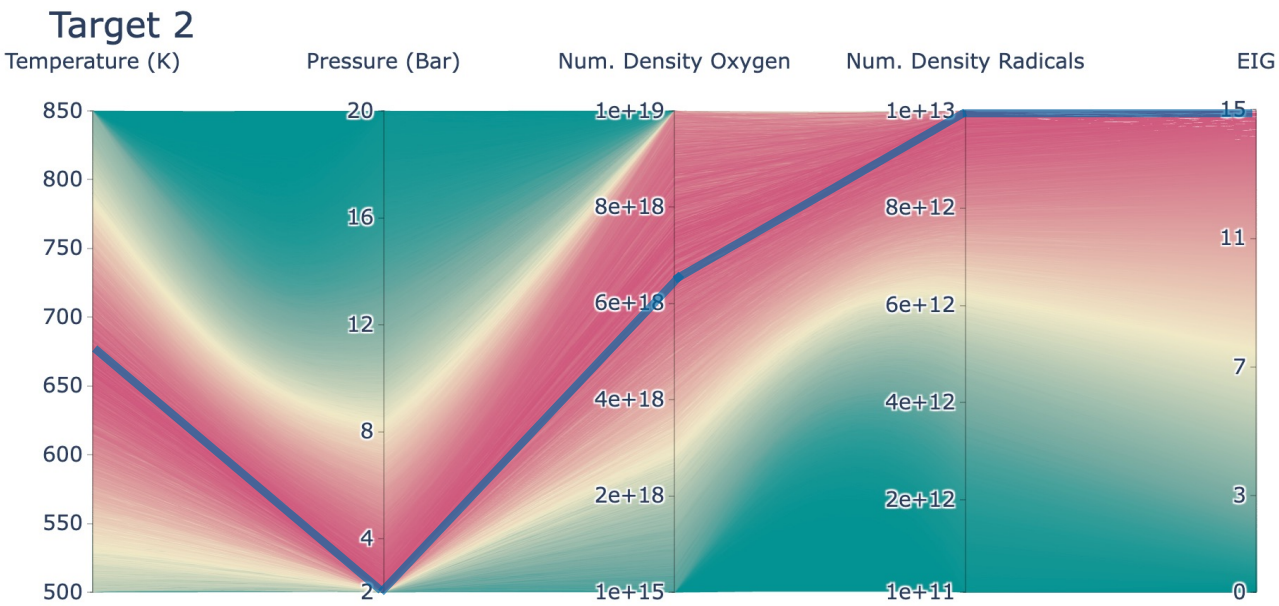


Cross-section of $U(d)$ for a fixed temperature and pressure.

Heatmap enables selection of a suboptimal design which can be desirable if resources are scarce or there are safety concerns



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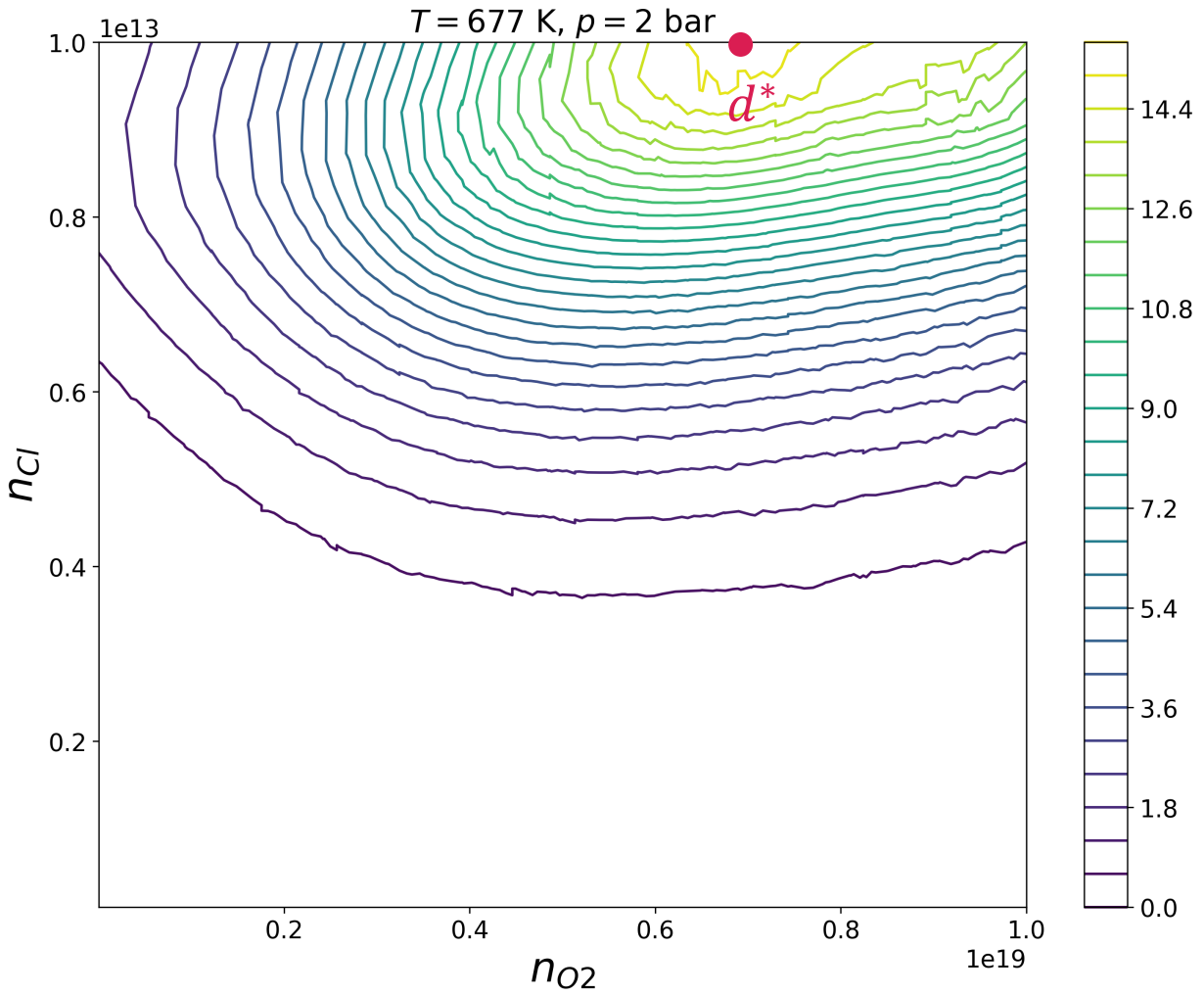


Cross-section of $U(d)$ for a fixed temperature and pressure.

Heatmap enables selection of a suboptimal design which can be desirable if resources are scarce or there are safety concerns

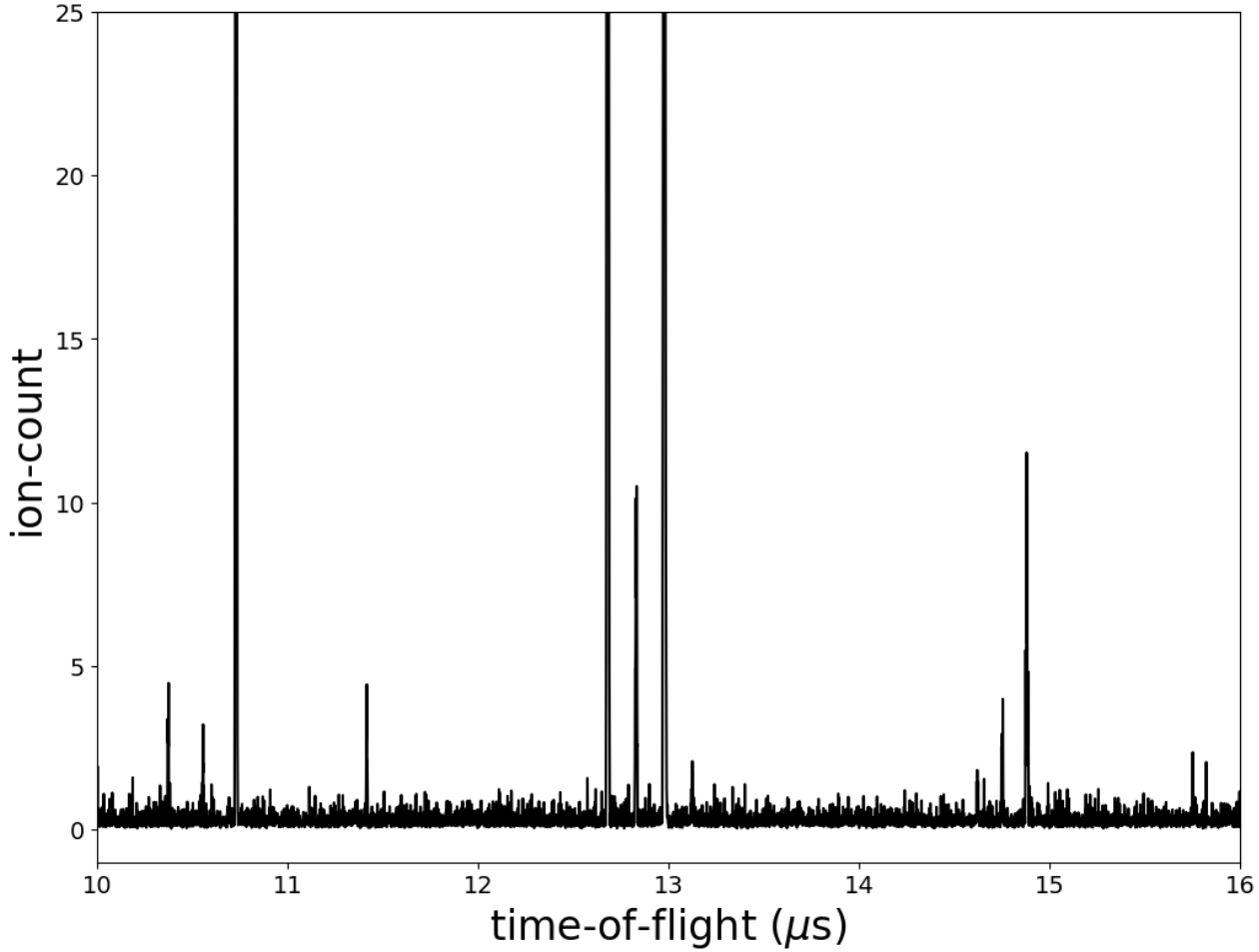
What does the posterior distribution look like after measuring data at d^*?

- Generate datasets using the probabilistic model at d^*
- Plausible posterior distributions are aggregated using logarithmic pooling:

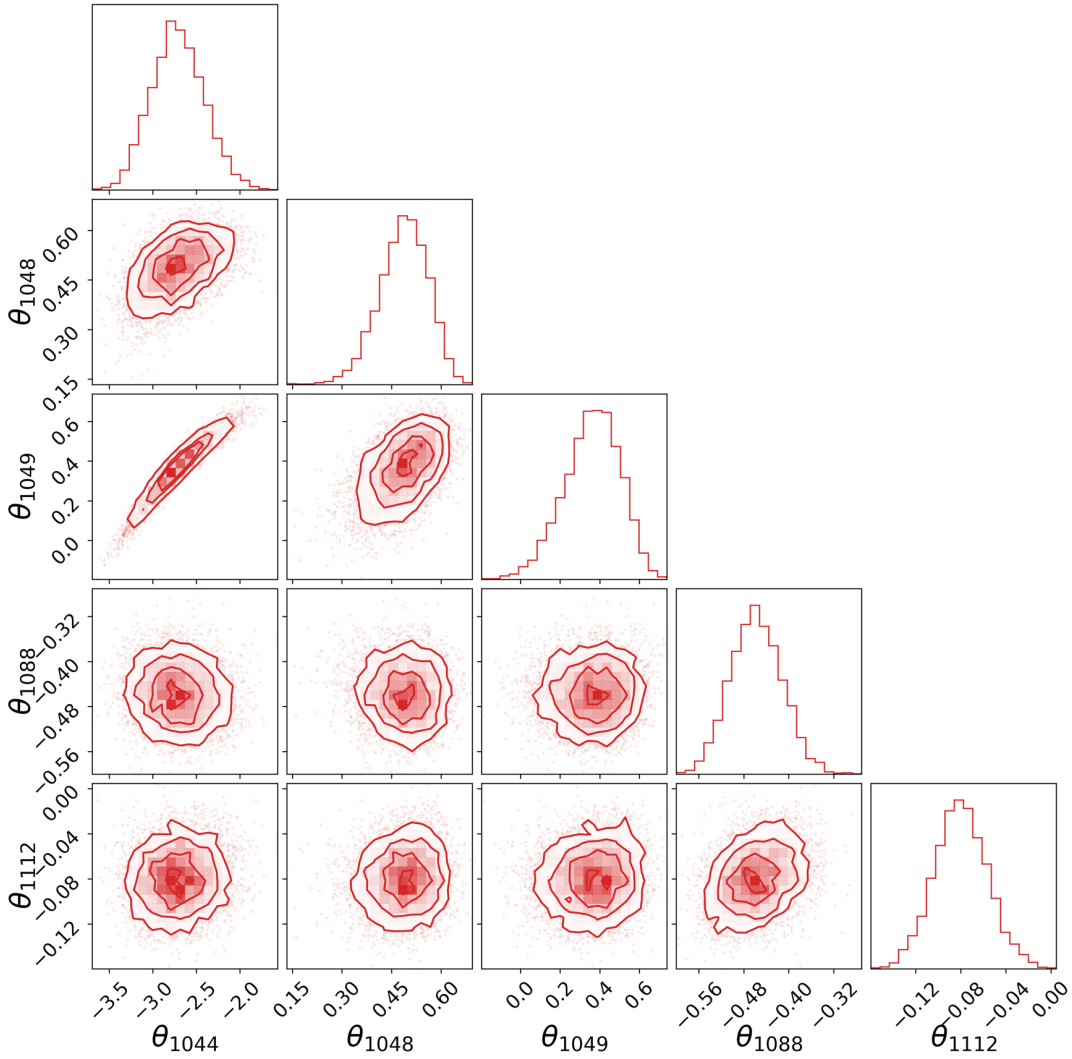
$$p(\theta)_{\log} \approx \left[\prod_{i=1}^{K} p(\theta|y_i)\right]^{1/K}$$

$$p(\theta|y_i) \propto p(\theta)p(y_i|\theta)$$

Results

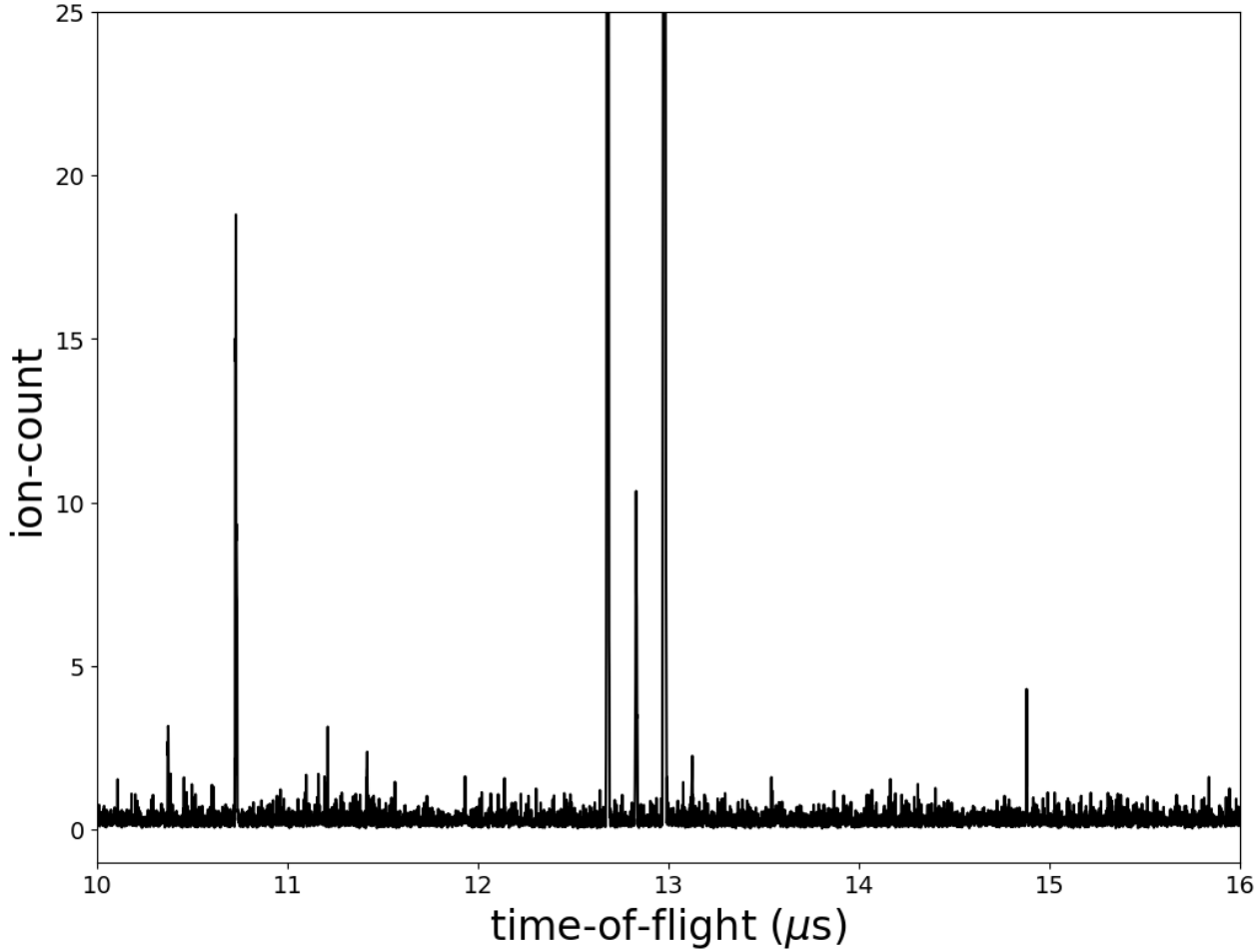


Visualization of the range of plausible datasets generated by the probabilistic model at d^* both time and VUV energy are fixed at 60ms and 11.3 eV, respectively. New species appear with significant changes to the peak magnitudes.



Estimated posterior after log pooling with $K = 10^3$ plausible datasets generated at d^*. Prior distributions are a standard normal.

Results



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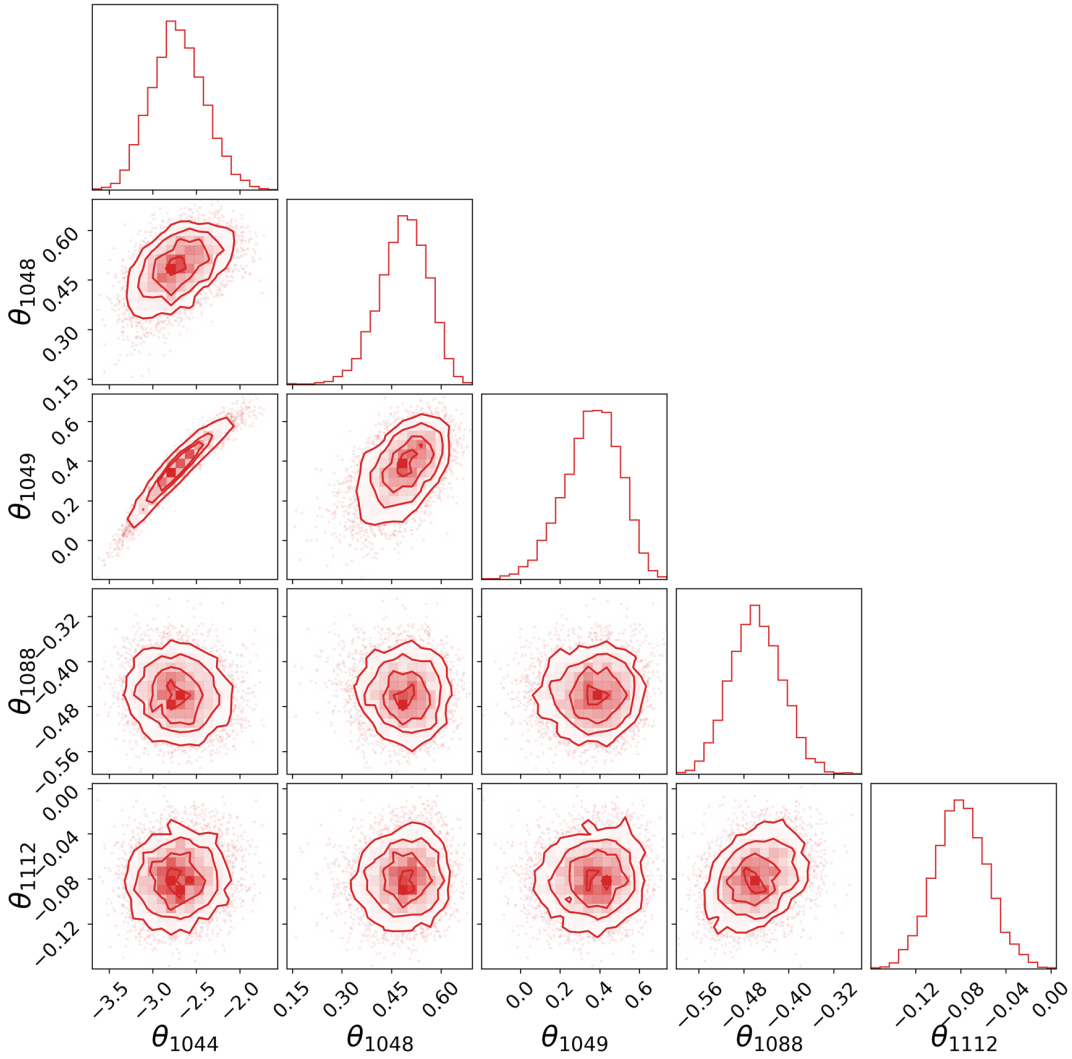

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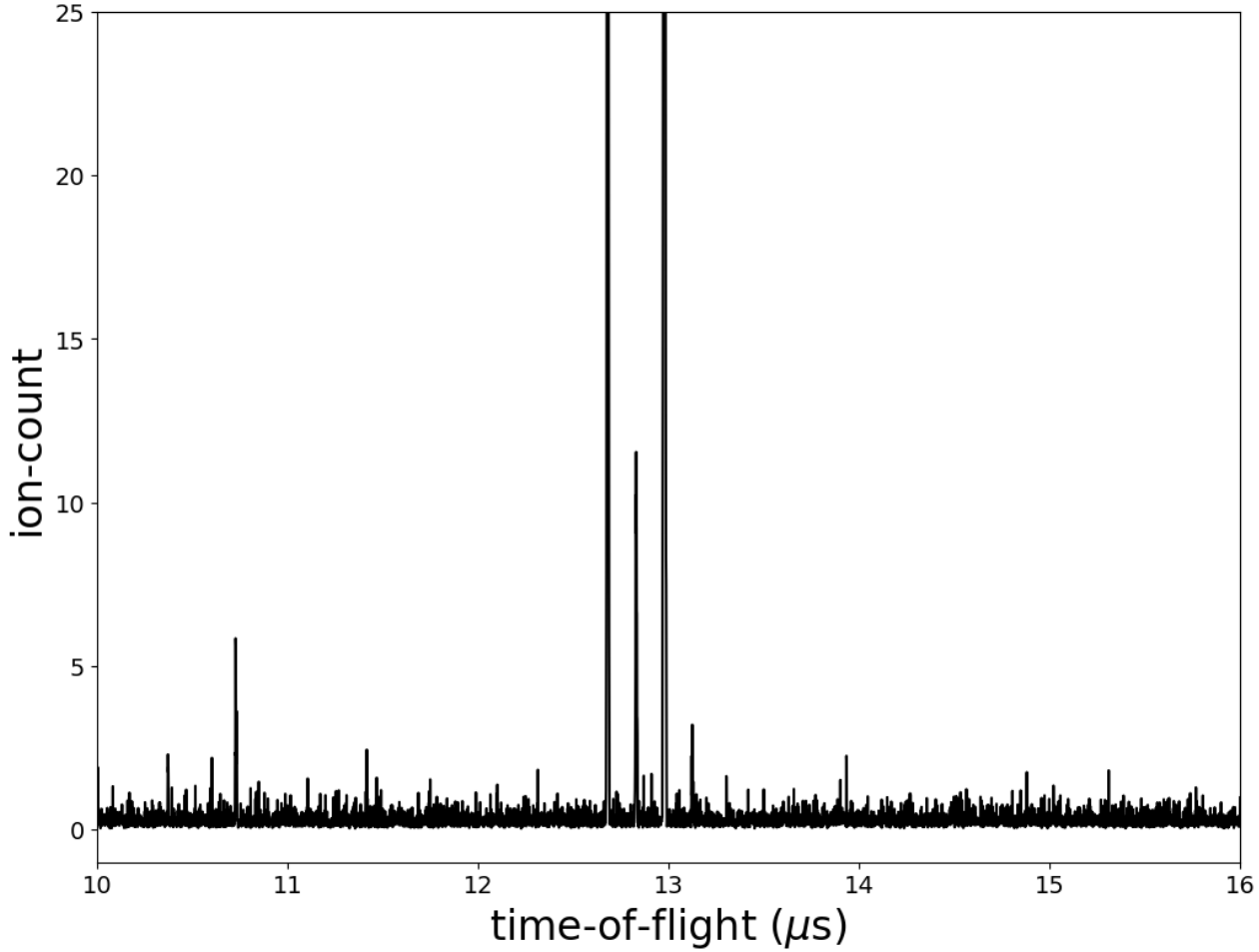

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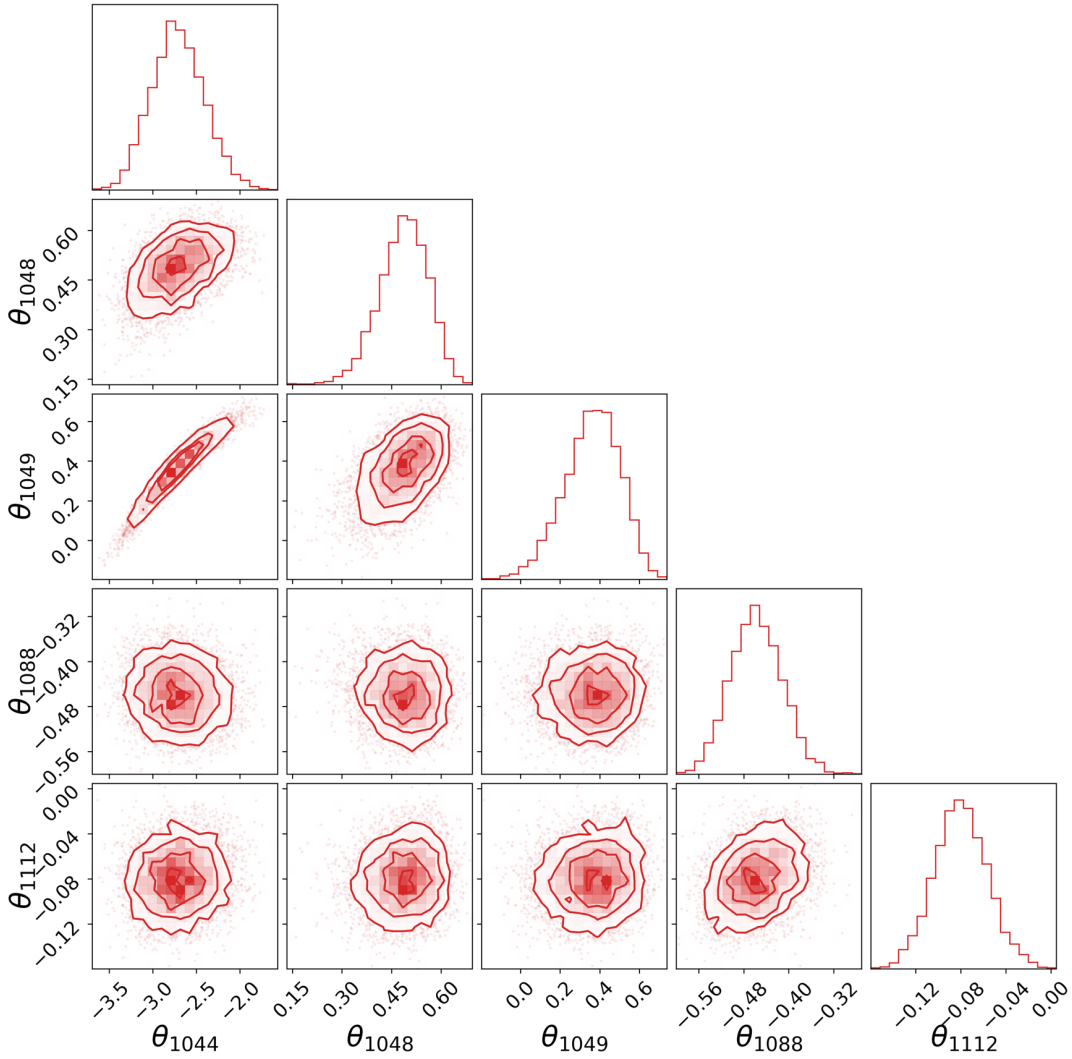

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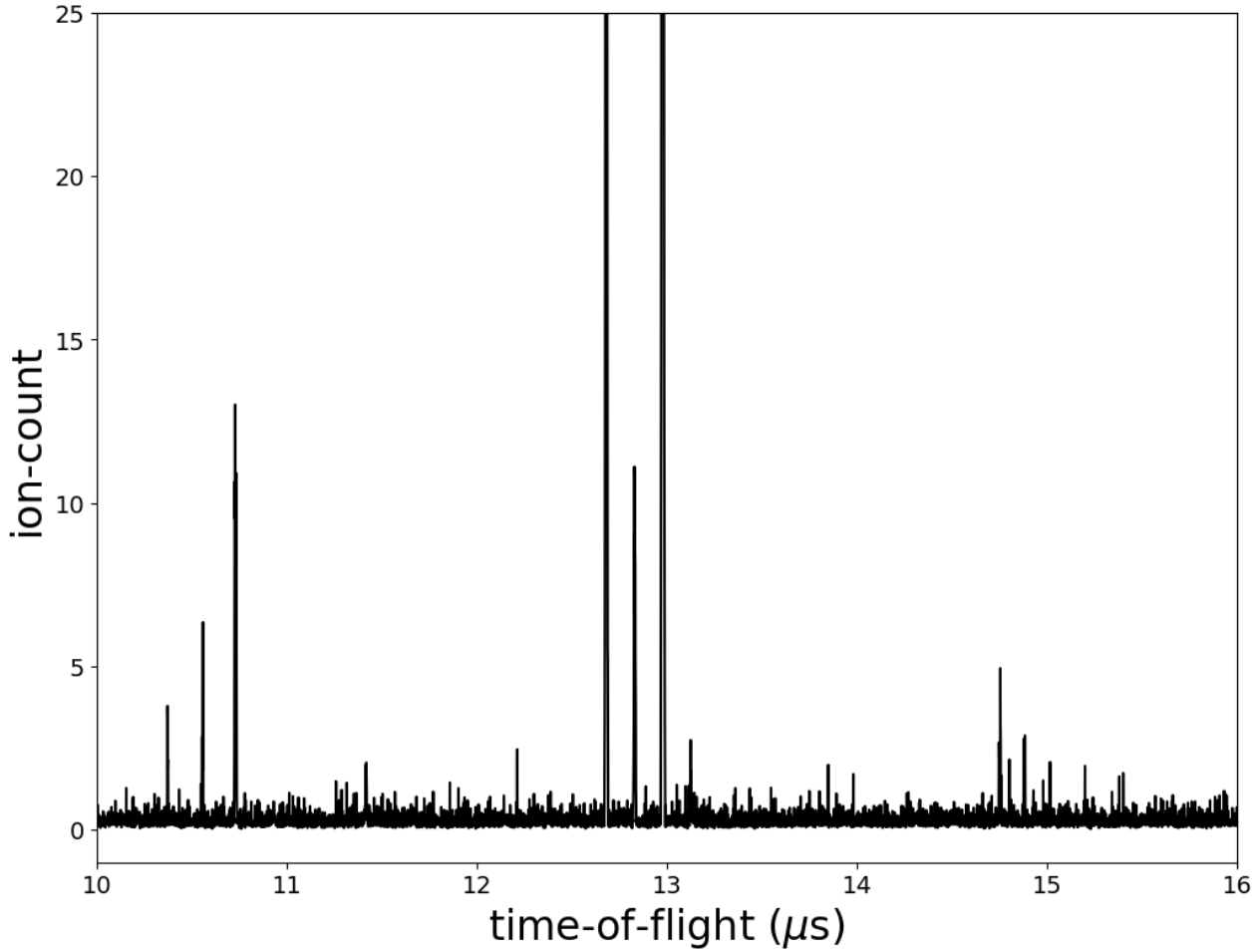

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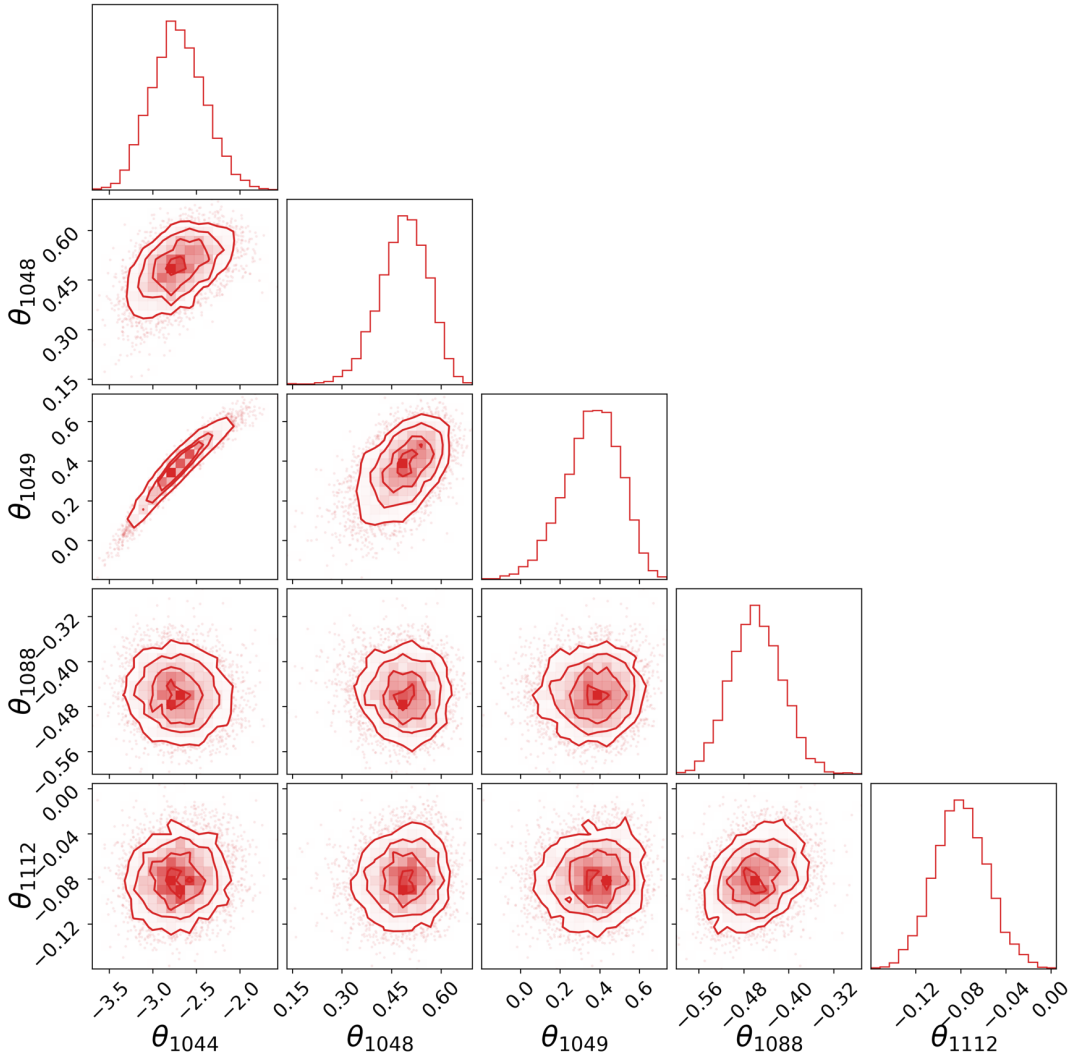
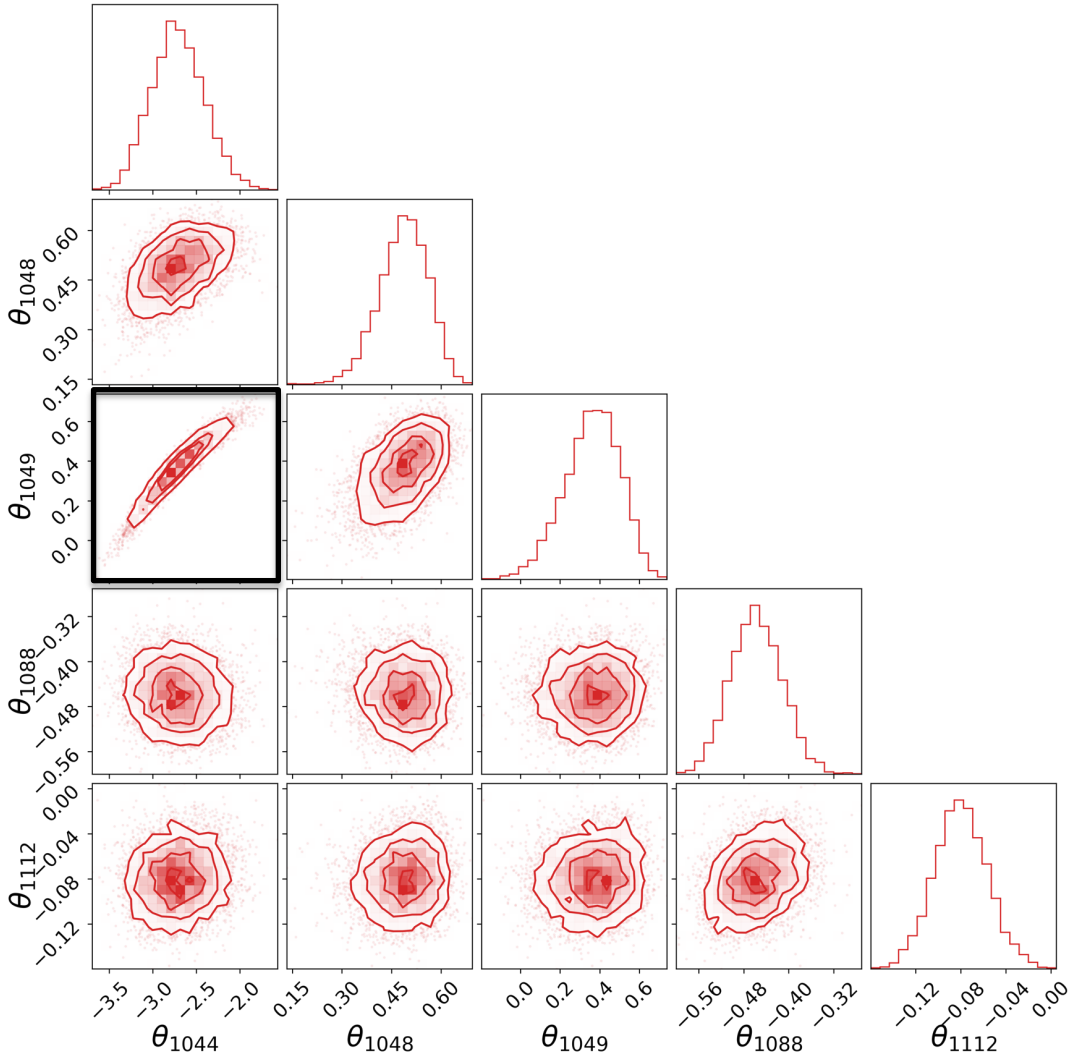


Estimated posterior after log pooling with $K = 10^3$ plausible datasets generated at d^*. Prior distributions are a standard normal.

Results

	n-R + O2 ⟶ propylene oxide
1044	O2 + CH3CH2CH2 ⟶ OH + C-CH2OCH(CH3)
	n-RO2 ⟶ products
1049	CH3CH2CH2OO ⟶ OH + C-CH2OCH(CH3)

Posterior distribution provides correlations between two of the sensitive reaction rates, both sharing propylene oxide as a product.



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Conclusion & Future Work

- Demonstrated feasibility and workflow for applying OED for high-dimensional physics-based models
- Bayesian optimization enables efficient exploration of noisy utility functions
- Low-dimensional surrogates of model output provide necessary computational savings for assessing complex models

Future Work

- Demonstrate improvement in the posterior density using measurements at optimal design compared to random designs
- Update initial prior estimates utilizing available measurement data

Acknowledgements

We would like to thank Oscar Diaz-Ibarra, Kyungjoo Kim, Arun Hegde, Cosmin Safta, and Khachik Sargsyan for helpful conversations about this work.

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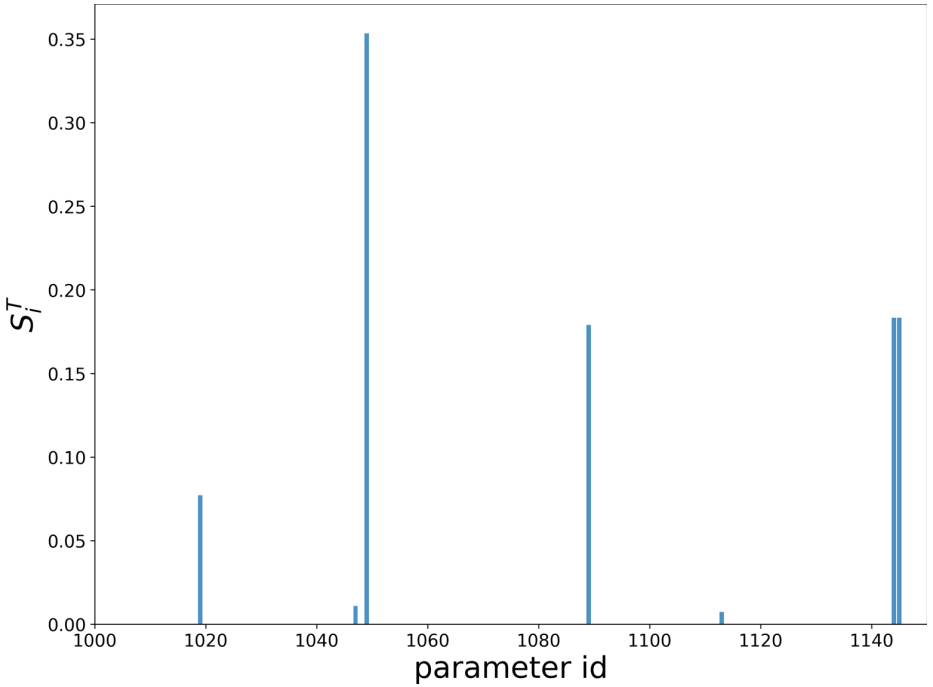
Additional Slides

Challenges

Large number of uncertain model parameters

- Physics and instrument model contain 1151 uncertain model parameters
- Only a *small fraction* have a significant impact on the spectrum at each design



Only a small number of nonzero total Sobol indices for design conditions 620 K, 2 bar, $n_{\mathrm{C3H8}} = 2\mathrm{e}14$, $n_{\mathrm{O2}} = 8\mathrm{e}18$, $n_{\mathrm{Cl}} = 1\mathrm{e}13$.

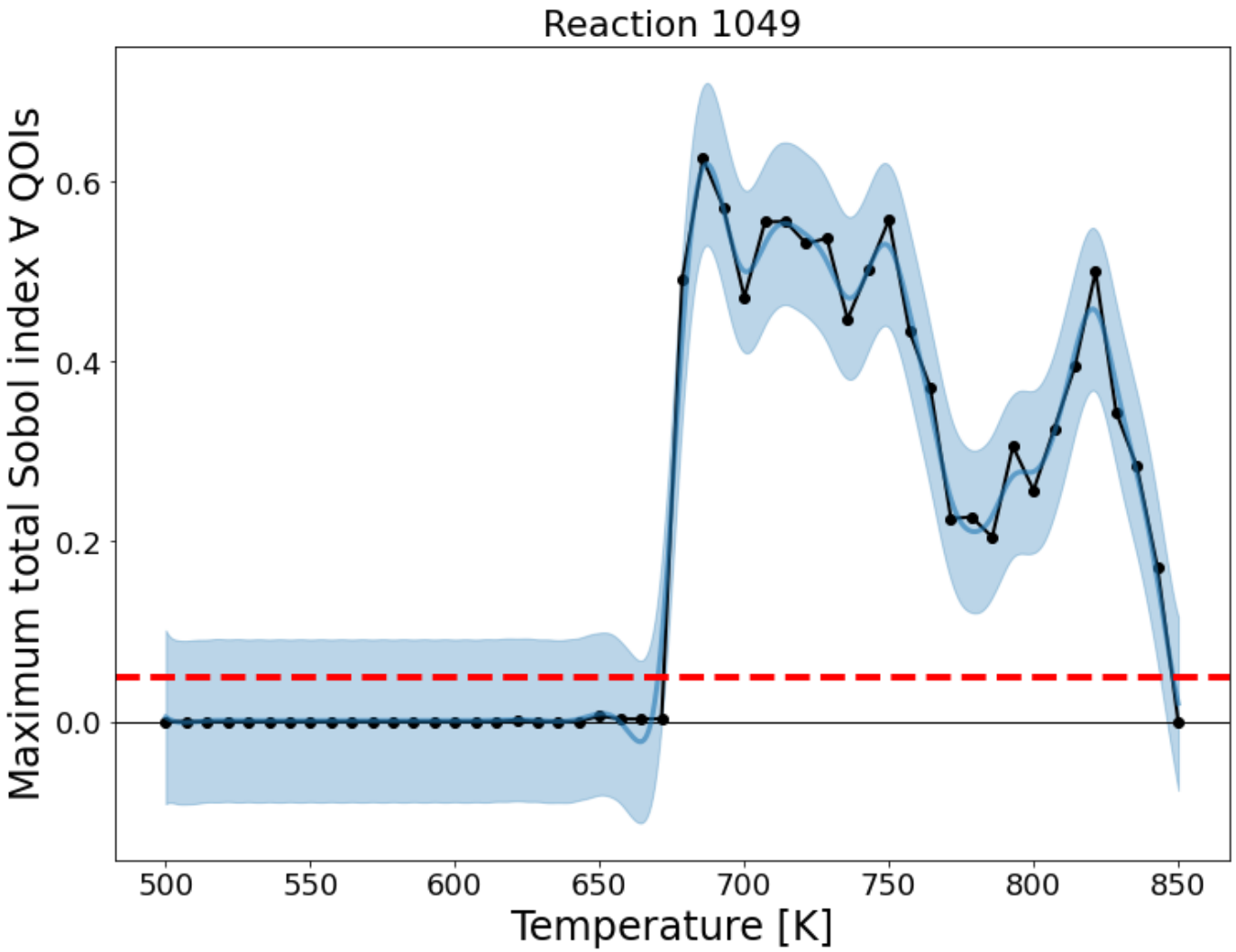
Variance-based Global Sensitivity Analysis

$$S_i^T = 1 - \frac{\mathrm{Var}_{\theta_{\sim i}}\left[\mathbb{E}_{\theta_i}\left(f\left(\boldsymbol{\theta}\right)|\theta_{\sim i}\right)\right]}{\mathrm{Var}_{\theta}\left[f(\boldsymbol{\theta})\right]}$$

- $\ell 1$ regularization of the polynomial chaos expansion coefficients

$$\boldsymbol{c}^{CS} = \arg\min_{\boldsymbol{c}} ||y - \boldsymbol{c}\boldsymbol{\Psi}||_2 + \gamma||\boldsymbol{c}||_1$$

Bruno Sudret. Global sensitivity analysis using polynomial chaos expansions. *Reliability engineering & system safety* 93, no. 7 (2008): 964-979


Reaction 1049
Maximum total Sobol index ∀ QOIs
0.0
0.2
0.4
0.6
500
550
600
650
700
750
800
850
Temperature [K]

Reducing output dimensionality

Goal: Map high-dimensional model output to a lower-dimensional space while minimizing loss of information

$$g : \mathbb{R}^J \mapsto \mathbb{R}^K$$

Autoencoder

Encoder network: $q = g_\psi(y)$ $\quad q \in \mathbb{R}^K$

Decoder network: $\widehat{y} = h_\phi(g_\psi(y))$ $\quad y \in \mathbb{R}^J$

$K \ll J$

Minimize reconstruction loss

$$L(\phi, \psi) = \min_{\phi,\psi} ||y - h_\phi(g_\psi(y))||_2^2$$

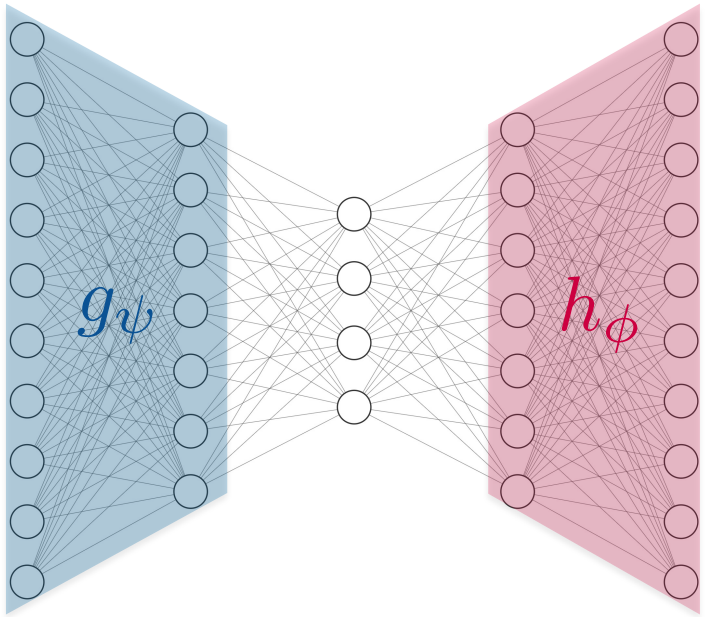
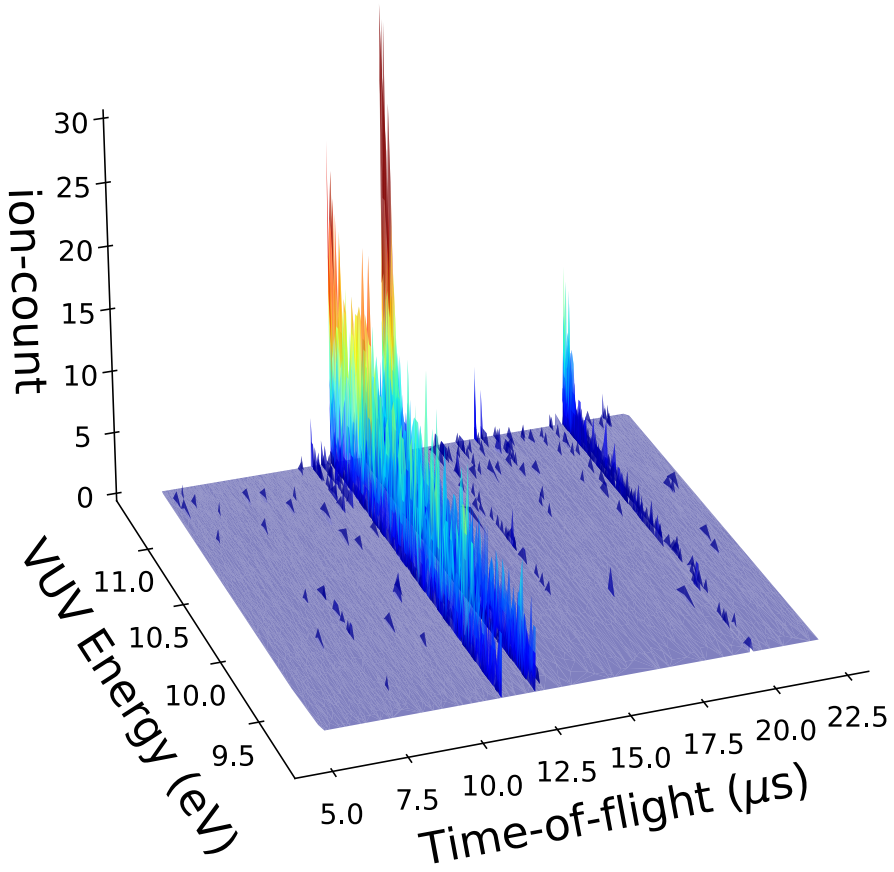
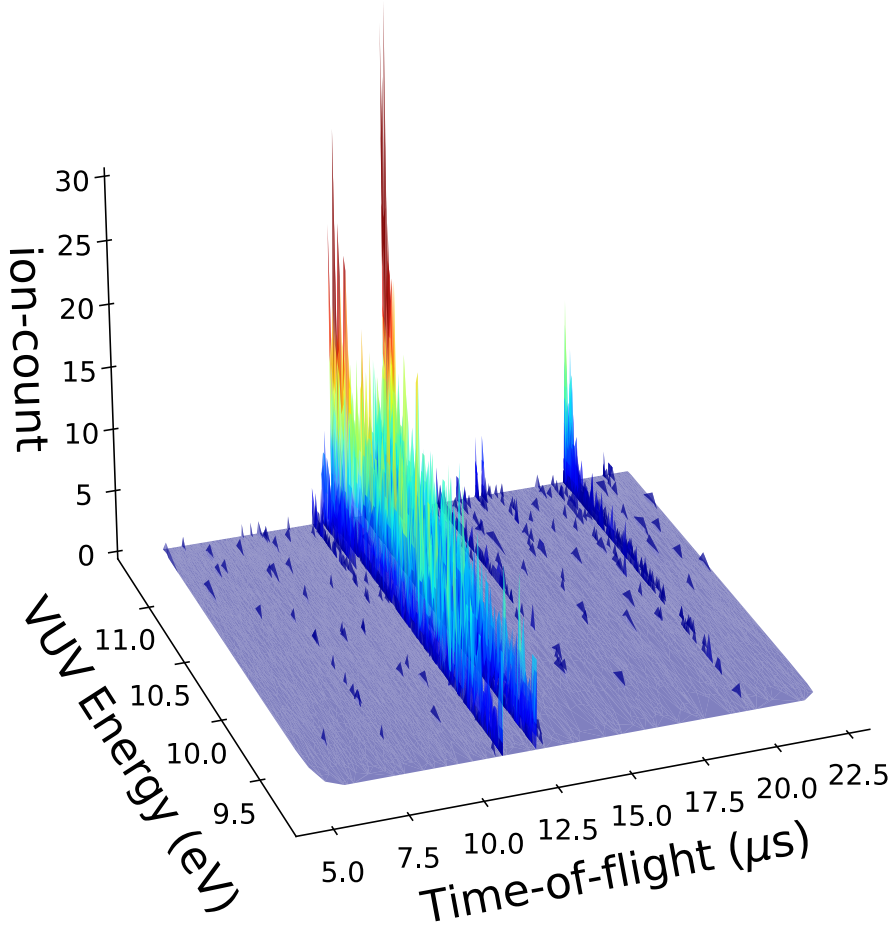


Illustration of an autoencoder architecture

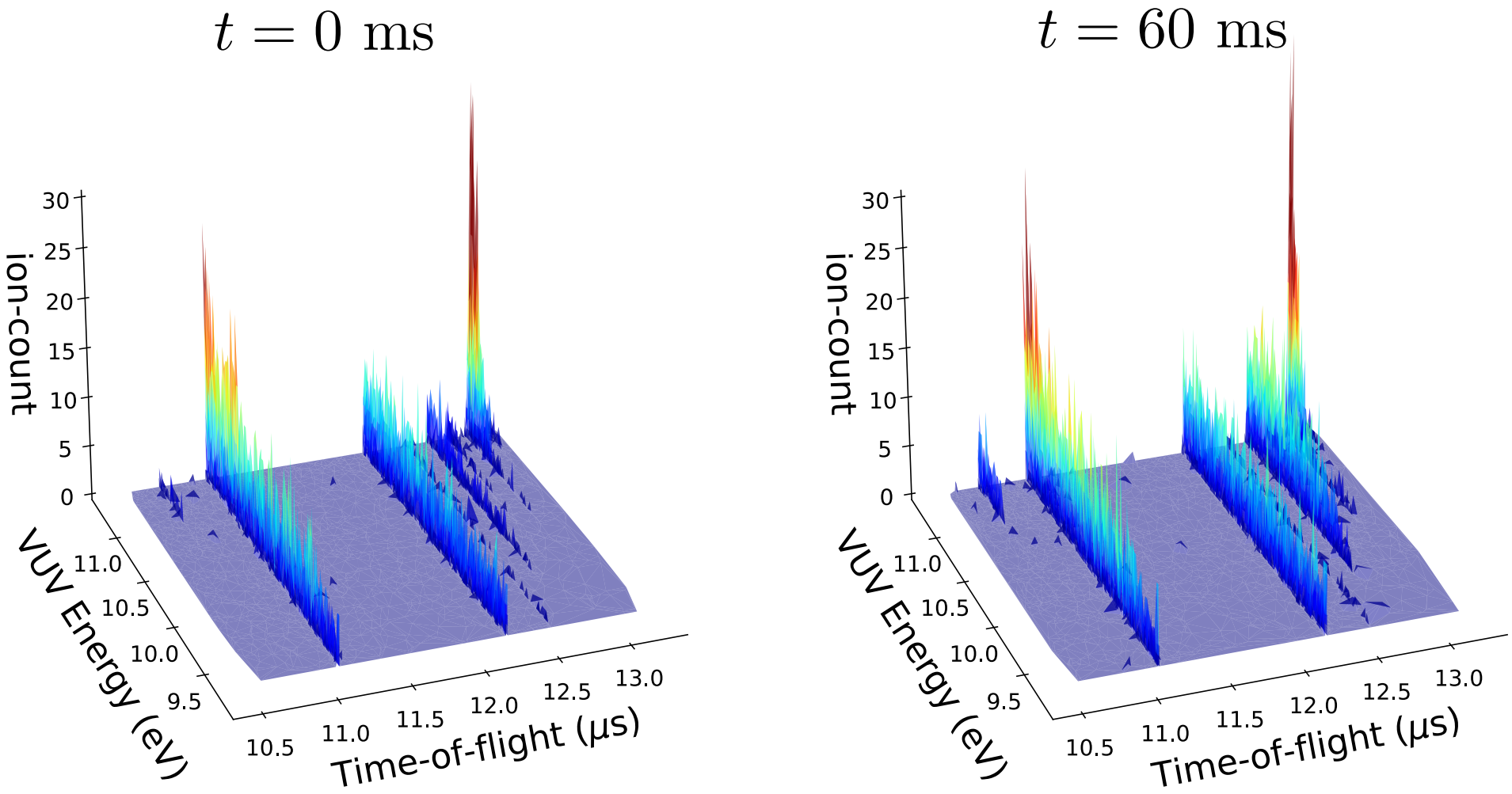
$t = 0$ ms



$t = 60$ ms



Experiment conducted at $T = 700$ K, p $= 10$ bar, $\chi_{C3H8} = 9.7 \times 10^{-7}$, $\chi_{O_2} = 2.9 \times 10^{-2}$, $\chi_{pre} = 2.2 \times 10^{-4}$ with a helium bath gas



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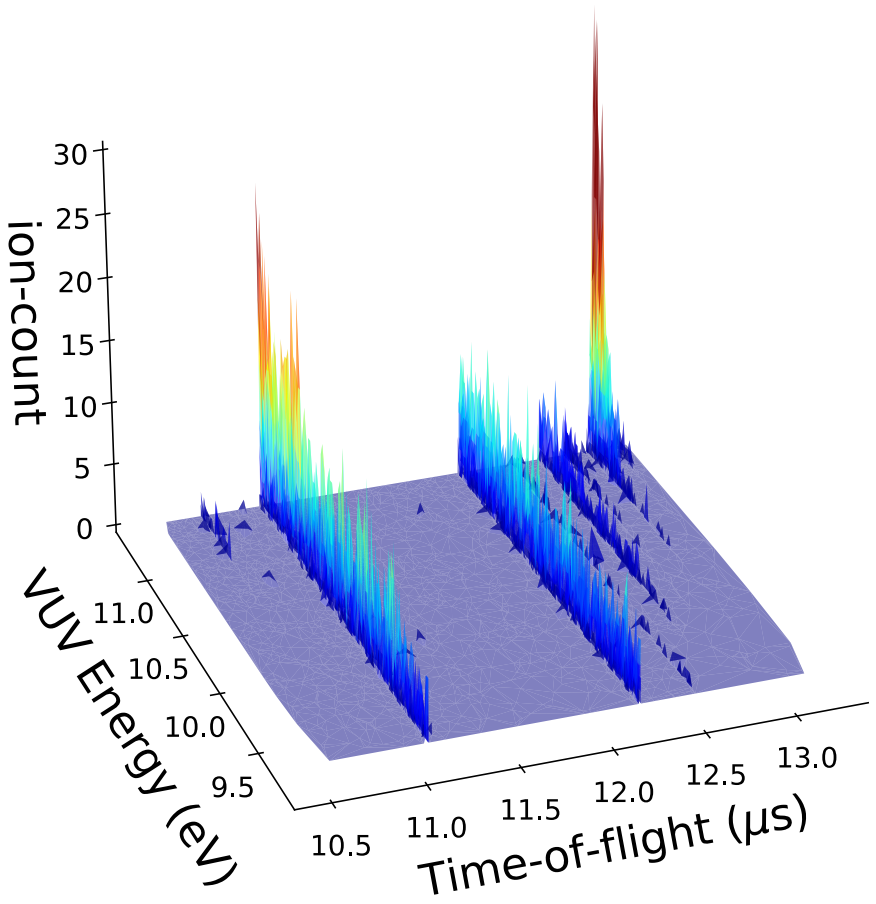
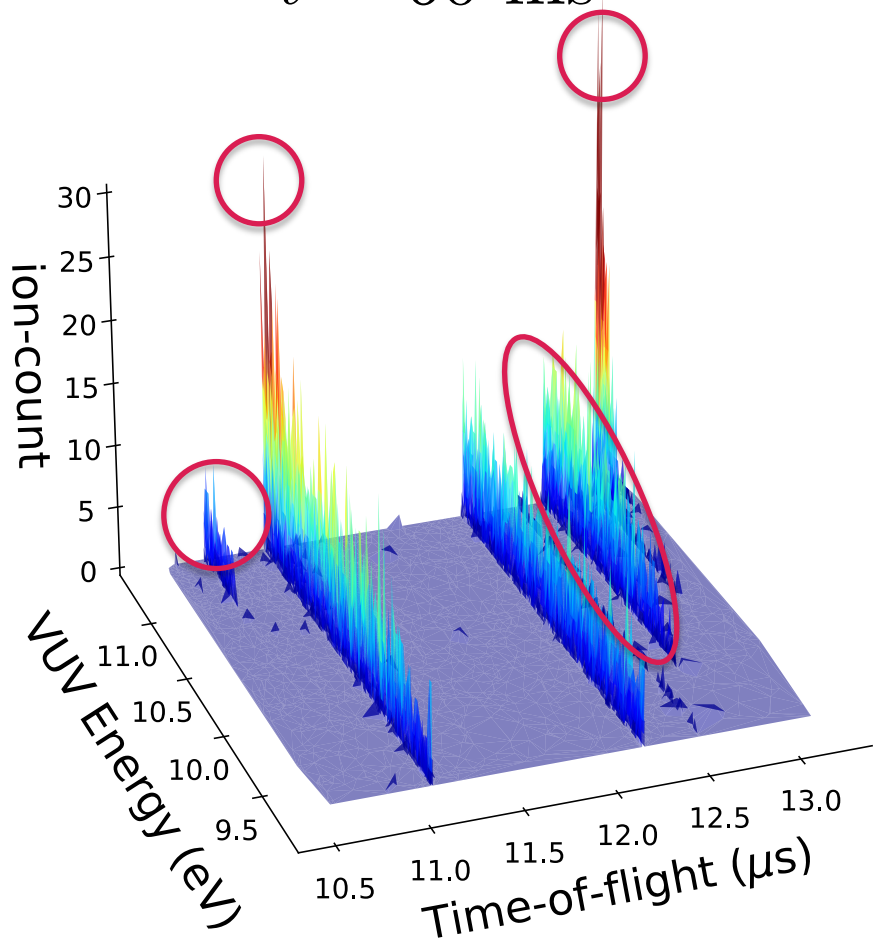
$t = 0$ ms



$t = 60$ ms



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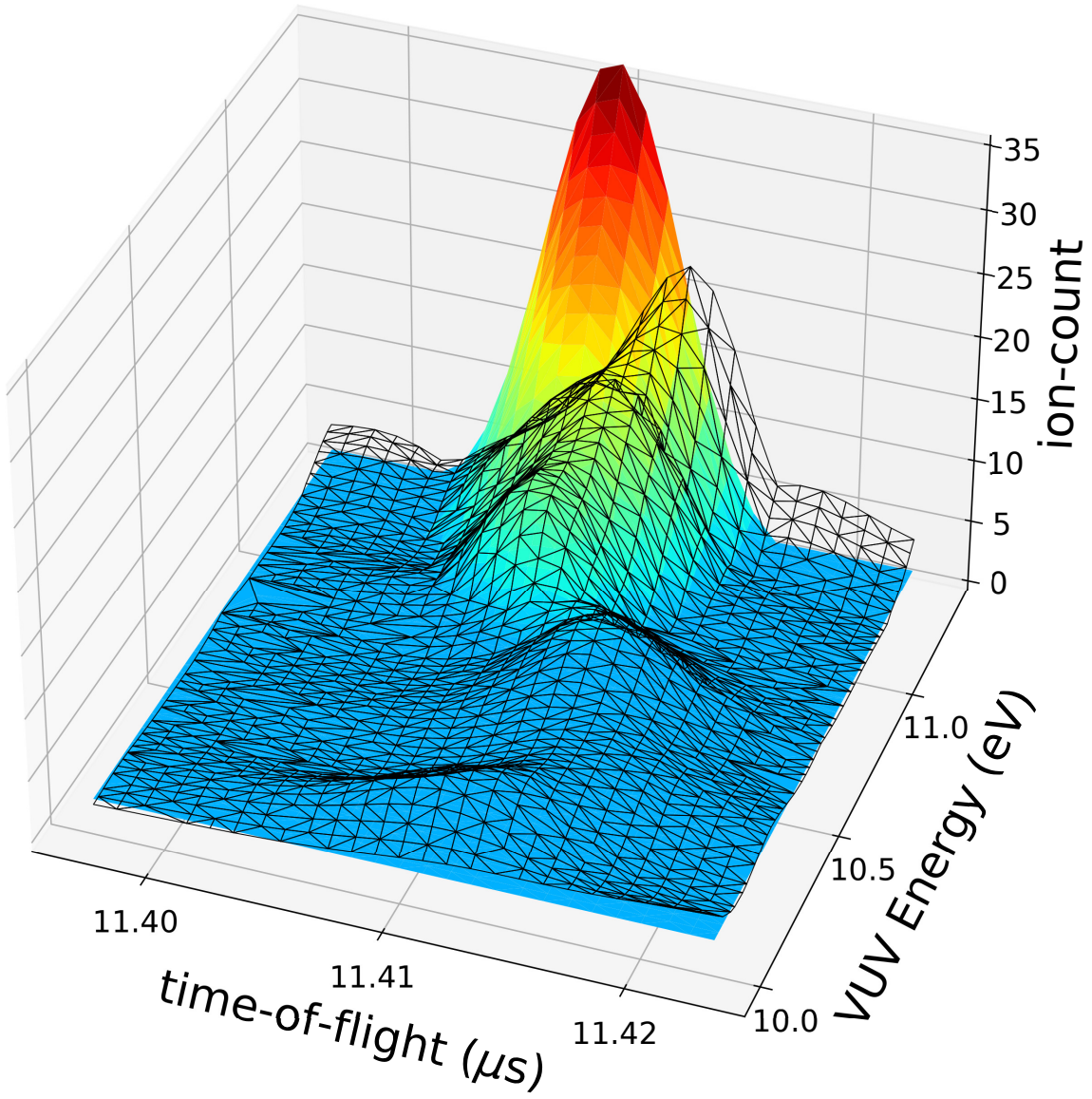
Modeling the HPR experiment



Solid surface is the prediction of $f(\theta_{MAP}, d, x)$ for one of the peaks in the time-of-flight spectrum (H_2O_2).
Mesh surface shows the prediction with model error, $f(\theta_{MAP}, d, x) + \mu_\delta(x)$ which increases the fidelity of the predictive model.

Numerical approximation

$$u(y, d, \theta) = D_{KL}(p(\theta|y, d)||p(\theta)) = \int_\Theta p(\theta|y, d) \log \left[\frac{p(\theta|y, d)}{p(\theta)} \right] d\theta = u(y, d)$$

$$\begin{aligned} U(d) &= \int_\mathcal{Y} \int_\Theta u(y, d) p(\theta|y, d) d\theta p(y|d) dy \\ &= \int_\mathcal{Y} u(y, d) p(y|d) dy \\ &= \int_\mathcal{Y} \left(\int_\Theta p(\theta|y, d) \log \left[\frac{p(\theta|y, d)}{p(\theta)} \right] d\theta \right) p(y|d) dy \end{aligned}$$

Using $p(\theta|y, d) = p(y|\theta, d)p(\theta)/p(y|d)$,

$$\begin{aligned} U(d) &= \int_\mathcal{Y} \int_\Theta \log \left[\frac{p(y|\theta, d)}{p(y|d)} \right] p(y|\theta, d) p(\theta) d\theta dy \\ &= \int_\mathcal{Y} \int_\Theta \left[\log p(y|\theta, d) - \log p(y|d) \right] p(y|\theta, d) p(\theta) d\theta dy \end{aligned}$$