Supplementary Information - Diagnostics of Data-Driven Models: Uncertainty Quantification of PM7 Semi-Empirical Quantum Chemical Method

James Oreluk[*], Zhenyuan Liu[*], Arun Hegde[*], Wenyu Li[*],
Andrew Packard[*], Michael Frenklach[†], Dmitry Zubarev[‡]

Feasible parameter vectors and feasibility labels are provided in two sets of CSV files. The feasibility labels files, `feasiblePoints_labels_1.csv` and `feasiblePoints_labels_2.csv` are of size nSamples-by-8, where column 1 is the feasibility label associated with methane, column 2 is the feasibility label associated with ethane, and so forth to column 8 which is feasibility label for octane. True indicates feasible and false indicates infeasible.

The associated feasible parameters vector files, `feasiblePoints_parameters_1.csv` and `feasiblePoints_parameters_2.csv` are of size nSamples-by-27, where each row is a parameter vector. The columns are the 27 parameters listed in Table S1. The feasibility of a parameter vector is found in the corresponding row of the associated feasibility labels file.

[*] Department of Mechanical Engineering, University of California at Berkeley, Berkeley, California 94720-1740

[†] Corresponding author: frenklach@berkeley.edu, Department of Mechanical Engineering, University of California at Berkeley, Berkeley, California 94720-1740

[‡] IBM Almaden Research Center, 650 Harry Road, San Jose, California 95136

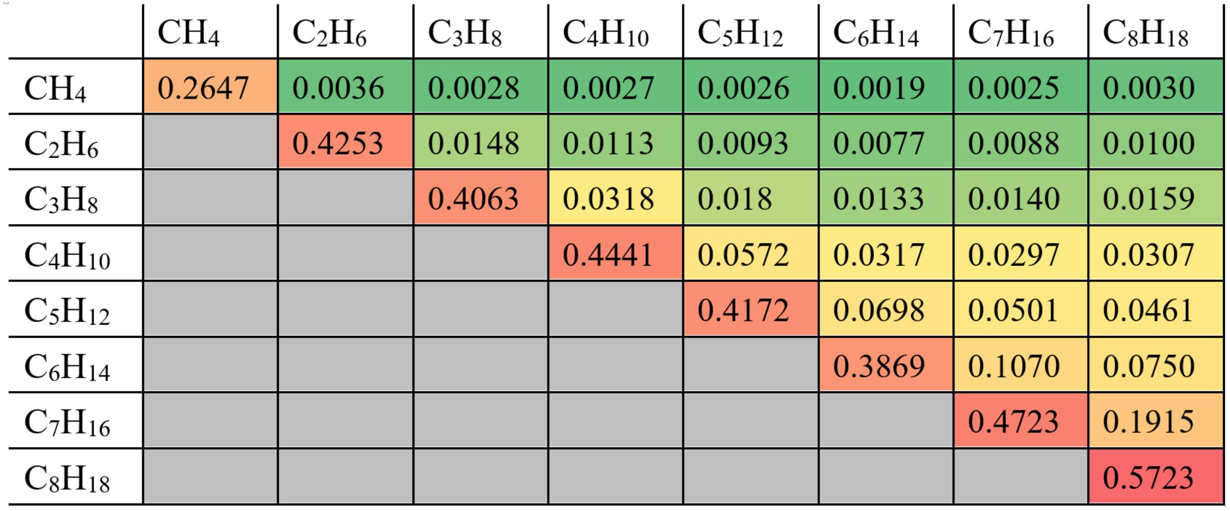
Table S1: PM7 nominal parameter vector and the δ associated with a 10 kcal/mol change in the heat of formation of C_4H_{10}, where each parameter value was perturbed, one-at-a-time from its nominal value.

Parameter	**Value**	$\boldsymbol{\delta}$
$\mathbf{USS_{H}}$	-11.07	0.29033
$\mathbf{BETAS_{H}}$	-8.3897	0.069276
$\mathbf{ZS_{H}}$	1.2602	0.017434
$\mathbf{GSS_{H}}$	14.15	0.36839
$\mathbf{FN11_{H}}$	0.17785	0.009959
$\mathbf{FN21_{H}}$	1.4287	0.33716
$\mathbf{FN31_{H}}$	0.99132	0.077376
$\mathbf{ALPB_{H}}$	4.0512	0.54877
$\mathbf{XFAC_{H}}$	2.8456	4.875
$\mathbf{USS_{C}}$	-51.373	0.11427
$\mathbf{UPP_{C}}$	-40.135	0.084482
$\mathbf{BETAS_{C}}$	-14.415	0.12922
$\mathbf{BETAP_{C}}$	-7.8937	0.067632
$\mathbf{ZS_{C}}$	1.9422	0.012886
$\mathbf{ZP_{C}}$	1.7087	0.007471
$\mathbf{GSS_{C}}$	12.347	0.13671
$\mathbf{GSP_{C}}$	11.933	0.18749
$\mathbf{GPP_{C}}$	10.452	0.8254
$\mathbf{GP2_{C}}$	9.3855	0.031369
$\mathbf{HSP_{C}}$	0.80263	0.11697
$\mathbf{FN11_{C}}$	0.045888	0.005604
$\mathbf{FN21_{C}}$	5.0371	1.5601
$\mathbf{FN31_{C}}$	1.5887	0.12723
$\mathbf{ALPB_{HC}}$	1.0387	0.007624
$\mathbf{XFAC_{HC}}$	0.20458	0.002002
$\mathbf{ALPC_{C}}$	2.6557	0.024061
$\mathbf{XFAC_{C}}$	0.93782	0.034605

Table S2: Parameter vector of $\mathcal{F}_{2:8}$ found via global optimization with a genetic algorithm using MATLAB's *ga* function.

Parameter	Value
$\mathbf{USS_H}$	-14.577
$\mathbf{BETAS_H}$	-7.3947
$\mathbf{ZS_H}$	1.1558
$\mathbf{GSS_H}$	15.096
$\mathbf{FN11_H}$	0.21973
$\mathbf{FN21_H}$	1.3555
$\mathbf{FN31_H}$	0.89818
$\mathbf{ALPB_H}$	4.0908
$\mathbf{XFAC_H}$	2.6264
$\mathbf{USS_C}$	-50.896
$\mathbf{UPP_C}$	-42.679
$\mathbf{BETAS_C}$	-12.204
$\mathbf{BETAP_C}$	-8.0123
$\mathbf{ZS_C}$	1.9807
$\mathbf{ZP_C}$	1.5976
$\mathbf{GSS_C}$	12.653
$\mathbf{GSP_C}$	10.104
$\mathbf{GPP_C}$	9.8942
$\mathbf{GP2_C}$	9.8562
$\mathbf{HSP_C}$	0.77597
$\mathbf{FN11_C}$	0.051751
$\mathbf{FN21_C}$	4.8582
$\mathbf{FN31_C}$	1.6624
$\mathbf{ALPB_{HC}}$	1.0878
$\mathbf{XFAC_{HC}}$	0.17728
$\mathbf{ALPC_C}$	2.5316
$\mathbf{XFAC_C}$	0.75887

Table S3: Percentage of feasible points found (out of 5.76 million samples) for each alkane and pair of alkanes.



	CH_4	C_2H_6	C_3H_8	C_4H_{10}	C_5H_{12}	C_6H_{14}	C_7H_{16}	C_8H_{18}
CH_4	0.2647	0.0036	0.0028	0.0027	0.0026	0.0019	0.0025	0.0030
C_2H_6		0.4253	0.0148	0.0113	0.0093	0.0077	0.0088	0.0100
C_3H_8			0.4063	0.0318	0.018	0.0133	0.0140	0.0159
C_4H_{10}				0.4441	0.0572	0.0317	0.0297	0.0307
C_5H_{12}					0.4172	0.0698	0.0501	0.0461
C_6H_{14}						0.3869	0.1070	0.0750
C_7H_{16}							0.4723	0.1915
C_8H_{18}								0.5723

Table S4: Largest percentage of feasible samples for three alkanes, indexed by QOI model.

$\mathcal{F}_{i,j,k}$	**Percentage of feasible samples**
6, 7, 8	0.0738
5, 6, 7	0.0488
5, 6, 8	0.0434
5, 7, 8	0.0432
4, 5, 6	0.0312
4, 5, 7	0.0285
4, 6, 7	0.0282
4, 5, 8	0.0278
4, 7, 8	0.0266
4, 6, 8	0.0260

Table S5: Feasible sets of consecutive alkanes which had a few or no feasible samples, indexed by the QOI model.

QOI Index	**Number of feasible samples**
1, 2, 3	7
1, 2, 3, 4	4
1, 2, 3, 4, 5	0
2, 3, 4, 5	7
5, 6, 7	2809
5, 6, 7, 8	2431
4, 5, 6, 7, 8	1431
3, 4, 5, 6, 7, 8	145

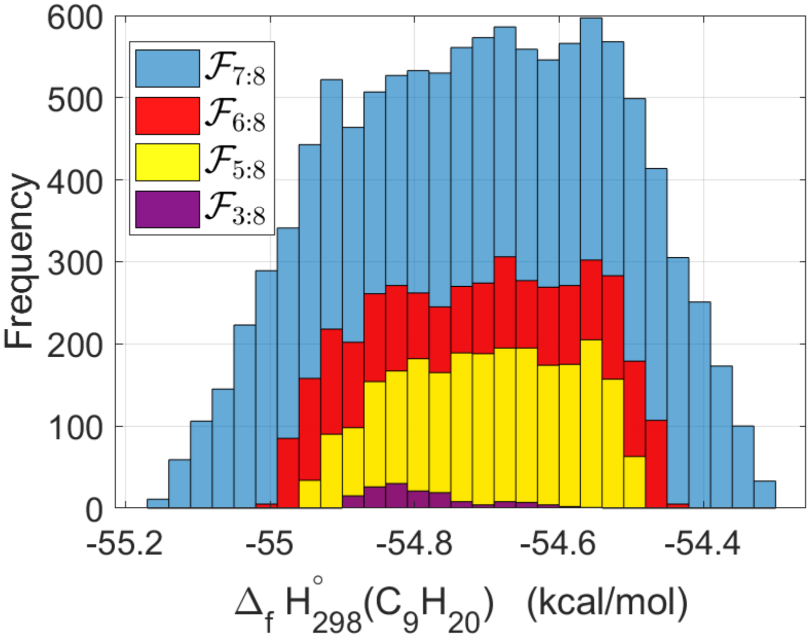


Figure S1: Predicted heat of formation of nonane by feasible samples of smaller alkanes. Using samples of $\mathcal{F}_{3:8}$ reduce the uncertainty in the predicted heat of formation by 40.5% compared to the prediction from samples of $\mathcal{F}_{7:8}$.

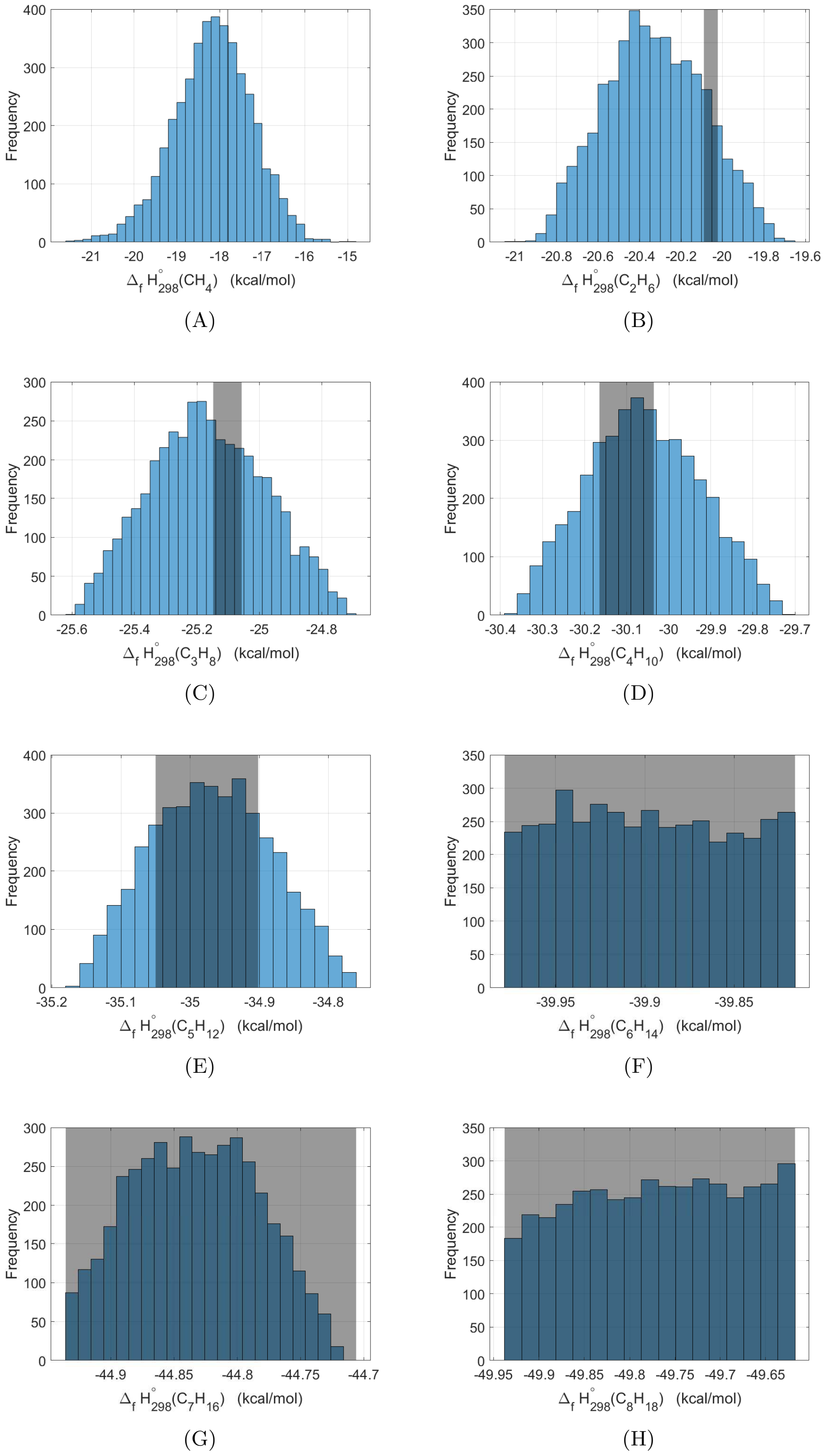


Figure S2: Predicted heat of formation of methane, ethane, to octane from samples of $\mathcal{F}_{6:8}$. The grey shaded region is the respective alkanes experimental interval shown in Table 1.

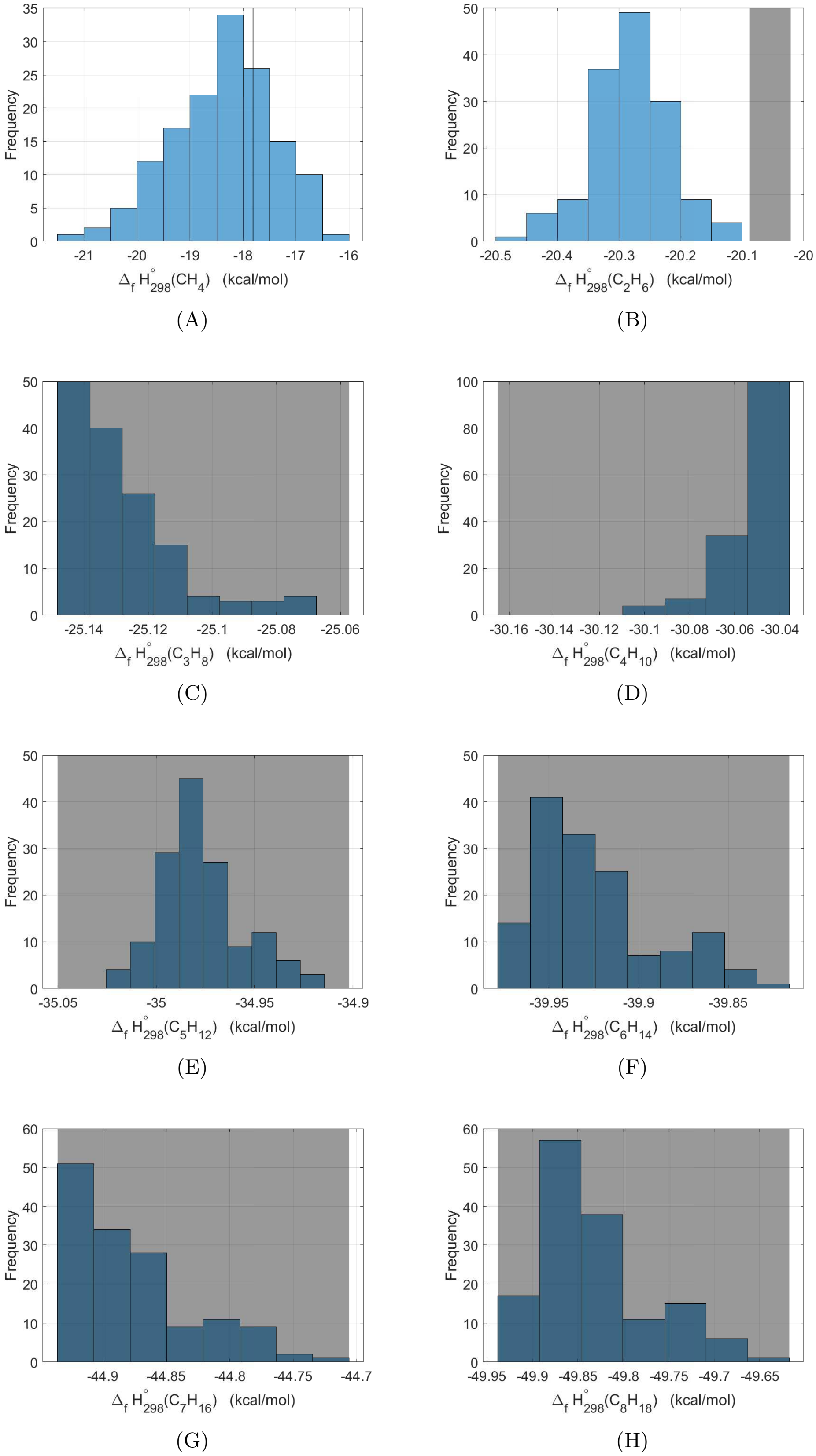


Figure S3: Predicted heat of formation of methane, ethane to octane from samples of $\mathcal{F}_{3:8}$. The grey shaded region is the respective alkanes experimental interval shown in Table 1.

Acknowledgments

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