

Supplementary Information

Exploring Chemical and Conformational Spaces by Batch Mode Deep Active Learning

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1 MD17

Here, additional results on the aspirin molecules from the MD17 data set¹⁻⁴ are presented. Table 1 and Table 2 present the numerical values for the mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients, as well as 95% quantile, evaluated on test data for total energies and forces. While Table 1 provides us with the errors averaged over the learning curve, Table 2 shows the performance of the final model. All results are given for the $(10, 100; 10)_{\text{MD17}}$ BMDAL experiment. The respective results for $(100, 1000; 100)_{\text{MD17}}$ can be found in Table 3 and Table 4. The measure of uncertainty for MAXDIST and LCMD is given by the distance to the closest point in the training set, where the distance is measured with respect to the corresponding kernel.

Fig. 1, Fig. 2, and Fig. 3 complement the results obtained in the main manuscript on atomic forces with the results for the total energy. Fig. 4, Fig. 5, and Fig. 6 present the batch size dependence and the learning curves for $(100, 1000; \{10, 25, 50, 100\})_{\text{MD17}}$ or $(100, 1000; 100)_{\text{MD17}}$, respectively.

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Table 1 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed for (10, 100; 10)_{MD17} as an average over AL steps. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy						Force						Time	
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MAXDIAG+RND	0.89 (0.03)	1.18 (0.05)	9.15 (0.91)	0.00 (0.00)	0.00 (0.00)	2.34 (0.09)	1.96 (0.06)	2.89 (0.09)	6.04 (0.44)	-0.00 (0.00)	-0.00 (0.00)	2.78 (0.09)	0.00 (0.00)	0.03 (0.00)
MAXDIAG+AE(E)	1.11 (0.06)	1.44 (0.07)	8.90 (0.81)	1.00 (0.00)	1.00 (0.00)	2.87 (0.15)	2.52 (0.09)	3.74 (0.14)	6.40 (0.40)	0.24 (0.02)	0.19 (0.02)	3.49 (0.13)	106.82 (2.68)	0.03 (0.00)
MAXDIAG+AE(F)	1.04 (0.05)	1.35 (0.06)	8.35 (0.73)	0.24 (0.02)	0.19 (0.02)	2.68 (0.13)	2.34 (0.08)	3.43 (0.12)	5.56 (0.26)	1.00 (0.00)	1.00 (0.00)	3.22 (0.11)	114.61 (11.21)	0.35 (0.33)
MAXDIAG+QBC(E)	1.09 (0.04)	1.41 (0.06)	6.92 (0.50)	0.44 (0.04)	0.47 (0.03)	2.32 (0.09)	2.46 (0.08)	3.64 (0.13)	5.60 (0.25)	0.29 (0.02)	0.26 (0.02)	3.27 (0.11)	278.63 (8.00)	0.05 (0.02)
MAXDIAG+QBC(F)	1.07 (0.04)	1.39 (0.05)	7.06 (0.63)	0.32 (0.02)	0.29 (0.02)	2.30 (0.09)	2.38 (0.05)	3.50 (0.08)	5.32 (0.25)	0.71 (0.01)	0.72 (0.01)	3.18 (0.07)	280.22 (3.69)	0.03 (0.00)
MAXDIAG+GP(LL)	1.07 (0.05)	1.39 (0.07)	9.12 (0.83)	0.08 (0.02)	0.05 (0.02)	2.75 (0.13)	2.38 (0.10)	3.51 (0.16)	5.94 (0.30)	0.23 (0.04)	0.19 (0.04)	3.30 (0.14)	56.21 (1.58)	0.03 (0.00)
MAXDIAG+GP(RP)	1.07 (0.05)	1.39 (0.07)	9.34 (1.04)	0.17 (0.01)	0.13 (0.01)	2.77 (0.15)	2.42 (0.08)	3.57 (0.12)	6.28 (0.47)	0.48 (0.01)	0.46 (0.01)	3.35 (0.10)	78.10 (2.96)	0.03 (0.00)
MAXDET+GP(LL)	0.87 (0.04)	1.13 (0.05)	7.37 (0.52)	0.08 (0.01)	0.05 (0.01)	2.25 (0.10)	1.91 (0.05)	2.79 (0.09)	5.02 (0.27)	0.23 (0.04)	0.19 (0.04)	2.64 (0.08)	66.54 (5.18)	0.89 (0.18)
MAXDET+GP(RP)	0.87 (0.03)	1.12 (0.04)	7.07 (0.53)	0.14 (0.01)	0.11 (0.01)	2.23 (0.09)	1.89 (0.06)	2.74 (0.09)	4.53 (0.21)	0.47 (0.01)	0.45 (0.01)	2.57 (0.08)	87.37 (2.67)	0.58 (0.04)
MAXDIST+GP(LL)	0.85 (0.03)	1.10 (0.04)	7.14 (0.52)	0.13 (0.01)	0.11 (0.01)	2.19 (0.09)	1.92 (0.06)	2.80 (0.09)	4.67 (0.22)	0.42 (0.02)	0.41 (0.02)	2.63 (0.08)	54.69 (1.29)	4.08 (0.31)
MAXDIST+GP(RP)	0.86 (0.04)	1.12 (0.05)	7.01 (0.57)	0.15 (0.01)	0.12 (0.01)	2.22 (0.11)	1.91 (0.07)	2.79 (0.11)	4.62 (0.27)	0.49 (0.01)	0.47 (0.01)	2.62 (0.09)	77.62 (1.20)	3.38 (0.18)
MAXDIST+FEAT(LL)	0.88 (0.04)	1.15 (0.05)	7.32 (0.58)	0.09 (0.01)	0.07 (0.01)	2.29 (0.10)	1.90 (0.05)	2.78 (0.08)	4.86 (0.25)	0.29 (0.02)	0.28 (0.02)	2.63 (0.07)	54.59 (4.18)	3.72 (0.31)
MAXDIST+FEAT(RP)	0.88 (0.03)	1.14 (0.04)	7.43 (0.67)	0.11 (0.01)	0.09 (0.01)	2.26 (0.09)	1.91 (0.06)	2.79 (0.10)	5.17 (0.48)	0.34 (0.02)	0.32 (0.02)	2.63 (0.09)	78.70 (7.36)	3.45 (0.36)
LCMD+FEAT(LL)	0.89 (0.04)	1.16 (0.05)	7.90 (0.67)	0.11 (0.01)	0.07 (0.01)	2.30 (0.10)	1.92 (0.05)	2.80 (0.08)	5.33 (0.32)	0.33 (0.02)	0.29 (0.02)	2.67 (0.07)	63.11 (1.69)	4.65 (0.22)
LCMD+FEAT(RP)	0.87 (0.03)	1.13 (0.04)	8.05 (0.70)	0.12 (0.01)	0.10 (0.01)	2.23 (0.09)	1.92 (0.05)	2.81 (0.09)	5.50 (0.55)	0.38 (0.02)	0.35 (0.02)	2.66 (0.08)	86.78 (1.71)	4.07 (0.21)

Table 2 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed employing the final model obtained for the (10, 100; 10)MD17 experiment. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. Standard errors on the mean are given in parentheses.

Method	Energy					Force						
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	0.61 (0.01)	0.79 (0.02)	6.06 (0.54)	0.00 (0.00)	0.00 (0.00)	1.56 (0.03)	1.26 (0.02)	1.83 (0.02)	4.12 (0.47)	-0.00 (0.00)	-0.00 (0.00)	1.81 (0.02)
MAXDIAG+AE(E)	0.78 (0.04)	1.00 (0.05)	5.63 (0.52)	1.00 (0.00)	1.00 (0.00)	1.98 (0.11)	1.81 (0.05)	2.66 (0.08)	4.60 (0.37)	0.21 (0.01)	0.16 (0.01)	2.47 (0.08)
MAXDIAG+AE(F)	0.72 (0.03)	0.92 (0.03)	4.77 (0.26)	0.20 (0.02)	0.15 (0.02)	1.82 (0.06)	1.64 (0.04)	2.38 (0.06)	3.84 (0.21)	1.00 (0.00)	1.00 (0.00)	2.22 (0.05)
MAXDIAG+QBC(E)	0.77 (0.02)	0.99 (0.03)	5.12 (0.40)	0.36 (0.02)	0.40 (0.02)	1.67 (0.03)	1.77 (0.04)	2.62 (0.07)	4.22 (0.15)	0.26 (0.02)	0.23 (0.02)	2.35 (0.05)
MAXDIAG+QBC(F)	0.73 (0.01)	0.95 (0.02)	5.48 (0.63)	0.30 (0.03)	0.27 (0.02)	1.63 (0.04)	1.65 (0.02)	2.41 (0.04)	3.75 (0.21)	0.70 (0.01)	0.71 (0.01)	2.21 (0.03)
MAXDIAG+GP(LL)	0.77 (0.01)	0.99 (0.01)	6.29 (0.47)	0.07 (0.02)	0.05 (0.01)	1.98 (0.03)	1.69 (0.04)	2.48 (0.07)	4.02 (0.11)	0.25 (0.01)	0.21 (0.01)	2.33 (0.06)
MAXDIAG+GP(RP)	0.72 (0.02)	0.93 (0.03)	5.61 (0.43)	0.15 (0.02)	0.12 (0.01)	1.84 (0.06)	1.64 (0.03)	2.38 (0.05)	4.74 (0.46)	0.50 (0.01)	0.48 (0.01)	2.24 (0.05)
MAXDET+GP(LL)	0.58 (0.01)	0.75 (0.01)	4.56 (0.29)	0.06 (0.01)	0.04 (0.00)	1.50 (0.03)	1.29 (0.02)	1.85 (0.03)	3.00 (0.08)	0.25 (0.04)	0.21 (0.04)	1.76 (0.03)
MAXDET+GP(RP)	0.59 (0.01)	0.75 (0.01)	4.91 (0.41)	0.13 (0.01)	0.11 (0.00)	1.50 (0.04)	1.24 (0.03)	1.78 (0.04)	2.98 (0.07)	0.49 (0.01)	0.47 (0.01)	1.68 (0.04)
MAXDIST+GP(LL)	0.58 (0.01)	0.74 (0.01)	4.67 (0.23)	0.12 (0.01)	0.11 (0.01)	1.48 (0.02)	1.24 (0.03)	1.79 (0.04)	2.91 (0.09)	0.43 (0.02)	0.42 (0.02)	1.69 (0.04)
MAXDIST+GP(RP)	0.56 (0.02)	0.73 (0.03)	4.57 (0.49)	0.13 (0.01)	0.11 (0.01)	1.45 (0.05)	1.25 (0.03)	1.79 (0.04)	2.95 (0.12)	0.50 (0.01)	0.49 (0.01)	1.70 (0.04)
MAXDIST+FEAT(LL)	0.60 (0.01)	0.77 (0.01)	4.66 (0.34)	0.08 (0.01)	0.06 (0.01)	1.53 (0.03)	1.25 (0.03)	1.79 (0.05)	3.43 (0.32)	0.30 (0.01)	0.28 (0.01)	1.71 (0.04)
MAXDIST+FEAT(RP)	0.58 (0.01)	0.75 (0.01)	4.56 (0.28)	0.08 (0.01)	0.07 (0.01)	1.48 (0.03)	1.22 (0.02)	1.75 (0.04)	3.38 (0.35)	0.34 (0.02)	0.33 (0.02)	1.68 (0.03)
LCMD+FEAT(LL)	0.59 (0.02)	0.77 (0.02)	5.36 (0.52)	0.10 (0.01)	0.07 (0.01)	1.52 (0.04)	1.22 (0.03)	1.75 (0.05)	3.83 (0.46)	0.37 (0.03)	0.32 (0.04)	1.70 (0.04)
LCMD+FEAT(RP)	0.58 (0.02)	0.74 (0.02)	4.21 (0.32)	0.10 (0.01)	0.08 (0.01)	1.46 (0.03)	1.24 (0.04)	1.79 (0.06)	3.50 (0.19)	0.40 (0.03)	0.38 (0.03)	1.71 (0.06)

Table 3 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed for (100,1000;100)_{MD17} as an average over AL steps. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy						Force						Time	
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MAXDIAG+RND	0.49 (0.01)	0.64 (0.01)	4.46 (0.26)	-0.00 (0.00)	-0.00 (0.00)	1.26 (0.03)	0.91 (0.02)	1.30 (0.03)	3.12 (0.25)	0.00 (0.00)	0.00 (0.00)	1.30 (0.03)	0.01 (0.00)	0.03 (0.00)
MAXDIAG+AE(E)	0.52 (0.01)	0.66 (0.01)	3.81 (0.26)	1.00 (0.00)	1.00 (0.00)	1.32 (0.03)	1.13 (0.02)	1.64 (0.03)	3.45 (0.24)	0.18 (0.01)	0.15 (0.01)	1.58 (0.03)	107.43 (8.03)	0.02 (0.00)
MAXDIAG+AE(F)	0.51 (0.01)	0.66 (0.01)	3.93 (0.21)	0.21 (0.01)	0.17 (0.01)	1.31 (0.03)	1.09 (0.02)	1.58 (0.03)	2.76 (0.16)	1.00 (0.00)	1.00 (0.00)	1.51 (0.03)	103.73 (2.92)	0.03 (0.00)
MAXDIAG+QBC(E)	0.52 (0.01)	0.67 (0.01)	3.62 (0.14)	0.26 (0.01)	0.29 (0.01)	1.19 (0.02)	1.08 (0.01)	1.57 (0.02)	2.76 (0.10)	0.27 (0.01)	0.24 (0.01)	1.47 (0.01)	254.17 (6.19)	0.03 (0.00)
MAXDIAG+QBC(F)	0.51 (0.01)	0.66 (0.01)	3.66 (0.18)	0.21 (0.01)	0.21 (0.01)	1.18 (0.02)	1.06 (0.01)	1.52 (0.02)	2.47 (0.07)	0.69 (0.01)	0.71 (0.01)	1.44 (0.01)	283.63 (5.86)	0.03 (0.00)
MAXDIAG+GP(LL)	0.51 (0.01)	0.66 (0.02)	3.98 (0.21)	0.13 (0.01)	0.10 (0.01)	1.31 (0.04)	1.04 (0.02)	1.50 (0.03)	2.88 (0.17)	0.49 (0.01)	0.47 (0.01)	1.45 (0.03)	61.45 (2.18)	0.03 (0.00)
MAXDIAG+GP(RP)	0.51 (0.01)	0.66 (0.02)	4.09 (0.30)	0.16 (0.01)	0.13 (0.01)	1.31 (0.04)	1.06 (0.02)	1.53 (0.04)	2.87 (0.17)	0.59 (0.01)	0.59 (0.01)	1.48 (0.03)	77.92 (2.19)	0.03 (0.00)
MAXDET+GP(LL)	0.49 (0.01)	0.63 (0.02)	3.81 (0.16)	0.11 (0.01)	0.09 (0.01)	1.25 (0.03)	0.93 (0.02)	1.32 (0.03)	2.28 (0.11)	0.39 (0.01)	0.39 (0.01)	1.28 (0.03)	63.51 (4.27)	11.68 (1.62)
MAXDET+GP(RP)	0.49 (0.01)	0.63 (0.02)	3.66 (0.13)	0.15 (0.01)	0.12 (0.01)	1.26 (0.04)	0.90 (0.02)	1.27 (0.02)	2.17 (0.09)	0.50 (0.01)	0.50 (0.01)	1.23 (0.02)	101.37 (13.68)	13.94 (4.14)
MAXDIST+GP(LL)	0.48 (0.01)	0.62 (0.01)	3.81 (0.24)	0.07 (0.01)	0.08 (0.01)	1.22 (0.03)	0.88 (0.02)	1.24 (0.03)	2.19 (0.11)	0.36 (0.01)	0.42 (0.01)	1.21 (0.03)	55.09 (1.17)	38.03 (1.47)
MAXDIST+GP(RP)	0.49 (0.01)	0.63 (0.02)	3.86 (0.15)	0.08 (0.01)	0.10 (0.01)	1.24 (0.03)	0.88 (0.02)	1.24 (0.03)	2.16 (0.09)	0.40 (0.01)	0.47 (0.01)	1.21 (0.03)	74.48 (1.62)	32.75 (1.74)
MAXDIST+FEAT(LL)	0.48 (0.01)	0.62 (0.01)	3.94 (0.19)	0.06 (0.01)	0.06 (0.00)	1.22 (0.03)	0.88 (0.02)	1.25 (0.03)	2.46 (0.14)	0.29 (0.01)	0.31 (0.01)	1.23 (0.03)	52.40 (1.25)	36.55 (2.47)
MAXDIST+FEAT(RP)	0.49 (0.01)	0.64 (0.02)	3.87 (0.21)	0.05 (0.01)	0.06 (0.01)	1.26 (0.04)	0.89 (0.02)	1.25 (0.03)	2.36 (0.12)	0.27 (0.01)	0.32 (0.01)	1.22 (0.02)	73.01 (0.69)	30.88 (0.69)
LCMID+FEAT(LL)	0.48 (0.01)	0.63 (0.01)	4.25 (0.38)	0.12 (0.01)	0.09 (0.00)	1.24 (0.03)	0.90 (0.02)	1.27 (0.02)	2.80 (0.15)	0.43 (0.02)	0.41 (0.01)	1.27 (0.02)	69.62 (1.17)	51.94 (1.09)
LCMID+FEAT(RP)	0.49 (0.01)	0.63 (0.01)	4.08 (0.23)	0.09 (0.01)	0.08 (0.00)	1.24 (0.02)	0.89 (0.02)	1.26 (0.02)	2.92 (0.30)	0.40 (0.01)	0.41 (0.01)	1.26 (0.02)	93.92 (2.46)	42.71 (1.35)

Table 4 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed employing the final model obtained for the (100,1000;100)_{MD17} experiment. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. Standard errors on the mean are given in parentheses.

Method	Energy					Force						
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	0.37 (0.00)	0.48 (0.01)	2.83 (0.12)	-0.00 (0.00)	-0.00 (0.00)	0.95 (0.01)	0.66 (0.01)	0.94 (0.01)	2.54 (0.19)	0.00 (0.00)	0.00 (0.00)	0.97 (0.01)
MAXDIAG+AE(E)	0.38 (0.00)	0.48 (0.00)	2.19 (0.10)	1.00 (0.00)	1.00 (0.00)	0.95 (0.01)	0.91 (0.02)	1.34 (0.03)	2.68 (0.15)	0.12 (0.01)	0.10 (0.01)	1.28 (0.03)
MAXDIAG+AE(F)	0.38 (0.01)	0.48 (0.01)	2.77 (0.21)	0.18 (0.01)	0.15 (0.01)	0.95 (0.01)	0.88 (0.01)	1.30 (0.01)	2.03 (0.13)	1.00 (0.00)	1.00 (0.00)	1.22 (0.01)
MAXDIAG+QBC(E)	0.39 (0.01)	0.50 (0.01)	2.50 (0.05)	0.22 (0.01)	0.25 (0.01)	0.90 (0.02)	0.87 (0.01)	1.28 (0.01)	1.98 (0.08)	0.25 (0.01)	0.23 (0.01)	1.18 (0.01)
MAXDIAG+QBC(F)	0.39 (0.01)	0.50 (0.01)	2.62 (0.09)	0.18 (0.00)	0.19 (0.00)	0.91 (0.02)	0.85 (0.01)	1.24 (0.01)	1.73 (0.01)	0.70 (0.00)	0.71 (0.01)	1.15 (0.01)
MAXDIAG+GP(LL)	0.38 (0.01)	0.49 (0.01)	2.59 (0.06)	0.14 (0.01)	0.12 (0.01)	0.98 (0.03)	0.83 (0.01)	1.22 (0.01)	1.96 (0.07)	0.51 (0.01)	0.51 (0.01)	1.17 (0.01)
MAXDIAG+GP(RP)	0.39 (0.01)	0.49 (0.01)	2.90 (0.15)	0.15 (0.01)	0.13 (0.01)	0.98 (0.02)	0.85 (0.02)	1.25 (0.03)	1.96 (0.05)	0.59 (0.01)	0.58 (0.01)	1.18 (0.03)
MAXDET+GP(LL)	0.37 (0.01)	0.48 (0.01)	2.54 (0.06)	0.13 (0.00)	0.11 (0.01)	0.95 (0.02)	0.72 (0.01)	1.03 (0.02)	1.56 (0.04)	0.38 (0.00)	0.41 (0.01)	0.99 (0.02)
MAXDET+GP(RP)	0.37 (0.01)	0.48 (0.01)	2.53 (0.09)	0.13 (0.01)	0.11 (0.01)	0.95 (0.03)	0.66 (0.01)	0.94 (0.01)	1.37 (0.02)	0.43 (0.01)	0.44 (0.01)	0.91 (0.01)
MAXDIST+GP(LL)	0.36 (0.01)	0.46 (0.01)	2.51 (0.10)	0.04 (0.00)	0.07 (0.00)	0.91 (0.03)	0.63 (0.01)	0.89 (0.02)	1.42 (0.07)	0.34 (0.02)	0.41 (0.01)	0.86 (0.02)
MAXDIST+GP(RP)	0.36 (0.01)	0.46 (0.01)	2.54 (0.06)	0.05 (0.00)	0.08 (0.00)	0.91 (0.02)	0.64 (0.01)	0.89 (0.01)	1.40 (0.02)	0.39 (0.01)	0.46 (0.01)	0.86 (0.01)
MAXDIST+FEAT(LL)	0.36 (0.01)	0.46 (0.01)	2.47 (0.10)	0.03 (0.00)	0.05 (0.00)	0.91 (0.03)	0.64 (0.01)	0.89 (0.01)	1.61 (0.07)	0.29 (0.01)	0.32 (0.01)	0.88 (0.01)
MAXDIST+FEAT(RP)	0.38 (0.01)	0.48 (0.01)	2.51 (0.08)	0.02 (0.01)	0.05 (0.01)	0.95 (0.03)	0.64 (0.01)	0.90 (0.01)	1.56 (0.03)	0.25 (0.01)	0.30 (0.01)	0.88 (0.01)
LCMD+FEAT(LL)	0.36 (0.01)	0.47 (0.01)	2.89 (0.25)	0.12 (0.01)	0.10 (0.00)	0.93 (0.02)	0.65 (0.01)	0.91 (0.01)	1.96 (0.05)	0.49 (0.02)	0.47 (0.02)	0.93 (0.01)
LCMD+FEAT(RP)	0.36 (0.00)	0.47 (0.00)	2.86 (0.14)	0.09 (0.01)	0.09 (0.01)	0.93 (0.01)	0.64 (0.01)	0.91 (0.01)	2.26 (0.24)	0.43 (0.02)	0.45 (0.01)	0.93 (0.01)

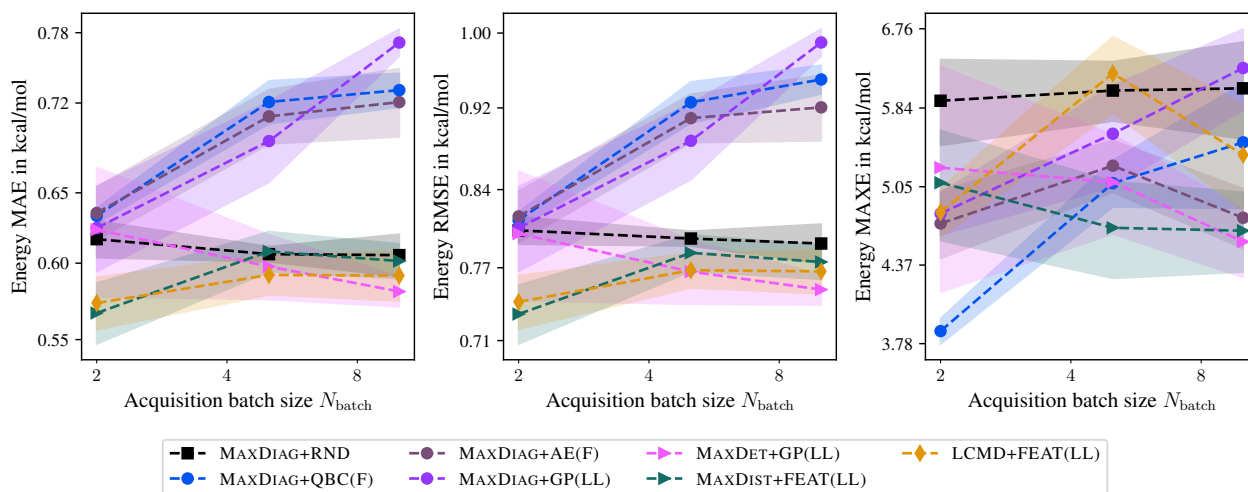


Figure 1 Dependence of the mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy on the acquired batch size N_{batch} . All errors are evaluated for the last BMDAL step on the aspirin molecule data from MD17.¹⁻⁴ All corresponding values are given for the $(10, 100; \{2, 5, 10\})_{\text{MD17}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

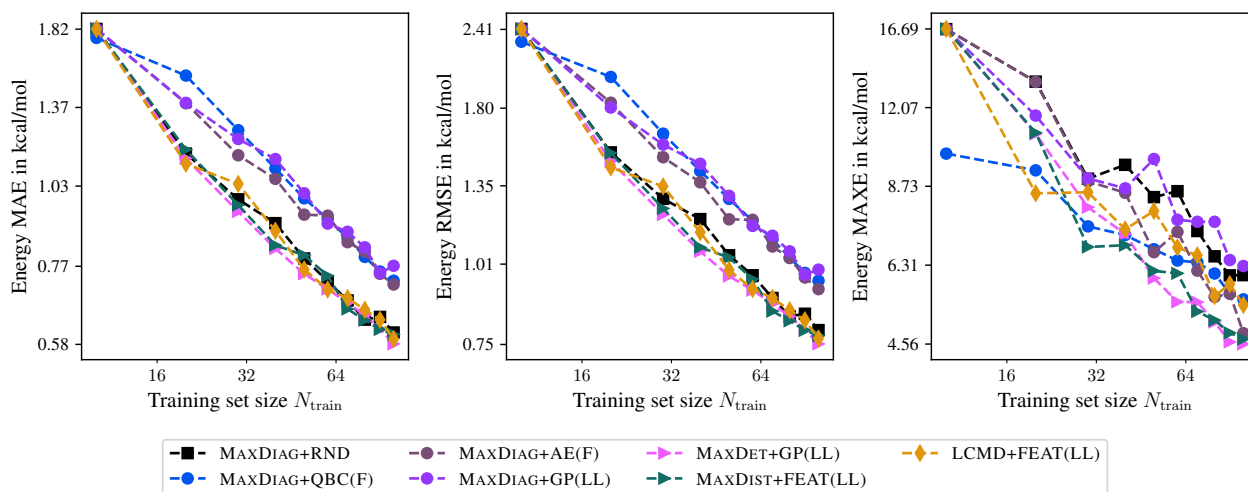


Figure 2 Learning curves for the aspirin molecule data from MD17.¹⁻⁴ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(10, 100; 10)_{\text{MD17}}$ BMDAL experiment. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

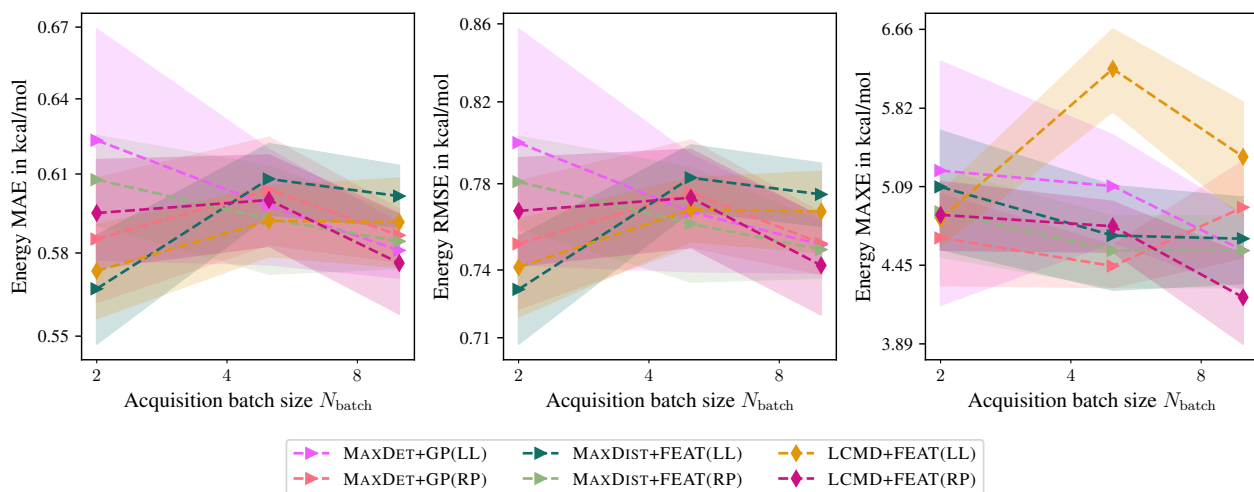


Figure 3 Comparison of the last layer and random projections gradient feature maps on the aspirin molecule data from MD17.¹⁻⁴ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(10, 100; \{2, 5, 10\})_{\text{MD17}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

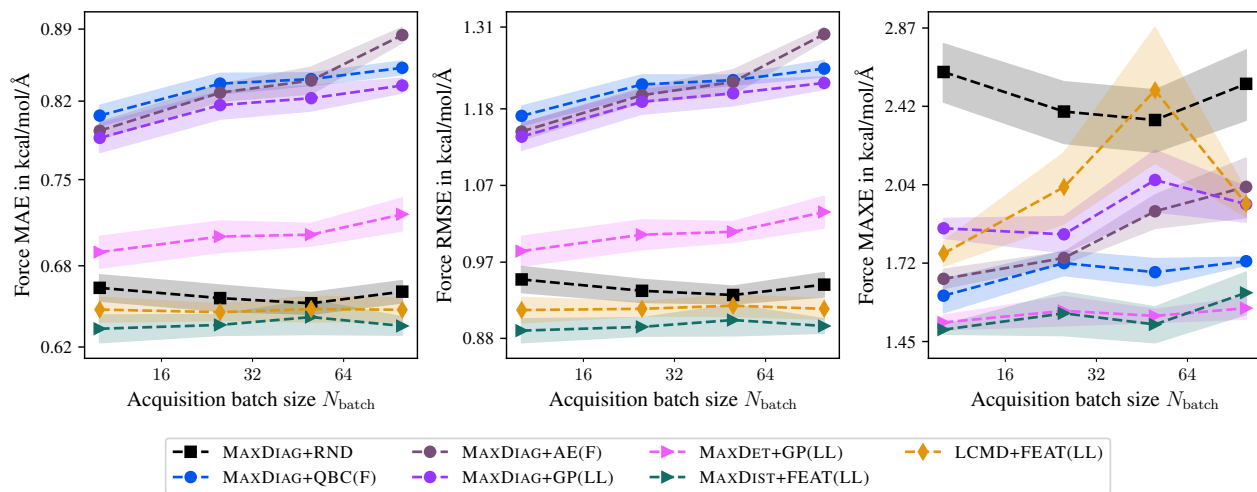


Figure 4 Dependence of the mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces on the acquired batch size N_{batch} . All errors are evaluated for the last BMDAL step on the aspirin molecule data from MD17.¹⁻⁴ All corresponding values are given for the $(100, 1000; \{10, 25, 50, 100\})_{\text{MD17}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

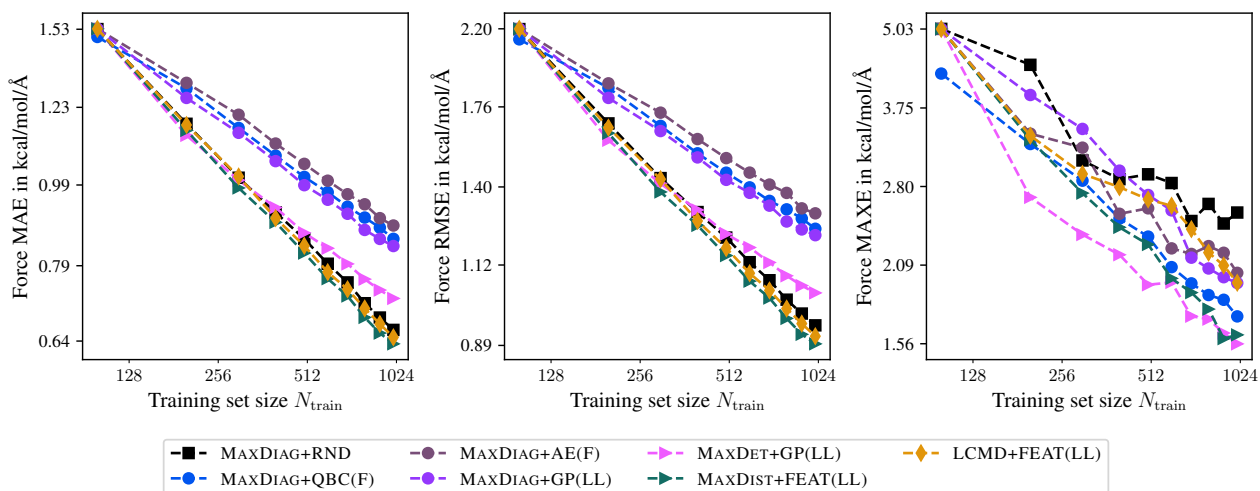


Figure 5 Learning curves for the aspirin molecule data from MD17.¹⁻⁴ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(100, 1000; 100)_{\text{MD17}}$ BMDAL experiment. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

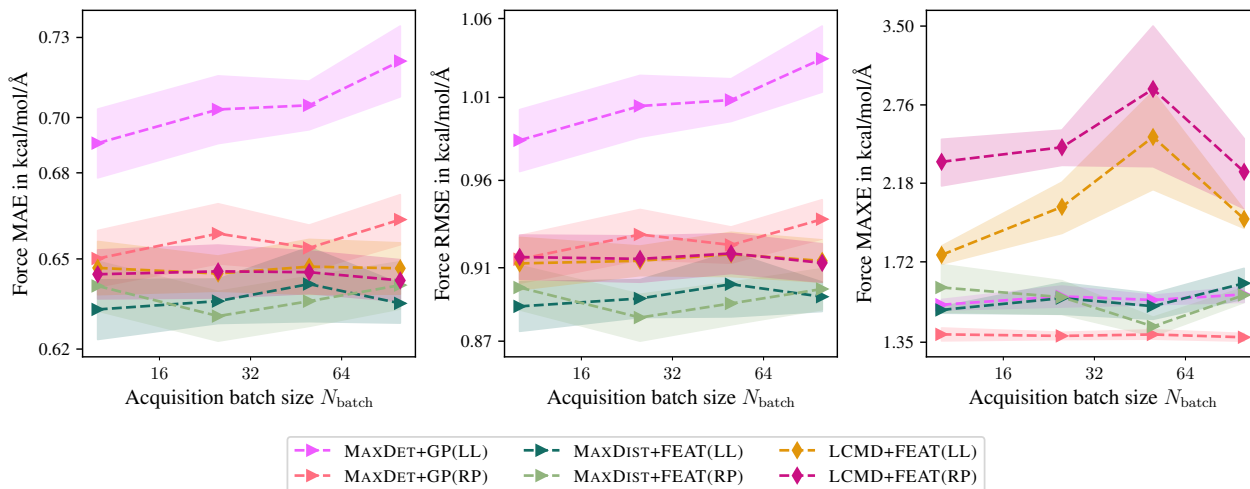


Figure 6 Comparison of the last layer and random projections gradient feature maps on the aspirin molecule data from MD17.¹⁻⁴ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(100, 1000; \{10, 25, 50, 100\})_{\text{MD17}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

2 QM9

Here, additional results on the QM9 data set⁵⁻⁷ are presented. Table 5 and Table 6 present the numerical values for the mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients, as well as 95% quantile, evaluated on test data for total energies. While Table 5 provides with the errors averaged over the learning curve, Table 6 shows the performance of the final model. All results are given for the $(1000, 10000; 250)_{\text{QM9}}$ BMDAL experiment. The respective results for $(5000, 25000; 250)_{\text{QM9}}$ can be found in Table 7 and Table 8. Fig. 7 and Fig. 8 present the learning curves for the $(5000, 25000; 250)_{\text{QM9}}$ experiment.

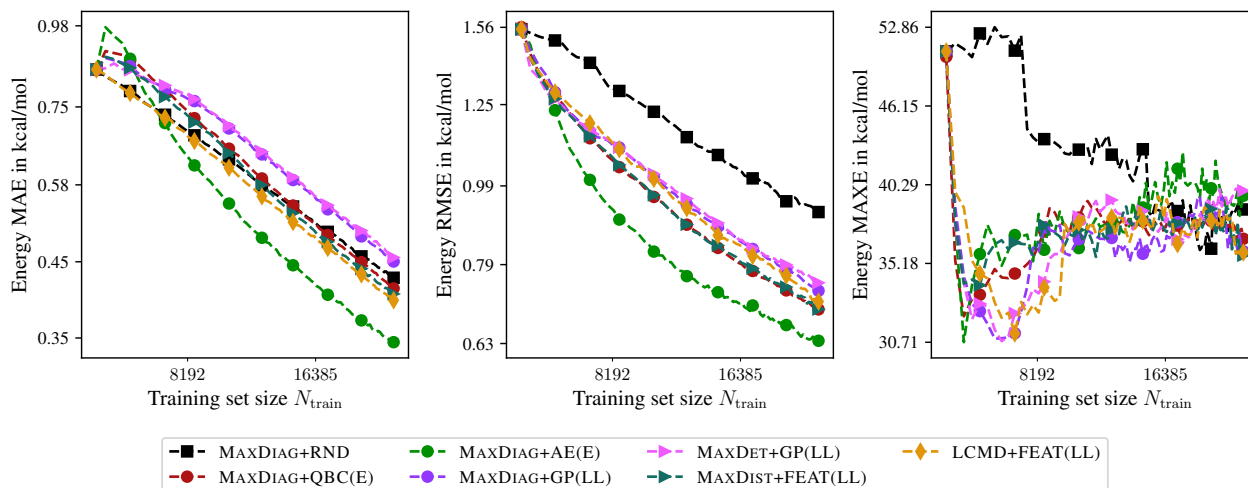


Figure 7 Learning curves for the QM9 data set.⁵⁻⁷ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the atomization energies are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(5000, 25000; 250)_{\text{QM9}}$ BMDAL experiment. The markers show only a subset of all BMDAL steps for better visibility, while the lines use all steps. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

Table 5 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies evaluated on test data. All values are computed for (1000,10000;250)_{QM9} as an average over AL steps. The energy errors are given in kcal/mol. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy					Time		
	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MAXDIAG+RND	0.91 (0.01)	1.69 (0.04)	60.88 (4.96)	0.00 (0.00)	-0.00 (0.00)	2.45 (0.03)	0.01 (0.00)	0.02 (0.01)
MAXDIAG+AE(E)	0.99 (0.02)	1.38 (0.02)	40.52 (3.71)	1.00 (0.00)	1.00 (0.00)	2.52 (0.04)	25.88 (0.40)	0.02 (0.00)
MAXDIAG+QBC(E)	1.11 (0.01)	1.53 (0.02)	38.50 (2.82)	0.28 (0.03)	0.17 (0.01)	2.72 (0.03)	45.78 (1.18)	0.02 (0.00)
MAXDIAG+GP(LL)	1.16 (0.01)	1.60 (0.02)	37.58 (2.00)	0.22 (0.01)	0.10 (0.01)	3.01 (0.04)	28.04 (0.36)	0.01 (0.00)
MAXDIAG+GP(RP)	1.15 (0.01)	1.60 (0.02)	38.73 (2.53)	0.25 (0.01)	0.11 (0.01)	2.99 (0.04)	40.00 (1.48)	0.01 (0.00)
MAXDET+GP(LL)	1.15 (0.01)	1.58 (0.02)	38.89 (2.28)	0.24 (0.01)	0.12 (0.01)	2.99 (0.03)	28.43 (0.47)	12.03 (0.37)
MAXDET+GP(RP)	1.14 (0.01)	1.57 (0.02)	38.77 (2.52)	0.25 (0.01)	0.12 (0.01)	2.97 (0.03)	38.75 (1.86)	12.55 (1.47)
MAXDIST+GP(LL)	1.07 (0.01)	1.48 (0.01)	39.12 (2.40)	0.24 (0.01)	0.10 (0.01)	2.75 (0.03)	27.60 (0.34)	175.37 (2.79)
MAXDIST+GP(RP)	1.09 (0.01)	1.50 (0.01)	38.68 (2.51)	0.25 (0.01)	0.10 (0.01)	2.79 (0.03)	39.77 (2.22)	169.87 (9.91)
MAXDIST+FEAT(LL)	1.05 (0.01)	1.49 (0.02)	40.32 (3.12)	0.14 (0.01)	0.07 (0.01)	2.72 (0.03)	33.32 (4.72)	231.35 (45.52)
MAXDIST+FEAT(RP)	1.05 (0.01)	1.50 (0.01)	40.91 (3.89)	0.14 (0.02)	0.07 (0.01)	2.74 (0.03)	37.68 (0.65)	162.21 (4.20)
LCMD+FEAT(LL)	0.91 (0.01)	1.44 (0.02)	47.96 (6.88)	0.29 (0.01)	0.16 (0.00)	2.44 (0.03)	27.35 (0.65)	180.74 (4.93)
LCMD+FEAT(RP)	0.91 (0.01)	1.46 (0.03)	55.52 (8.17)	0.28 (0.01)	0.16 (0.01)	2.42 (0.04)	38.32 (1.03)	172.90 (4.86)

Table 6 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies evaluated on test data. All values are computed employing the final model obtained for the (1000,10000;250)_{QM9} experiment. The energy errors are given in kcal/mol. Standard errors on the mean are given in parentheses.

Method	Energy						
	MAE	RMSE	MAXE	PCC	SRC	95%	
MAXDIAG+RND	0.64 (0.01)	1.25 (0.02)	62.77 (7.42)	0.00 (0.00)	0.00 (0.00)	1.75 (0.03)	
MAXDIAG+AE(E)	0.55 (0.01)	0.83 (0.01)	40.26 (4.51)	1.00 (0.00)	1.00 (0.00)	1.35 (0.02)	
MAXDIAG+QBC(E)	0.71 (0.01)	1.01 (0.01)	39.42 (3.25)	0.40 (0.06)	0.15 (0.01)	1.70 (0.02)	
MAXDIAG+GP(LL)	0.79 (0.00)	1.11 (0.01)	39.91 (1.85)	0.23 (0.02)	0.03 (0.00)	2.03 (0.01)	
MAXDIAG+GP(RP)	0.78 (0.01)	1.10 (0.01)	41.23 (2.66)	0.28 (0.03)	0.03 (0.00)	1.98 (0.01)	
MAXDET+GP(LL)	0.80 (0.00)	1.12 (0.00)	40.55 (2.69)	0.22 (0.02)	0.03 (0.01)	2.04 (0.01)	
MAXDET+GP(RP)	0.80 (0.01)	1.11 (0.00)	40.79 (3.02)	0.26 (0.02)	0.04 (0.01)	2.02 (0.01)	
MAXDIST+GP(LL)	0.70 (0.00)	1.00 (0.00)	39.73 (2.25)	0.26 (0.01)	0.06 (0.01)	1.76 (0.01)	
MAXDIST+GP(RP)	0.71 (0.01)	1.01 (0.01)	39.32 (3.51)	0.31 (0.02)	0.05 (0.01)	1.78 (0.01)	
MAXDIST+FEAT(LL)	0.68 (0.01)	1.00 (0.01)	40.64 (3.14)	0.18 (0.02)	0.05 (0.00)	1.73 (0.02)	
MAXDIST+FEAT(RP)	0.68 (0.00)	1.00 (0.01)	44.80 (4.54)	0.18 (0.03)	0.05 (0.00)	1.72 (0.01)	
LCMD+FEAT(LL)	0.62 (0.01)	1.01 (0.01)	40.67 (3.89)	0.30 (0.02)	0.16 (0.00)	1.65 (0.01)	
LCMD+FEAT(RP)	0.62 (0.01)	1.02 (0.03)	47.35 (6.63)	0.31 (0.02)	0.17 (0.00)	1.64 (0.02)	

Table 7 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies evaluated on test data. All values are computed for (5000, 25000; 250)_{QM9} as an average over AL steps. The energy errors are given in kcal/mol. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy						Time	
	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MAXDIAG+RND	0.56 (0.01)	1.12 (0.02)	41.55 (3.59)	0.00 (0.00)	-0.00 (0.00)	1.54 (0.02)	0.00 (0.00)	0.02 (0.00)
MAXDIAG+AE(E)	0.49 (0.00)	0.79 (0.03)	38.72 (5.46)	1.00 (0.00)	1.00 (0.00)	1.25 (0.01)	25.41 (0.61)	0.02 (0.00)
MAXDIAG+QBC(E)	0.58 (0.00)	0.89 (0.02)	37.34 (4.75)	0.42 (0.03)	0.24 (0.01)	1.40 (0.01)	48.58 (1.02)	0.02 (0.01)
MAXDIAG+GP(LL)	0.61 (0.00)	0.93 (0.02)	36.65 (4.90)	0.30 (0.01)	0.12 (0.00)	1.60 (0.01)	29.78 (0.57)	0.01 (0.00)
MAXDIAG+GP(RP)	0.61 (0.00)	0.92 (0.02)	37.71 (5.25)	0.33 (0.01)	0.12 (0.00)	1.58 (0.01)	38.58 (0.50)	0.01 (0.00)
MAXDET+GP(LL)	0.62 (0.00)	0.94 (0.02)	37.38 (4.59)	0.30 (0.01)	0.12 (0.00)	1.61 (0.01)	30.59 (0.82)	12.70 (1.17)
MAXDET+GP(RP)	0.61 (0.00)	0.93 (0.02)	38.02 (4.89)	0.34 (0.01)	0.13 (0.00)	1.60 (0.01)	38.80 (0.57)	10.29 (0.29)
MAXDIST+GP(LL)	0.57 (0.00)	0.89 (0.02)	38.98 (4.87)	0.37 (0.02)	0.14 (0.00)	1.49 (0.01)	30.07 (0.76)	540.91 (28.03)
MAXDIST+GP(RP)	0.57 (0.00)	0.89 (0.02)	40.35 (5.42)	0.39 (0.02)	0.14 (0.00)	1.48 (0.01)	39.31 (0.57)	456.22 (10.12)
MAXDIST+FEAT(LL)	0.56 (0.00)	0.89 (0.02)	37.32 (4.65)	0.23 (0.01)	0.10 (0.00)	1.47 (0.01)	30.98 (1.13)	580.00 (37.60)
MAXDIST+FEAT(RP)	0.56 (0.00)	0.88 (0.02)	38.11 (4.61)	0.23 (0.01)	0.10 (0.00)	1.47 (0.01)	40.20 (0.70)	484.96 (23.72)
LCMD+FEAT(LL)	0.54 (0.00)	0.92 (0.03)	36.95 (6.30)	0.30 (0.01)	0.17 (0.01)	1.44 (0.01)	35.06 (4.23)	695.98 (114.62)
LCMD+FEAT(RP)	0.54 (0.01)	0.92 (0.02)	37.78 (4.63)	0.30 (0.00)	0.18 (0.00)	1.44 (0.02)	47.81 (5.30)	695.31 (137.75)

Table 8 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies evaluated on test data. All values are computed employing the final model obtained for the (5000,25000;250)_{QM9} experiment. The energy errors are given in kcal/mol. Standard errors on the mean are given in parentheses.

Method	Energy					
	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	0.43 (0.01)	0.92 (0.02)	38.61 (5.42)	-0.00 (0.00)	-0.00 (0.00)	1.18 (0.01)
MAXDIAG+AE(E)	0.35 (0.00)	0.64 (0.03)	39.53 (5.49)	1.00 (0.00)	1.00 (0.00)	0.87 (0.01)
MAXDIAG+QBC(E)	0.41 (0.00)	0.69 (0.02)	36.71 (4.81)	0.51 (0.04)	0.23 (0.01)	0.97 (0.01)
MAXDIAG+GP(LL)	0.45 (0.00)	0.73 (0.02)	35.92 (4.71)	0.30 (0.02)	0.02 (0.00)	1.17 (0.00)
MAXDIAG+GP(RP)	0.45 (0.00)	0.72 (0.02)	38.00 (4.84)	0.35 (0.02)	0.05 (0.01)	1.15 (0.01)
MAXDET+GP(LL)	0.46 (0.00)	0.75 (0.02)	39.88 (4.87)	0.31 (0.01)	0.02 (0.01)	1.18 (0.01)
MAXDET+GP(RP)	0.45 (0.00)	0.73 (0.02)	37.12 (4.82)	0.36 (0.02)	0.05 (0.01)	1.15 (0.01)
MAXDIST+GP(LL)	0.41 (0.00)	0.70 (0.02)	37.93 (4.28)	0.43 (0.02)	0.08 (0.00)	1.05 (0.01)
MAXDIST+GP(RP)	0.41 (0.00)	0.70 (0.03)	39.17 (5.31)	0.46 (0.02)	0.09 (0.00)	1.04 (0.01)
MAXDIST+FEAT(LL)	0.40 (0.00)	0.70 (0.02)	35.67 (4.53)	0.28 (0.01)	0.07 (0.00)	1.05 (0.01)
MAXDIST+FEAT(RP)	0.40 (0.00)	0.69 (0.02)	38.31 (4.94)	0.29 (0.01)	0.08 (0.00)	1.03 (0.01)
LCMD+FEAT(LL)	0.40 (0.00)	0.71 (0.03)	35.85 (5.17)	0.31 (0.01)	0.15 (0.00)	1.05 (0.00)
LCMD+FEAT(RP)	0.40 (0.00)	0.73 (0.04)	39.83 (6.90)	0.32 (0.00)	0.17 (0.00)	1.06 (0.01)

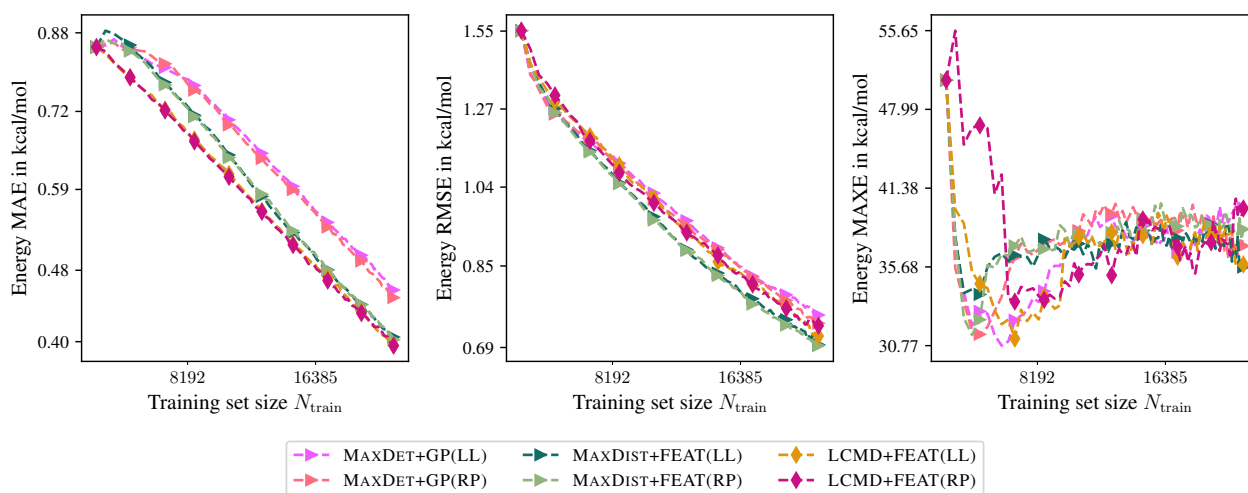


Figure 8 Comparison of the last layer and random projections gradient feature maps on the QM9 data set.⁵⁻⁷ The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the atomization energies are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(5000, 25000; 250)_{\text{QM9}}$ BMDAL experiment. The markers show only a subset of all BMDAL steps for better visibility, while the lines use all steps. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

3 TiO₂

Here, additional results on the TiO₂ data set^{8,9} are presented. Table 9 and Table 10 present the numerical values for the mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients, as well as 95% quantile, evaluated on test data for total energies and forces. While Table 9 provides with the errors averaged over the learning curve, Table 10 shows the performance of the final model. All results are given for the (10, 250; 10)_{TiO₂} BMDAL experiment. The respective results for (500, 2500; 250)_{TiO₂} can be found in Table 11 and Table 12.

Fig. 9, Fig. 10, and Fig. 11 complement the results obtained in the main manuscript on atomic forces with the results for the total energy. Fig. 12, Fig. 13, and Fig. 14 present the batch size dependence and the learning curves for (500, 2500; {50, 100, 250})_{TiO₂} or (500, 2500; 250)_{TiO₂}, respectively.

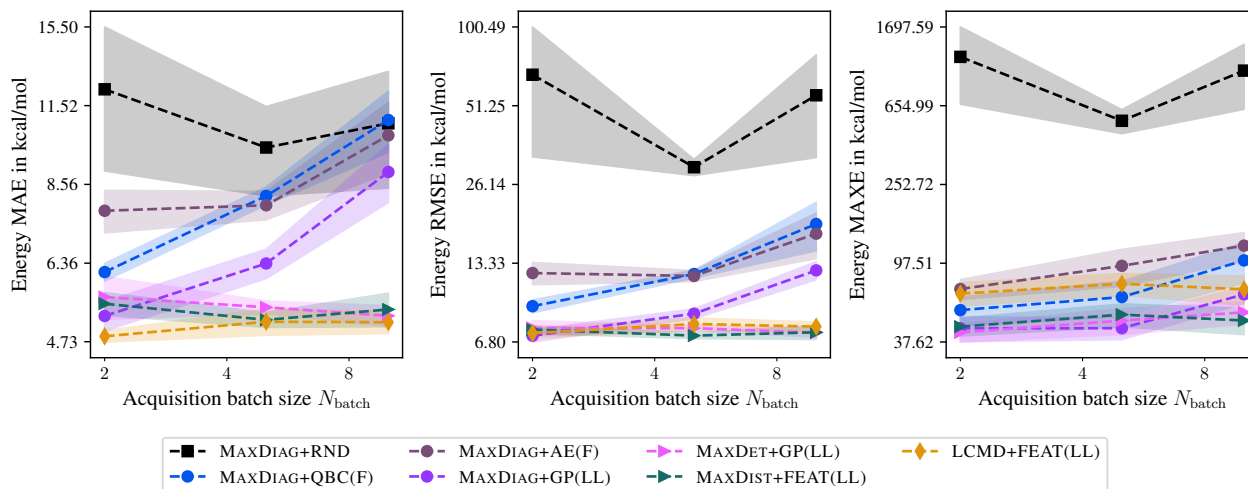


Figure 9 Dependence of the mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy on the acquired batch size N_{batch} . All errors are evaluated for the last BMDAL step on the TiO₂ data set.^{8,9} All corresponding values are given for the (10, 250; {2, 5, 10})_{TiO₂} BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

Table 9 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed for (10,250;10)_{TiO₂} as an average over AL steps. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy					Force					Time			
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MaxDiag+RND	25.49 (3.69)	109.70 (26.01)	1687.74 (426.35)	-0.01 (0.01)	0.00 (0.01)	92.09 (12.59)	6.08 (0.41)	35.68 (6.93)	360.23 (66.47)	-0.01 (0.01)	-0.01 (0.01)	15.57 (1.32)	0.00 (0.00)	0.00 (0.00)
MaxDiag+AE(E)	32.50 (7.43)	54.45 (11.05)	400.36 (69.10)	1.00 (0.00)	1.00 (0.00)	107.93 (23.55)	7.22 (0.65)	14.76 (1.75)	103.62 (18.02)	0.34 (0.04)	0.30 (0.04)	15.62 (1.30)	15.79 (2.01)	0.01 (0.01)
MaxDiag+AE(F)	45.84 (8.51)	89.50 (15.77)	777.08 (182.93)	0.30 (0.04)	0.32 (0.05)	173.19 (33.82)	8.17 (0.44)	15.07 (1.16)	85.93 (13.18)	1.00 (0.00)	1.00 (0.00)	16.45 (0.98)	18.37 (4.05)	0.00 (0.00)
MaxDiag+QBC(E)	42.65 (8.43)	70.68 (11.82)	454.54 (63.86)	0.52 (0.07)	0.63 (0.05)	130.98 (27.46)	7.45 (0.32)	16.54 (1.08)	133.13 (13.68)	0.30 (0.05)	0.33 (0.05)	15.44 (0.67)	33.81 (0.69)	0.00 (0.00)
MaxDiag+QBC(F)	47.79 (7.00)	91.47 (14.62)	676.32 (146.42)	0.22 (0.05)	0.40 (0.04)	177.38 (36.72)	8.07 (0.25)	14.76 (0.83)	86.69 (12.00)	0.73 (0.03)	0.84 (0.02)	15.83 (0.77)	37.43 (0.40)	0.00 (0.00)
MaxDiag+GP(LL)	90.74 (26.09)	139.23 (38.49)	724.86 (177.08)	0.31 (0.05)	0.44 (0.05)	288.28 (87.02)	7.66 (0.55)	16.47 (1.49)	130.15 (18.58)	0.48 (0.05)	0.55 (0.03)	16.89 (1.16)	7.09 (0.14)	0.00 (0.00)
MaxDiag+GP(RP)	76.63 (22.47)	118.28 (33.41)	657.21 (157.42)	0.32 (0.05)	0.45 (0.04)	240.68 (73.60)	7.57 (0.46)	16.78 (1.40)	137.10 (20.94)	0.46 (0.04)	0.53 (0.03)	17.37 (0.99)	8.69 (0.14)	0.00 (0.00)
MaxDET+GP(LL)	32.98 (6.91)	53.56 (10.28)	337.58 (57.94)	0.26 (0.03)	0.38 (0.03)	106.66 (21.71)	5.35 (0.24)	11.15 (1.00)	82.02 (14.27)	0.54 (0.04)	0.34 (0.03)	11.33 (0.50)	7.58 (0.45)	0.04 (0.00)
MaxDET+GP(RP)	32.07 (6.70)	52.47 (9.86)	323.57 (50.24)	0.27 (0.04)	0.39 (0.03)	105.54 (21.76)	5.37 (0.27)	11.09 (1.04)	79.57 (14.23)	0.54 (0.04)	0.31 (0.02)	11.27 (0.50)	9.14 (0.22)	0.04 (0.00)
MaxDist+GP(LL)	30.46 (5.78)	51.03 (9.00)	368.66 (69.51)	0.18 (0.05)	0.18 (0.04)	99.07 (17.72)	5.20 (0.26)	11.20 (1.17)	84.86 (16.19)	0.58 (0.04)	0.42 (0.05)	10.94 (0.51)	8.39 (0.83)	0.48 (0.09)
MaxDist+GP(RP)	28.88 (5.95)	48.94 (8.33)	333.94 (42.94)	0.16 (0.05)	0.17 (0.05)	94.06 (18.45)	5.16 (0.23)	11.35 (1.08)	89.06 (15.35)	0.59 (0.04)	0.43 (0.06)	10.83 (0.47)	10.02 (0.23)	0.43 (0.03)
MaxDist+FEAT(LL)	30.55 (6.46)	52.60 (10.12)	405.80 (63.28)	0.18 (0.06)	0.16 (0.06)	101.15 (20.89)	5.55 (0.30)	12.64 (1.15)	100.52 (14.20)	0.34 (0.06)	0.24 (0.05)	12.06 (0.68)	8.07 (0.44)	0.44 (0.04)
MaxDist+FEAT(RP)	28.33 (6.27)	50.95 (10.61)	399.13 (67.16)	0.17 (0.06)	0.14 (0.05)	102.26 (25.45)	5.47 (0.30)	12.52 (1.19)	99.20 (15.90)	0.33 (0.05)	0.24 (0.05)	12.01 (0.56)	11.65 (0.87)	0.53 (0.07)
LCMD+FEAT(LL)	16.18 (2.24)	34.50 (5.39)	372.09 (68.08)	0.37 (0.04)	0.28 (0.04)	60.95 (10.60)	4.97 (0.28)	13.24 (1.30)	119.18 (17.42)	0.49 (0.06)	0.18 (0.03)	10.80 (0.60)	7.44 (0.36)	0.43 (0.03)
LCMD+FEAT(RP)	17.02 (2.63)	36.05 (5.10)	399.53 (71.21)	0.37 (0.04)	0.25 (0.03)	62.21 (12.21)	5.00 (0.27)	13.63 (1.42)	127.16 (22.02)	0.45 (0.06)	0.18 (0.04)	10.98 (0.63)	29.22 (19.49)	0.79 (0.35)

Table 10 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed employing the final model obtained for the (10,250;10) TiO_2 experiment. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. Standard errors on the mean are given in parentheses.

Method	Energy					Force						
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	10.77 (2.33)	56.04 (23.13)	1001.04 (373.47)	-0.02 (0.01)	-0.00 (0.01)	25.52 (3.65)	3.81 (0.36)	24.68 (7.17)	219.66 (48.89)	-0.01 (0.01)	-0.01 (0.01)	7.04 (0.31)
MAXDIAG+AE(E)	6.87 (0.43)	9.34 (0.44)	46.81 (7.65)	1.00 (0.00)	1.00 (0.00)	19.79 (0.81)	3.78 (0.14)	6.43 (0.37)	41.81 (4.51)	0.25 (0.03)	0.16 (0.05)	7.06 (0.26)
MAXDIAG+AE(F)	10.30 (1.36)	17.16 (3.29)	120.57 (20.60)	0.12 (0.02)	0.11 (0.04)	35.37 (7.27)	4.10 (0.08)	6.30 (0.19)	33.74 (5.10)	1.00 (0.00)	1.00 (0.00)	6.40 (0.11)
MAXDIAG+QBC(E)	8.74 (0.79)	11.76 (1.08)	57.98 (7.01)	0.46 (0.04)	0.48 (0.04)	21.27 (2.59)	4.07 (0.06)	7.00 (0.22)	44.58 (4.01)	0.21 (0.04)	0.11 (0.05)	7.91 (0.22)
MAXDIAG+QBC(F)	10.91 (1.25)	18.64 (3.80)	100.88 (24.30)	0.12 (0.03)	0.20 (0.02)	36.27 (9.16)	4.32 (0.13)	6.62 (0.24)	33.65 (4.20)	0.74 (0.02)	0.84 (0.02)	6.91 (0.32)
MAXDIAG+GP(LL)	8.97 (0.98)	12.54 (0.95)	66.93 (6.93)	0.24 (0.03)	0.38 (0.04)	25.46 (2.50)	3.59 (0.06)	6.16 (0.15)	41.00 (2.04)	0.58 (0.04)	0.49 (0.01)	6.93 (0.19)
MAXDIAG+GP(RP)	10.77 (1.39)	14.41 (1.53)	71.04 (5.92)	0.24 (0.02)	0.38 (0.03)	29.28 (2.70)	3.69 (0.03)	6.39 (0.20)	42.72 (3.72)	0.58 (0.03)	0.48 (0.04)	7.31 (0.19)
MAXDET+GP(LL)	5.21 (0.20)	7.36 (0.27)	53.84 (7.21)	0.30 (0.03)	0.29 (0.02)	15.06 (0.35)	2.86 (0.04)	5.10 (0.24)	36.96 (5.83)	0.65 (0.07)	0.10 (0.02)	5.28 (0.12)
MAXDET+GP(RP)	5.59 (0.18)	7.73 (0.28)	45.56 (7.78)	0.25 (0.05)	0.32 (0.02)	16.15 (0.76)	2.88 (0.06)	4.92 (0.18)	33.55 (4.00)	0.66 (0.03)	0.10 (0.02)	5.32 (0.06)
MAXDIST+GP(LL)	4.89 (0.18)	7.02 (0.24)	44.24 (9.09)	0.15 (0.02)	0.05 (0.03)	14.76 (0.41)	2.67 (0.02)	4.78 (0.23)	34.77 (5.14)	0.71 (0.04)	0.38 (0.05)	4.83 (0.02)
MAXDIST+GP(RP)	5.21 (0.22)	7.40 (0.31)	43.83 (3.02)	0.15 (0.05)	0.01 (0.04)	15.75 (0.62)	2.69 (0.02)	4.72 (0.15)	36.06 (3.78)	0.77 (0.03)	0.38 (0.04)	4.76 (0.05)
MAXDIST+FEAT(LL)	5.34 (0.34)	7.39 (0.44)	48.75 (7.64)	0.23 (0.03)	0.15 (0.02)	15.06 (0.97)	3.00 (0.06)	5.26 (0.13)	35.66 (2.89)	0.53 (0.05)	0.23 (0.00)	6.02 (0.14)
MAXDIST+FEAT(RP)	5.79 (0.58)	8.15 (0.91)	48.10 (9.02)	0.20 (0.06)	0.10 (0.04)	18.12 (2.56)	3.07 (0.10)	5.38 (0.24)	35.07 (4.22)	0.49 (0.05)	0.23 (0.01)	6.15 (0.14)
LCMD+FEAT(LL)	5.09 (0.21)	7.75 (0.31)	71.09 (12.35)	0.42 (0.03)	0.25 (0.03)	15.10 (0.41)	2.80 (0.04)	6.11 (0.27)	51.81 (5.18)	0.52 (0.05)	0.10 (0.02)	5.54 (0.20)
LCMD+FEAT(RP)	4.75 (0.24)	7.43 (0.24)	85.70 (13.10)	0.42 (0.03)	0.26 (0.02)	14.00 (0.55)	2.77 (0.07)	6.19 (0.26)	51.92 (4.60)	0.52 (0.06)	0.13 (0.02)	5.56 (0.08)

Table 11 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed for (500,2500;250)TiO₂ as an average over AL steps. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy					Force					Time			
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MaxDiag+RND	3.08 (0.17)	11.96 (1.96)	228.33 (53.46)	-0.00 (0.01)	-0.01 (0.02)	7.76 (0.44)	1.96 (0.06)	9.44 (1.48)	97.07 (21.34)	-0.01 (0.02)	-0.01 (0.03)	4.06 (0.11)	0.00 (0.00)	0.00 (0.00)
MaxDiag+AE(E)	2.58 (0.10)	6.07 (0.43)	101.41 (18.99)	1.00 (0.00)	1.00 (0.00)	6.98 (0.27)	1.90 (0.04)	5.40 (0.49)	49.15 (7.04)	0.61 (0.06)	0.09 (0.03)	4.08 (0.07)	12.70 (1.09)	0.00 (0.00)
MaxDiag+AE(F)	3.04 (0.11)	6.85 (0.42)	104.58 (19.85)	0.44 (0.07)	-0.03 (0.02)	9.37 (0.55)	1.80 (0.03)	5.18 (0.46)	48.33 (7.16)	1.00 (0.00)	1.00 (0.00)	3.44 (0.08)	13.80 (1.26)	0.00 (0.00)
MaxDiag+QBC(E)	2.58 (0.09)	5.80 (0.34)	89.25 (16.40)	0.64 (0.05)	0.64 (0.02)	6.37 (0.33)	1.92 (0.04)	5.42 (0.44)	47.87 (6.48)	0.55 (0.04)	-0.01 (0.03)	4.13 (0.07)	31.22 (0.41)	0.00 (0.00)
MaxDiag+QBC(F)	2.81 (0.11)	6.18 (0.32)	88.99 (15.73)	0.59 (0.05)	0.06 (0.02)	7.69 (0.49)	1.78 (0.04)	5.15 (0.40)	46.68 (5.81)	0.88 (0.01)	0.90 (0.01)	3.47 (0.07)	31.06 (0.67)	0.00 (0.00)
MaxDiag+GP(LL)	2.86 (0.13)	6.38 (0.45)	100.10 (18.36)	0.54 (0.07)	0.33 (0.03)	8.22 (0.45)	1.86 (0.04)	5.35 (0.48)	48.72 (6.86)	0.82 (0.03)	0.24 (0.02)	3.83 (0.09)	7.25 (0.07)	0.00 (0.00)
MaxDiag+GP(RP)	2.72 (0.11)	6.18 (0.42)	98.01 (17.59)	0.52 (0.07)	0.35 (0.03)	7.95 (0.38)	1.85 (0.04)	5.32 (0.46)	48.68 (6.60)	0.83 (0.02)	0.26 (0.02)	3.82 (0.08)	10.00 (0.65)	0.00 (0.00)
MaxDef+GP(LL)	2.60 (0.10)	6.11 (0.41)	100.84 (19.84)	0.58 (0.06)	0.42 (0.02)	7.42 (0.36)	1.74 (0.03)	5.30 (0.53)	49.78 (7.38)	0.82 (0.02)	0.08 (0.02)	3.50 (0.06)	8.26 (0.34)	1.93 (0.06)
MaxDef+GP(RP)	2.51 (0.11)	5.92 (0.42)	98.75 (18.11)	0.59 (0.05)	0.41 (0.02)	7.11 (0.34)	1.72 (0.03)	5.26 (0.50)	48.85 (7.06)	0.83 (0.02)	0.10 (0.02)	3.50 (0.05)	9.41 (0.14)	1.77 (0.04)
MaxDist+GP(LL)	2.68 (0.13)	6.28 (0.43)	101.66 (20.13)	0.49 (0.08)	0.01 (0.02)	8.04 (0.46)	1.69 (0.03)	5.16 (0.42)	48.66 (6.19)	0.85 (0.02)	0.62 (0.01)	3.39 (0.05)	8.12 (0.19)	5.64 (0.24)
MaxDist+GP(RP)	2.63 (0.11)	6.18 (0.43)	100.73 (19.39)	0.52 (0.07)	0.03 (0.02)	7.83 (0.46)	1.69 (0.03)	5.18 (0.45)	48.63 (6.53)	0.87 (0.02)	0.61 (0.01)	3.40 (0.06)	13.12 (1.44)	8.15 (1.35)
MaxDist+FEAT(LL)	2.64 (0.09)	6.11 (0.42)	99.59 (19.17)	0.47 (0.06)	0.17 (0.02)	7.53 (0.37)	1.76 (0.02)	5.23 (0.46)	48.32 (6.56)	0.75 (0.03)	0.32 (0.02)	3.66 (0.06)	11.39 (2.12)	7.14 (0.45)
MaxDist+FEAT(RP)	2.61 (0.10)	6.10 (0.42)	99.61 (18.56)	0.49 (0.07)	0.16 (0.02)	7.42 (0.32)	1.74 (0.03)	5.22 (0.47)	48.38 (6.84)	0.78 (0.02)	0.33 (0.02)	3.65 (0.07)	12.57 (0.74)	7.80 (1.55)
LCMD+FEAT(LL)	2.57 (0.12)	6.12 (0.39)	101.17 (18.94)	0.50 (0.06)	0.21 (0.03)	7.38 (0.40)	1.71 (0.03)	5.23 (0.44)	49.26 (6.49)	0.75 (0.03)	0.30 (0.02)	3.65 (0.06)	38.58 (28.39)	42.56 (35.25)
LCMD+FEAT(RP)	2.56 (0.15)	6.09 (0.50)	101.36 (21.09)	0.51 (0.07)	0.20 (0.03)	7.28 (0.41)	1.71 (0.03)	5.25 (0.47)	48.68 (6.93)	0.78 (0.02)	0.30 (0.02)	3.65 (0.06)	12.91 (2.27)	6.49 (0.34)

Table 12 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed employing the final model obtained for the (500,2500;250)_{TiO₂} experiment. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. Standard errors on the mean are given in parentheses.

Method	Energy						Force					
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	1.94 (0.16)	8.25 (2.59)	178.60 (82.59)	-0.01 (0.01)	-0.01 (0.02)	4.64 (0.14)	1.66 (0.11)	11.11 (4.94)	152.93 (94.70)	-0.01 (0.02)	-0.00 (0.02)	3.56 (0.13)
MAXDIAG+AE(E)	1.41 (0.06)	3.10 (0.22)	54.51 (10.02)	1.00 (0.00)	1.00 (0.00)	3.43 (0.18)	1.46 (0.03)	4.39 (0.58)	40.55 (6.57)	0.73 (0.05)	0.13 (0.04)	3.34 (0.08)
MAXDIAG+AE(F)	1.99 (0.11)	3.86 (0.09)	57.57 (11.20)	0.57 (0.07)	-0.09 (0.01)	6.53 (0.51)	1.35 (0.03)	4.10 (0.45)	40.36 (6.39)	1.00 (0.00)	1.00 (0.00)	2.73 (0.05)
MAXDIAG+QBC(E)	1.50 (0.07)	3.17 (0.28)	53.36 (11.44)	0.61 (0.07)	0.60 (0.02)	3.28 (0.09)	1.52 (0.03)	4.44 (0.47)	38.69 (5.80)	0.57 (0.04)	-0.00 (0.02)	3.40 (0.04)
MAXDIAG+QBC(F)	1.64 (0.06)	3.22 (0.19)	48.95 (9.08)	0.71 (0.04)	-0.03 (0.03)	4.12 (0.15)	1.34 (0.02)	4.16 (0.43)	38.04 (5.49)	0.89 (0.01)	0.92 (0.01)	2.73 (0.07)
MAXDIAG+GP(LL)	1.56 (0.09)	3.05 (0.33)	53.02 (12.34)	0.67 (0.06)	0.28 (0.02)	4.44 (0.34)	1.42 (0.03)	4.42 (0.56)	41.50 (7.16)	0.83 (0.06)	0.16 (0.02)	3.04 (0.06)
MAXDIAG+GP(RP)	1.42 (0.05)	2.83 (0.24)	50.71 (7.06)	0.66 (0.05)	0.29 (0.02)	3.90 (0.15)	1.41 (0.03)	4.35 (0.53)	39.67 (6.49)	0.88 (0.01)	0.27 (0.02)	2.97 (0.04)
MAXDET+GP(LL)	1.52 (0.07)	3.20 (0.21)	54.74 (9.58)	0.69 (0.02)	0.35 (0.02)	4.17 (0.26)	1.35 (0.02)	4.30 (0.53)	40.42 (6.63)	0.83 (0.02)	0.00 (0.03)	2.79 (0.04)
MAXDET+GP(RP)	1.38 (0.10)	2.80 (0.32)	46.51 (13.29)	0.69 (0.04)	0.30 (0.02)	3.76 (0.24)	1.33 (0.03)	4.47 (0.59)	43.01 (6.80)	0.86 (0.02)	0.00 (0.05)	2.78 (0.02)
MAXDIST+GP(LL)	1.58 (0.10)	3.28 (0.29)	54.74 (13.09)	0.66 (0.07)	-0.03 (0.03)	4.59 (0.28)	1.29 (0.04)	4.26 (0.40)	39.65 (4.69)	0.85 (0.04)	0.65 (0.01)	2.72 (0.03)
MAXDIST+GP(RP)	1.52 (0.04)	3.18 (0.28)	56.24 (13.95)	0.69 (0.06)	-0.00 (0.01)	4.56 (0.22)	1.28 (0.03)	4.26 (0.40)	39.17 (5.42)	0.92 (0.01)	0.65 (0.00)	2.70 (0.04)
MAXDIST+FEAT(LL)	1.56 (0.06)	2.98 (0.24)	49.94 (11.10)	0.61 (0.04)	0.11 (0.04)	4.52 (0.29)	1.34 (0.02)	4.32 (0.53)	39.78 (5.35)	0.81 (0.02)	0.31 (0.03)	2.85 (0.04)
MAXDIST+FEAT(RP)	1.56 (0.06)	3.22 (0.22)	54.05 (11.97)	0.66 (0.03)	0.11 (0.02)	4.56 (0.20)	1.34 (0.02)	4.39 (0.57)	40.94 (7.17)	0.83 (0.02)	0.31 (0.03)	2.85 (0.05)
LCMD+FEAT(LL)	1.51 (0.08)	3.12 (0.12)	54.27 (8.68)	0.58 (0.03)	0.17 (0.02)	4.23 (0.21)	1.30 (0.03)	4.21 (0.47)	40.92 (5.34)	0.79 (0.02)	0.32 (0.01)	2.87 (0.03)
LCMD+FEAT(RP)	1.59 (0.12)	3.28 (0.41)	60.18 (17.22)	0.58 (0.07)	0.15 (0.03)	4.32 (0.33)	1.33 (0.02)	4.35 (0.60)	41.57 (8.13)	0.81 (0.02)	0.33 (0.03)	2.96 (0.02)

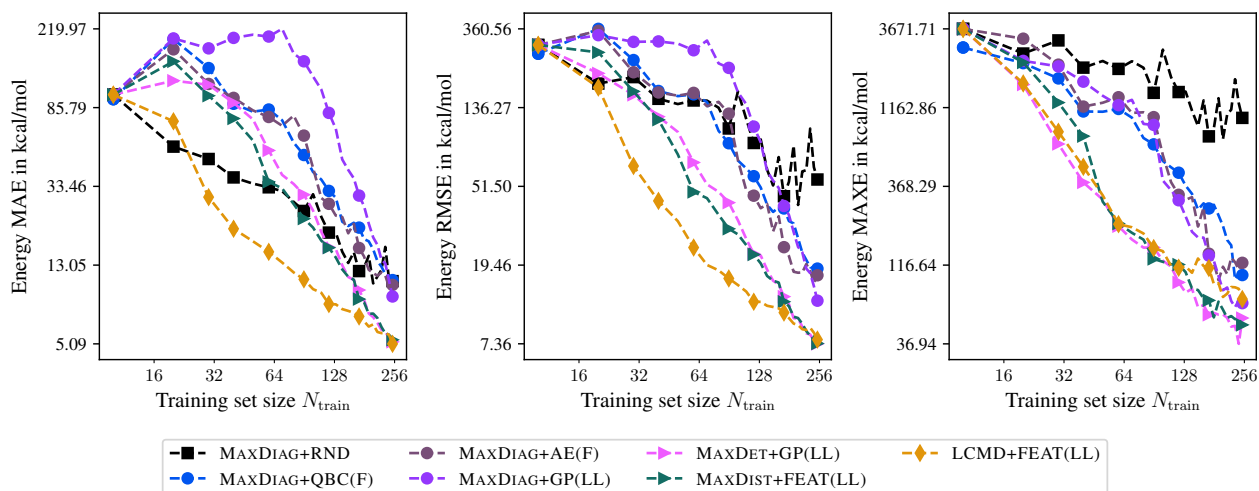


Figure 10 Learning curves for the TiO_2 data set.^{8,9} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(10, 250; 10)_{\text{TiO}_2}$ BMDAL experiment. The markers show only a subset of all BMDAL steps for better visibility, while the lines use all steps. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

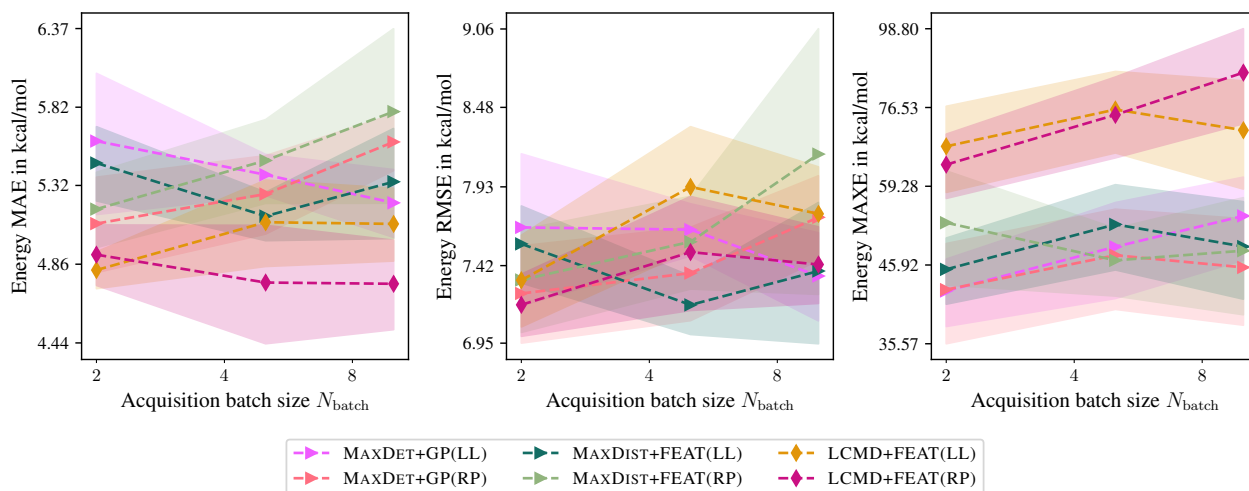


Figure 11 Comparison of the last layer and random projections gradient feature maps on the TiO_2 data set.^{8,9} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(10, 250; \{2, 5, 10\})_{\text{TiO}_2}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

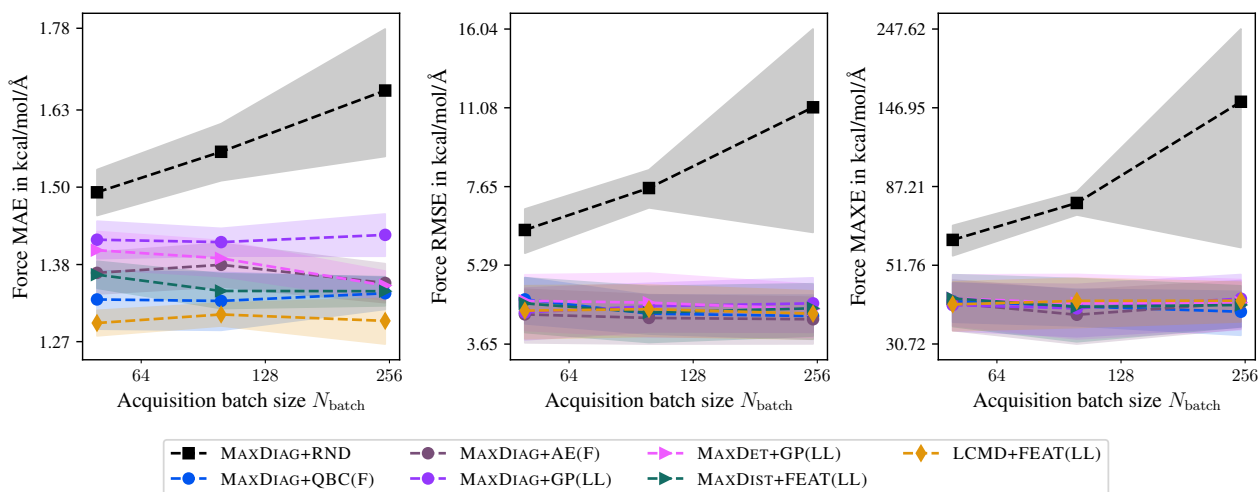


Figure 12 Dependence of the mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces on the acquired batch size N_{batch} . All errors are evaluated for the last BMDAL step on the TiO_2 data set.^{8,9} All corresponding values are given for the $(500, 2500; \{50, 100, 250\})_{\text{TiO}_2}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

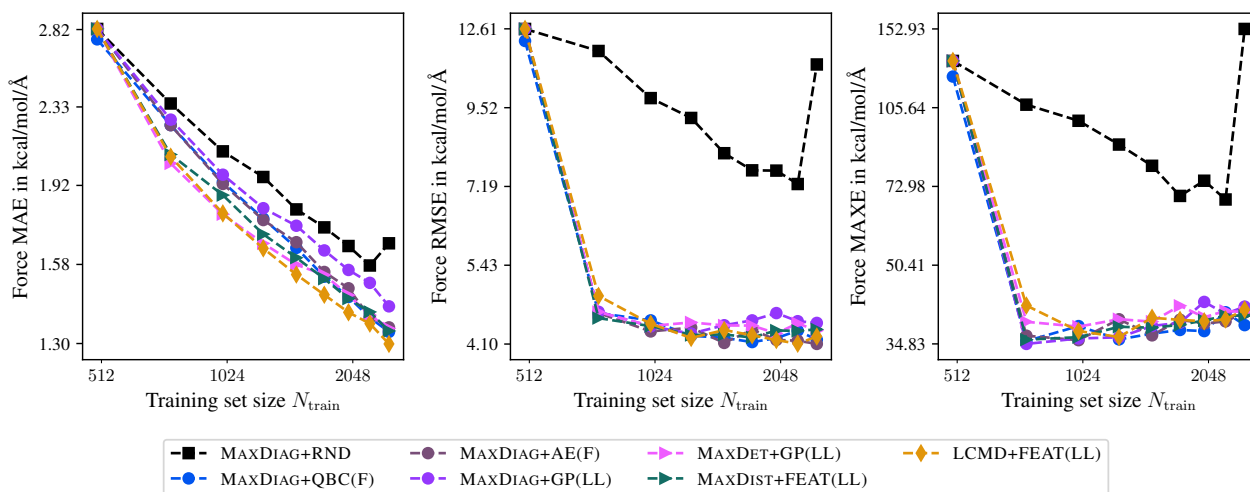


Figure 13 Learning curves for the TiO_2 data set.^{8,9} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(500, 2500; 250)_{\text{TiO}_2}$ BMDAL experiment. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

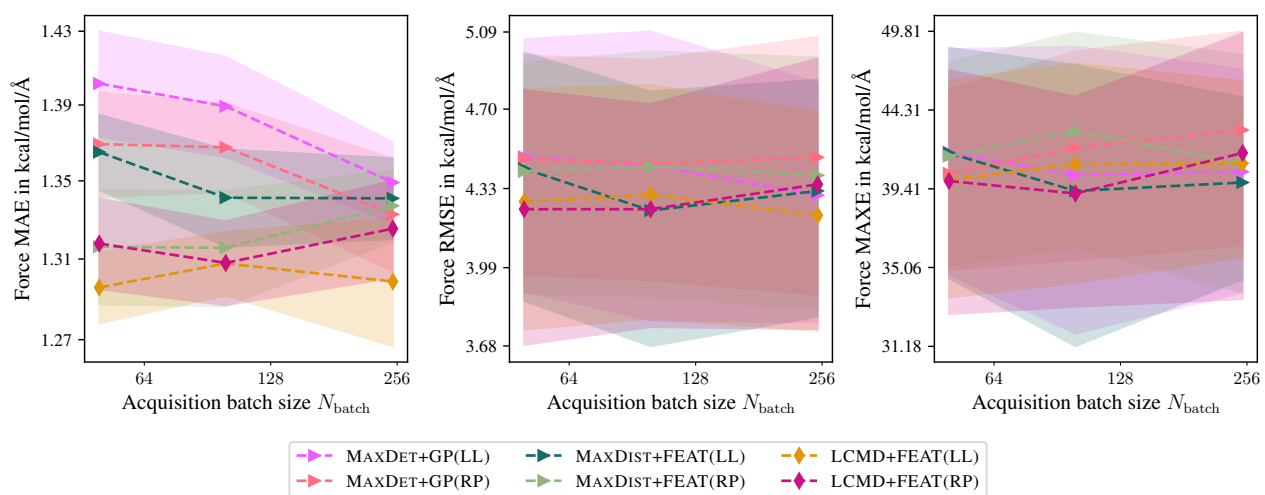


Figure 14 Comparison of the last layer and random projections gradient feature maps on the TiO_2 data set.^{8,9} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(500, 2500; \{50, 100, 250\})_{\text{TiO}_2}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

4 LMNTO

Here, additional results on the LMNTO data set^{10,11} are presented. Table 13 and Table 14 present the numerical values for the mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients, as well as 95% quantile, evaluated on test data for total energies and forces. While Table 13 provides with the errors averaged over the learning curve, Table 14 shows the performance of the final model. All results are given for the $(100, 1000; 100)_{\text{LMNTO}}$ BMDAL experiment.

Fig. 15, and Fig. 16 present the the learning curves and batch size dependence for the $(100, 1000; 100)_{\text{LMNTO}}$ or $(100, 1000; \{25, 50, 100\})_{\text{LMNTO}}$ experiments, respectively. Fig. 17, Fig. 18, and Fig. 19 complement the results obtained on atomic forces with the results for the total energy.

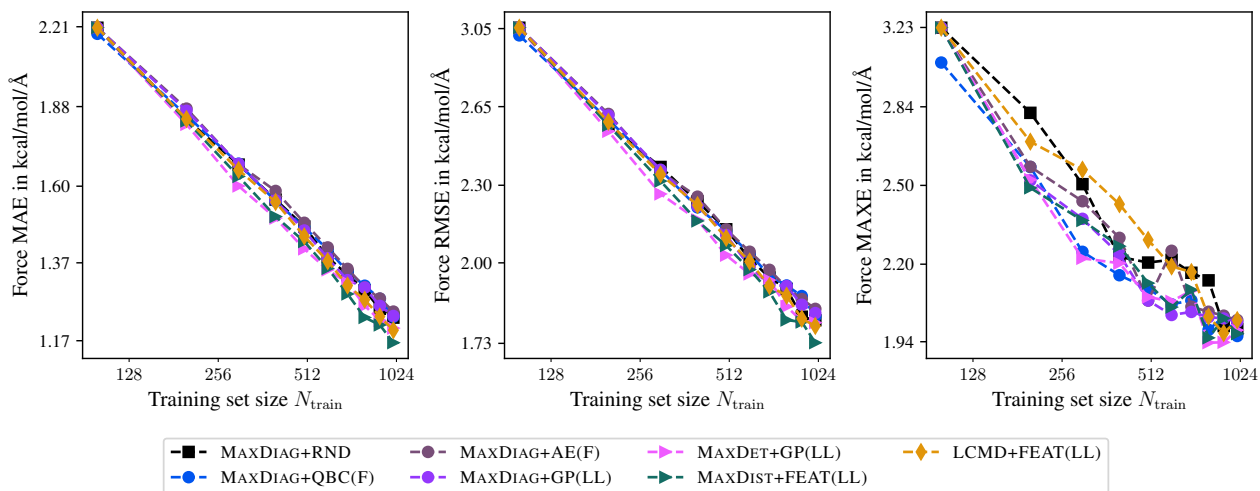


Figure 15 Learning curves for the LMNTO data set.^{10,11} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(100, 1000; 100)_{\text{LMNTO}}$ BMDAL experiment. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

Table 13 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed for (100,1000;100)_{LMNTO} as an average over AL steps. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. All times are given in seconds. Standard errors on the mean are given in parentheses.

Method	Energy					Force					Time			
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%	Kernel	Selection
MAXDIAG+RND	0.90 (0.03)	1.13 (0.03)	3.72 (0.20)	0.01 (0.02)	0.01 (0.02)	2.22 (0.08)	1.52 (0.03)	2.18 (0.04)	2.35 (0.15)	-0.02 (0.03)	-0.01 (0.03)	1.90 (0.05)	0.01 (0.00)	0.00 (0.00)
MAXDIAG+AE(E)	0.88 (0.03)	1.10 (0.04)	3.69 (0.32)	1.00 (0.00)	1.00 (0.00)	2.13 (0.08)	1.56 (0.04)	2.24 (0.05)	2.39 (0.18)	0.06 (0.03)	0.05 (0.03)	1.94 (0.06)	19.32 (3.11)	0.00 (0.00)
MAXDIAG+AE(F)	0.90 (0.02)	1.14 (0.03)	3.78 (0.30)	0.12 (0.03)	0.11 (0.03)	2.20 (0.06)	1.54 (0.03)	2.20 (0.04)	2.30 (0.16)	1.00 (0.00)	1.00 (0.00)	1.88 (0.05)	16.64 (1.86)	0.00 (0.00)
MAXDIAG+QBC(E)	0.89 (0.02)	1.12 (0.03)	3.49 (0.35)	0.29 (0.03)	0.34 (0.02)	1.89 (0.08)	1.54 (0.02)	2.21 (0.02)	2.26 (0.13)	0.19 (0.03)	0.17 (0.03)	1.87 (0.03)	33.74 (0.71)	0.01 (0.01)
MAXDIAG+QBC(F)	0.89 (0.02)	1.13 (0.02)	3.31 (0.21)	0.13 (0.04)	0.13 (0.03)	1.88 (0.06)	1.53 (0.02)	2.19 (0.02)	2.22 (0.16)	0.84 (0.01)	0.77 (0.02)	1.84 (0.03)	33.65 (0.74)	0.01 (0.00)
MAXDIAG+GP(LL)	0.90 (0.02)	1.13 (0.03)	3.73 (0.27)	0.13 (0.04)	0.08 (0.02)	2.24 (0.06)	1.53 (0.03)	2.19 (0.04)	2.25 (0.13)	0.37 (0.04)	0.44 (0.03)	1.87 (0.04)	10.06 (0.34)	0.00 (0.00)
MAXDIAG+GP(RP)	0.91 (0.02)	1.15 (0.03)	3.82 (0.28)	0.13 (0.04)	0.08 (0.02)	2.22 (0.06)	1.53 (0.03)	2.20 (0.04)	2.28 (0.14)	0.60 (0.02)	0.59 (0.02)	1.89 (0.04)	186.09 (173.20)	0.00 (0.00)
MAXDET+GP(LL)	0.89 (0.02)	1.12 (0.03)	3.71 (0.22)	0.14 (0.05)	0.08 (0.03)	2.18 (0.07)	1.49 (0.03)	2.14 (0.04)	2.23 (0.13)	0.36 (0.04)	0.44 (0.03)	1.84 (0.04)	12.57 (2.62)	0.51 (0.07)
MAXDET+GP(RP)	0.87 (0.02)	1.10 (0.03)	3.67 (0.22)	0.11 (0.05)	0.06 (0.03)	2.18 (0.06)	1.48 (0.03)	2.12 (0.04)	2.20 (0.13)	0.60 (0.03)	0.60 (0.02)	1.82 (0.04)	13.55 (1.03)	0.47 (0.02)
MAXDIST+GP(LL)	0.86 (0.03)	1.09 (0.03)	3.64 (0.28)	0.07 (0.03)	0.05 (0.02)	2.12 (0.07)	1.48 (0.03)	2.14 (0.04)	2.24 (0.13)	0.66 (0.03)	0.55 (0.03)	1.84 (0.04)	11.67 (0.83)	1.16 (0.14)
MAXDIST+GP(RP)	0.87 (0.02)	1.10 (0.03)	3.69 (0.22)	0.05 (0.02)	0.05 (0.02)	2.16 (0.06)	1.48 (0.03)	2.13 (0.04)	2.23 (0.14)	0.67 (0.02)	0.56 (0.02)	1.83 (0.04)	13.14 (1.13)	1.15 (0.15)
MAXDIST+FEAT(LL)	0.87 (0.02)	1.10 (0.03)	3.74 (0.30)	0.06 (0.04)	0.02 (0.02)	2.14 (0.08)	1.48 (0.03)	2.14 (0.04)	2.26 (0.16)	0.61 (0.03)	0.49 (0.03)	1.85 (0.05)	11.76 (1.27)	1.07 (0.12)
MAXDIST+FEAT(RP)	0.87 (0.02)	1.10 (0.03)	3.64 (0.26)	0.05 (0.03)	0.04 (0.02)	2.15 (0.06)	1.48 (0.02)	2.13 (0.03)	2.30 (0.15)	0.62 (0.03)	0.47 (0.03)	1.84 (0.04)	13.45 (0.69)	1.17 (0.12)
LCMID+FEAT(LL)	0.88 (0.02)	1.11 (0.03)	3.83 (0.34)	0.12 (0.04)	0.08 (0.02)	2.17 (0.06)	1.51 (0.03)	2.17 (0.04)	2.36 (0.18)	0.65 (0.03)	0.56 (0.02)	1.88 (0.05)	10.85 (0.26)	1.46 (0.05)
LCMID+FEAT(RP)	0.88 (0.02)	1.12 (0.03)	4.10 (0.29)	0.12 (0.03)	0.10 (0.03)	2.18 (0.06)	1.50 (0.03)	2.16 (0.03)	2.35 (0.15)	0.67 (0.02)	0.56 (0.02)	1.88 (0.04)	13.19 (0.25)	1.46 (0.05)

Table 14 The mean absolute error (MAE), root-mean-square error (RMSE), maximum error (MAXE), Pearson (PCC) and Spearman (SRC) correlation coefficients for total energies and atomic forces evaluated on test data. All values are computed employing the final model obtained for the (100, 1000; 100)_{LMNTO} experiment. The energy errors are given in kcal/mol, while the force errors are in kcal/mol/Å. Standard errors on the mean are given in parentheses.

Method	Energy					Force						
	MAE	RMSE	MAXE	PCC	SRC	95%	MAE	RMSE	MAXE	PCC	SRC	95%
MAXDIAG+RND	0.72 (0.02)	0.90 (0.02)	2.96 (0.16)	0.03 (0.02)	0.02 (0.01)	1.79 (0.04)	1.23 (0.02)	1.80 (0.02)	1.98 (0.14)	-0.02 (0.03)	-0.01 (0.03)	1.57 (0.03)
MAXDIAG+AE(E)	0.64 (0.02)	0.82 (0.03)	3.58 (0.73)	1.00 (0.00)	1.00 (0.00)	1.58 (0.04)	1.25 (0.03)	1.85 (0.04)	2.07 (0.20)	0.05 (0.03)	0.03 (0.02)	1.62 (0.05)
MAXDIAG+AE(F)	0.68 (0.02)	0.86 (0.02)	2.96 (0.22)	0.11 (0.05)	0.12 (0.03)	1.69 (0.02)	1.24 (0.02)	1.84 (0.03)	2.01 (0.21)	1.00 (0.00)	1.00 (0.00)	1.59 (0.03)
MAXDIAG+QBC(E)	0.67 (0.02)	0.85 (0.03)	2.72 (0.43)	0.27 (0.01)	0.33 (0.01)	1.39 (0.06)	1.25 (0.01)	1.85 (0.02)	1.96 (0.16)	0.18 (0.02)	0.16 (0.03)	1.58 (0.02)
MAXDIAG+QBC(F)	0.69 (0.01)	0.87 (0.01)	2.57 (0.10)	0.09 (0.02)	0.11 (0.03)	1.42 (0.04)	1.23 (0.01)	1.82 (0.02)	1.96 (0.20)	0.90 (0.01)	0.81 (0.02)	1.54 (0.02)
MAXDIAG+GP(LL)	0.69 (0.02)	0.87 (0.02)	2.98 (0.26)	0.19 (0.06)	0.09 (0.02)	1.71 (0.06)	1.23 (0.03)	1.83 (0.05)	2.00 (0.21)	0.38 (0.04)	0.46 (0.04)	1.58 (0.06)
MAXDIAG+GP(RP)	0.68 (0.02)	0.84 (0.02)	2.90 (0.18)	0.11 (0.03)	0.08 (0.02)	1.63 (0.05)	1.21 (0.02)	1.80 (0.03)	1.89 (0.16)	0.63 (0.02)	0.67 (0.01)	1.56 (0.03)
MAXDEF+GP(LL)	0.67 (0.01)	0.86 (0.02)	3.07 (0.37)	0.17 (0.08)	0.08 (0.02)	1.69 (0.05)	1.20 (0.03)	1.80 (0.04)	1.98 (0.17)	0.37 (0.03)	0.49 (0.03)	1.56 (0.05)
MAXDEF+GP(RP)	0.67 (0.01)	0.85 (0.01)	2.90 (0.17)	0.13 (0.04)	0.07 (0.03)	1.75 (0.05)	1.17 (0.02)	1.75 (0.03)	1.88 (0.12)	0.63 (0.02)	0.68 (0.01)	1.52 (0.03)
MAXDIST+GP(LL)	0.63 (0.02)	0.81 (0.03)	2.95 (0.25)	0.11 (0.04)	0.06 (0.01)	1.53 (0.06)	1.17 (0.02)	1.77 (0.03)	1.96 (0.16)	0.81 (0.01)	0.72 (0.02)	1.55 (0.03)
MAXDIST+GP(RP)	0.65 (0.02)	0.82 (0.02)	2.64 (0.13)	0.07 (0.02)	0.07 (0.01)	1.60 (0.05)	1.16 (0.02)	1.74 (0.02)	1.87 (0.10)	0.84 (0.01)	0.72 (0.01)	1.54 (0.02)
MAXDIST+FEAT(LL)	0.65 (0.02)	0.84 (0.03)	3.07 (0.47)	0.13 (0.07)	0.06 (0.03)	1.59 (0.04)	1.17 (0.03)	1.73 (0.04)	1.97 (0.24)	0.75 (0.02)	0.64 (0.02)	1.52 (0.03)
MAXDIST+FEAT(RP)	0.64 (0.01)	0.80 (0.02)	2.80 (0.19)	0.08 (0.04)	0.07 (0.03)	1.59 (0.04)	1.17 (0.02)	1.77 (0.03)	2.13 (0.26)	0.77 (0.01)	0.64 (0.02)	1.56 (0.04)
LCMD+FEAT(LL)	0.65 (0.01)	0.84 (0.02)	2.94 (0.32)	0.18 (0.04)	0.11 (0.01)	1.68 (0.05)	1.20 (0.03)	1.79 (0.04)	2.01 (0.18)	0.76 (0.03)	0.68 (0.02)	1.57 (0.04)
LCMD+FEAT(RP)	0.69 (0.02)	0.89 (0.03)	3.27 (0.18)	0.09 (0.02)	0.06 (0.01)	1.80 (0.07)	1.18 (0.02)	1.76 (0.03)	1.94 (0.15)	0.78 (0.01)	0.68 (0.01)	1.55 (0.04)

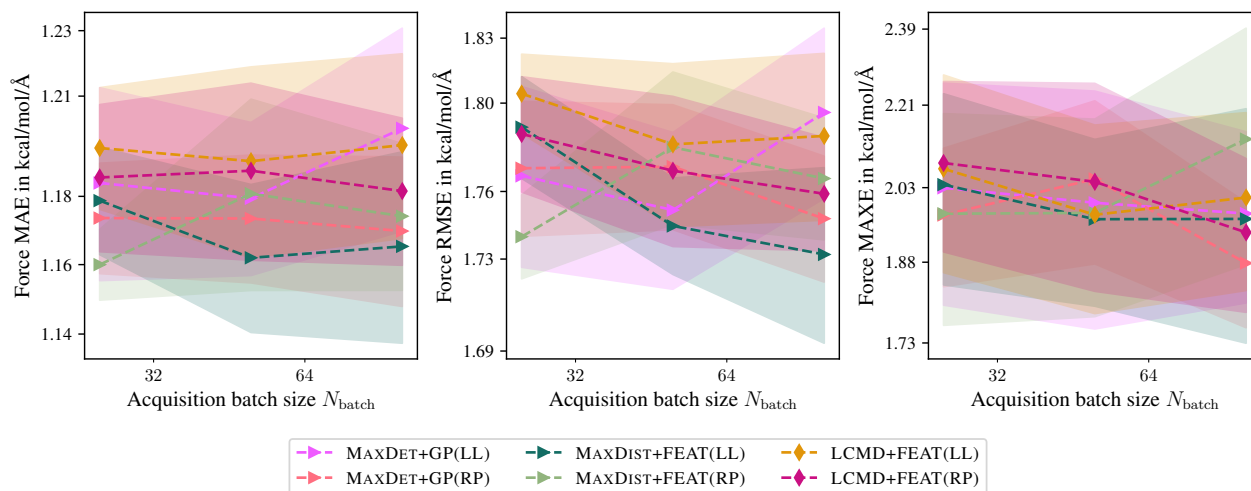


Figure 16 Comparison of the last layer and random projections gradient feature maps on the LMNTO data set.^{10,11} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of atomic forces are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(100, 1000; \{25, 50, 100\})_{\text{LMNTO}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

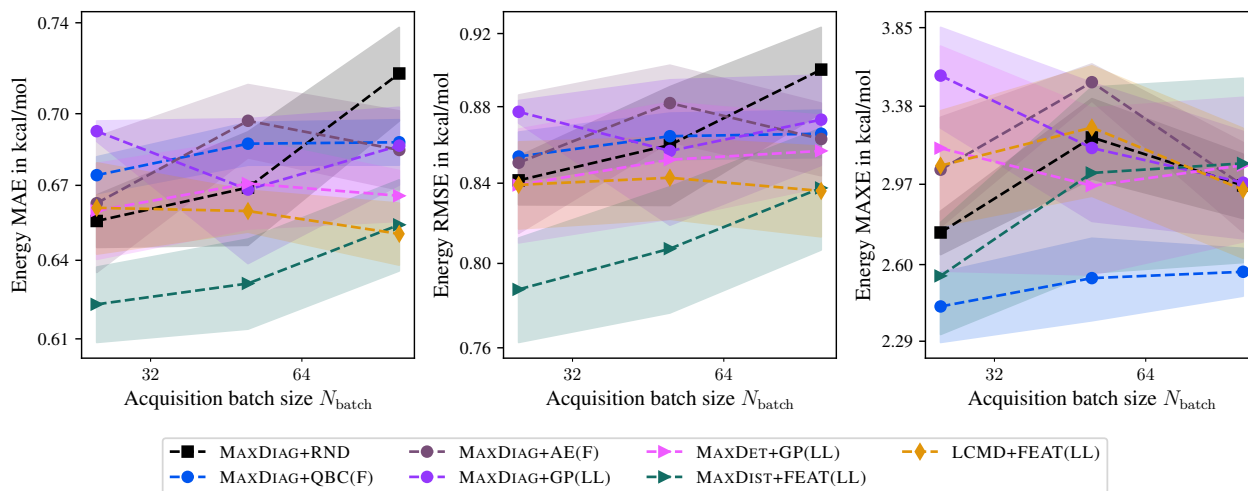


Figure 17 Dependence of the mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy on the acquired batch size N_{batch} . All errors are evaluated for the last BMDAL step on the LMNTO data set.^{10,11} All corresponding values are given for the $(100, 1000; \{25, 50, 100\})_{\text{LMNTO}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

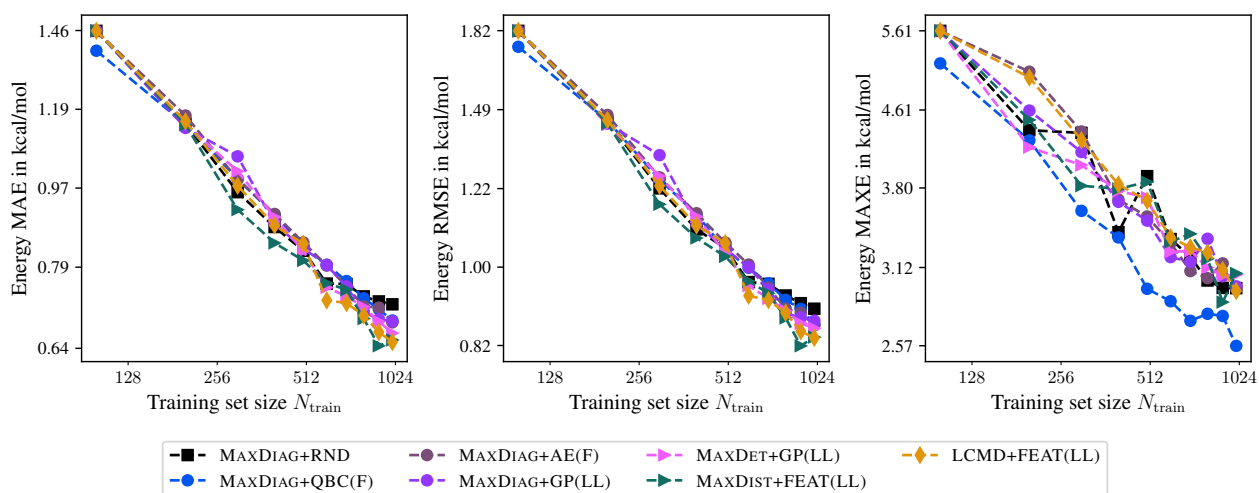


Figure 18 Learning curves for the LMNTO data set.^{10,11} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the training set size acquired during BMDAL. All corresponding values are given for the $(100,1000;100)_{\text{LMNTO}}$ BMDAL experiment. The training errors before the first BMDAL step are identical for most methods since they use the same random seed. This does not apply to QbC, where more models are trained.

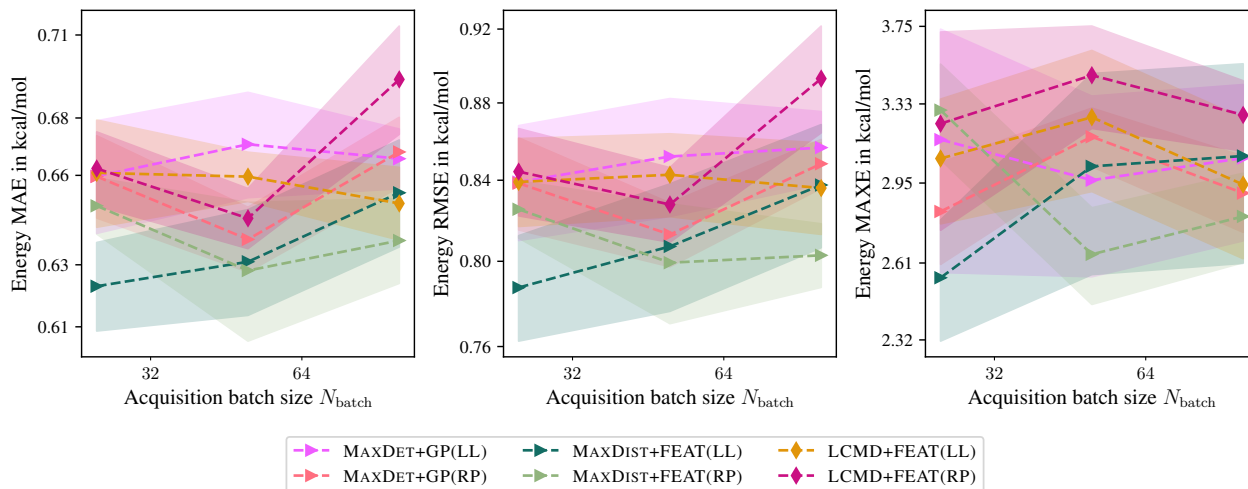


Figure 19 Comparison of the last layer and random projections gradient feature maps on the LMNTO data set.^{10,11} The mean absolute errors (MAE), root-mean-square errors (RMSE), and maximum errors (MAXE) of the total energy are plotted against the acquired batch size N_{batch} . All corresponding values are given for the $(100,1000;\{25,50,100\})_{\text{LMNTO}}$ BMDAL experiments. Shaded areas denote the standard error on the mean evaluated over five independent runs.

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