Supplementary Material

Using the Gini coefficient to characterize the shape of computational chemistry error distributions

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Case	Property (units)	N	K	Source	Figure
BOR2019	Band gaps (eV)	471	15	[1]	1
NAR2019	Enthalpies of formation (kcal/mol)	469	4	[2]	2
PER2018	Intensive atomization energies (kcal/mol)	222	9	[3]	3
SCH2018	Chemisorption energies (eV)	195	7	[4]	4
THA2015	Polarizability (relative errors, in %)	135	7	[5]	5
WU2015	Polarizability (relative errors, in %)	145	36	[6]	6, 7
ZAS2019	Effective atomization energies (kcal/mol)	6211	3	[7]	8
ZHA2018	Solid formation enthalpies (kcal/mol)	196	2	[8]	9

Table 1: Case studies: N is the number of systems in the dataset and K is the number of methods.

1 Statistics

Statistics for the datasets listed in Table 1 are provided in the companion file "allStats.csv" for four combinations of the correction level (ctd = -1/1) and outliers treatment (out = FALSE/TRUE). The headers in the table are defined as follows (prefix 'u_' refers to uncertainty, obtained by bootstrapping with 1000 samples):

Dataset name of the dataset

Methods name of the method

ctd -1 = raw data; 1 = linear correction

out FALSE = raw data; TRUE = removal of global outliers

mue mean unsigned error

mse mean signed error

rmsd root mean squared deviation

q95hd 95th quantile of the absolute errors Q_{95} (Harrell-Davis estimator [9])

hrmode mode of the errors distribution (HRM estimator [10])

skew skewness (moments-based formula)

skewgm robust skewness (Groeneveld and Meeden estimator [11])

skewgm_mcf robust skewness of mode-centered errors

kurt kurtosis (moments-based formula)

kurtcs robust kurtosis excess [12]

gini Gini coefficient of the absolute errors G_F

gimc Gini coefficient of the mode-centered error distribution

bmax value of the error bias maximizing the Gini coefficient

gmax maximal value of the Gini coefficient (data centered on bmax)

2 ECDF and Lorenz curves

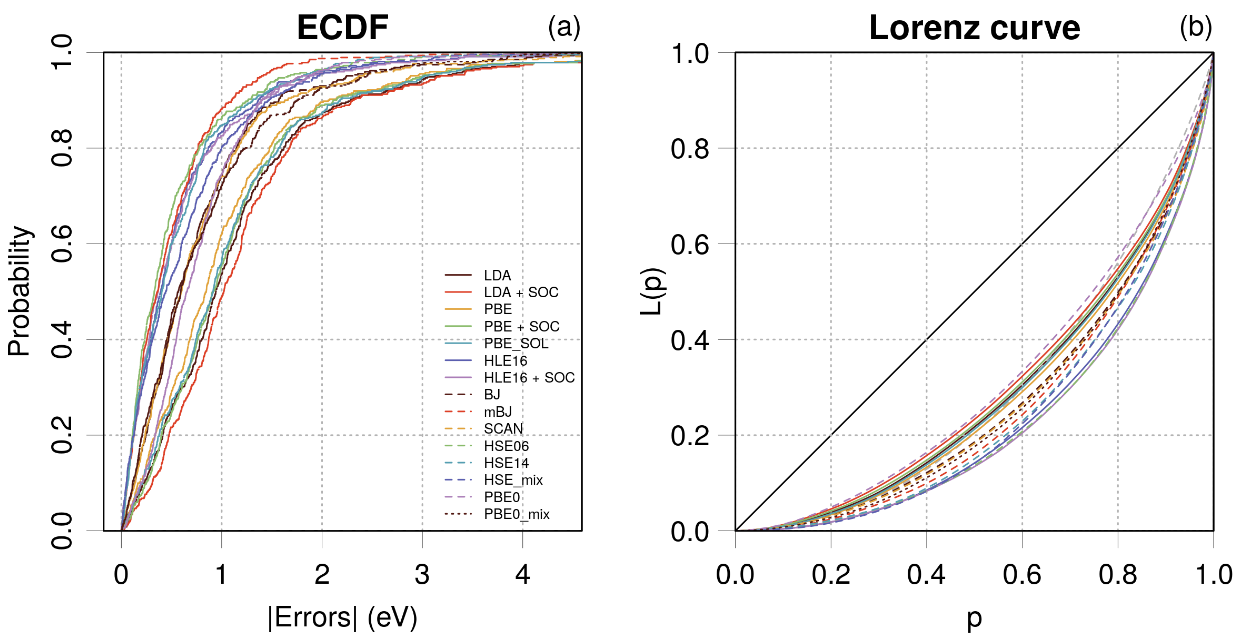

Figure 1: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from BOR2019.

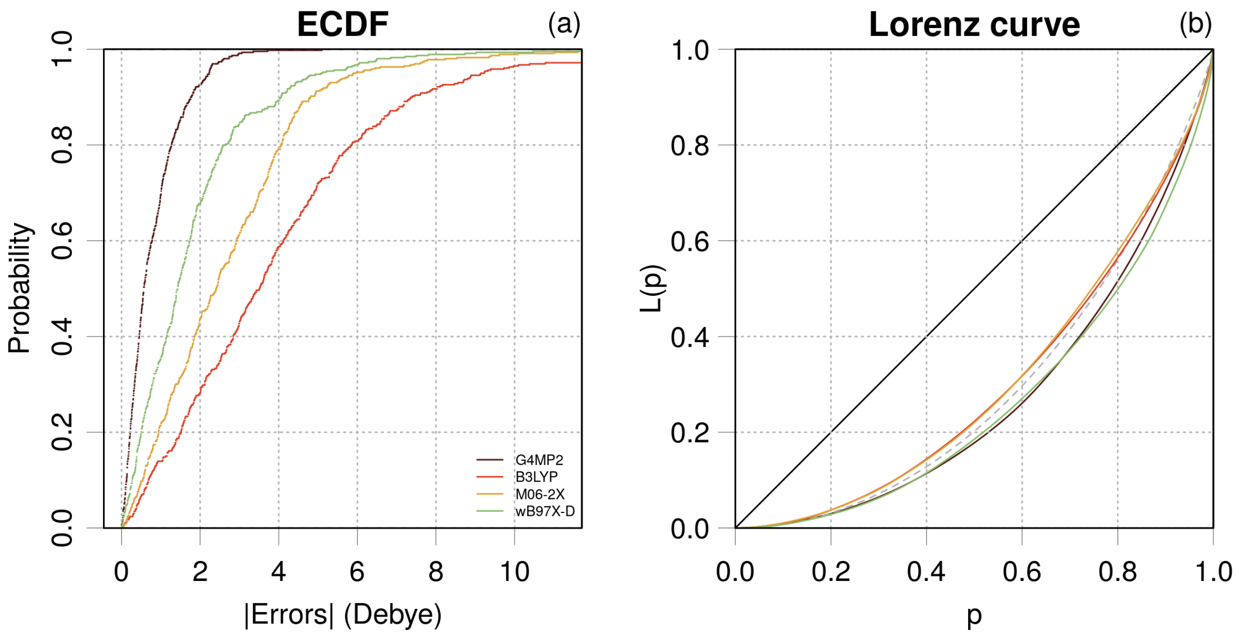

Figure 2: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from NAR2019.

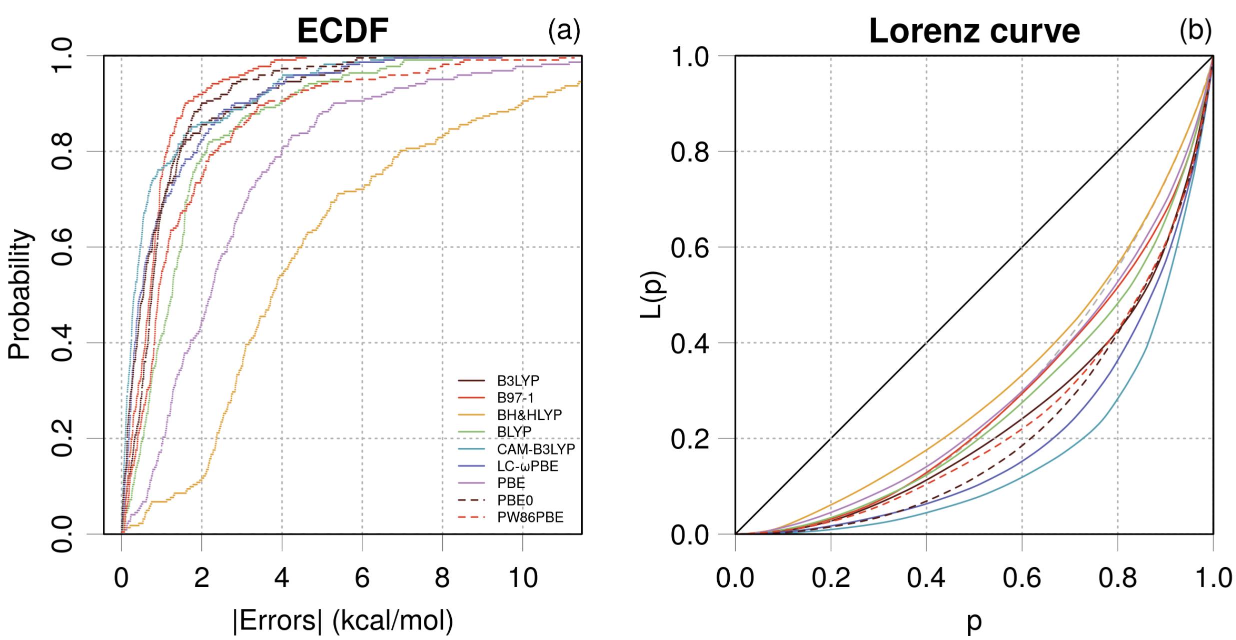

Figure 3: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from PER2018.

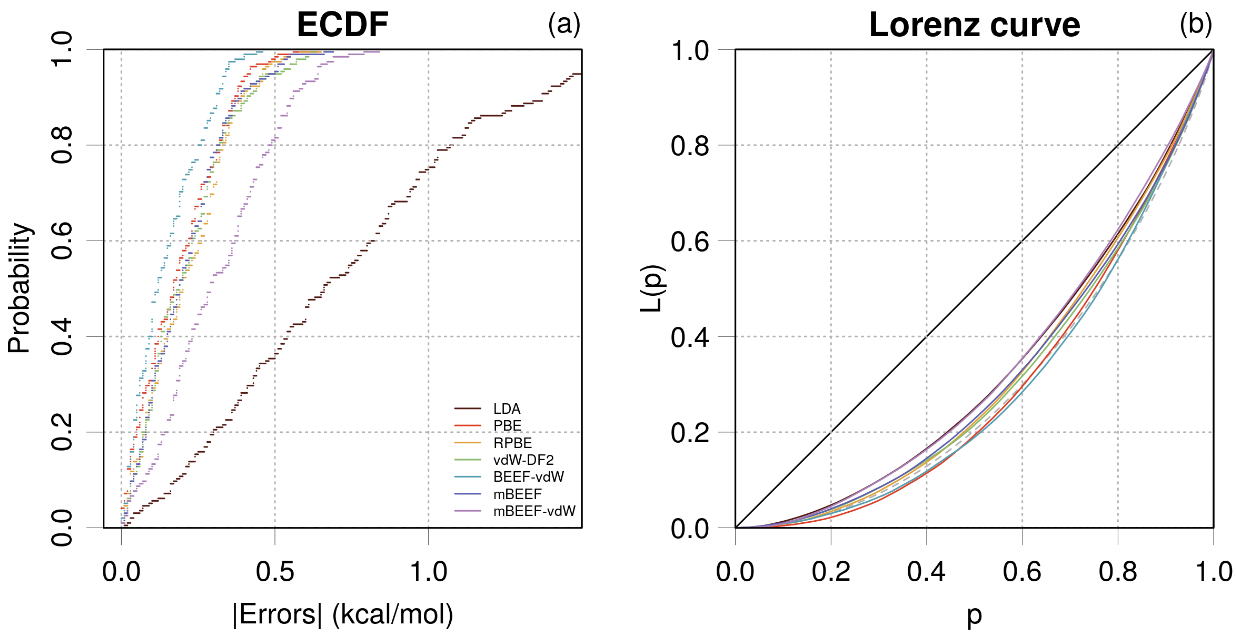

Figure 4: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from SCH2018.

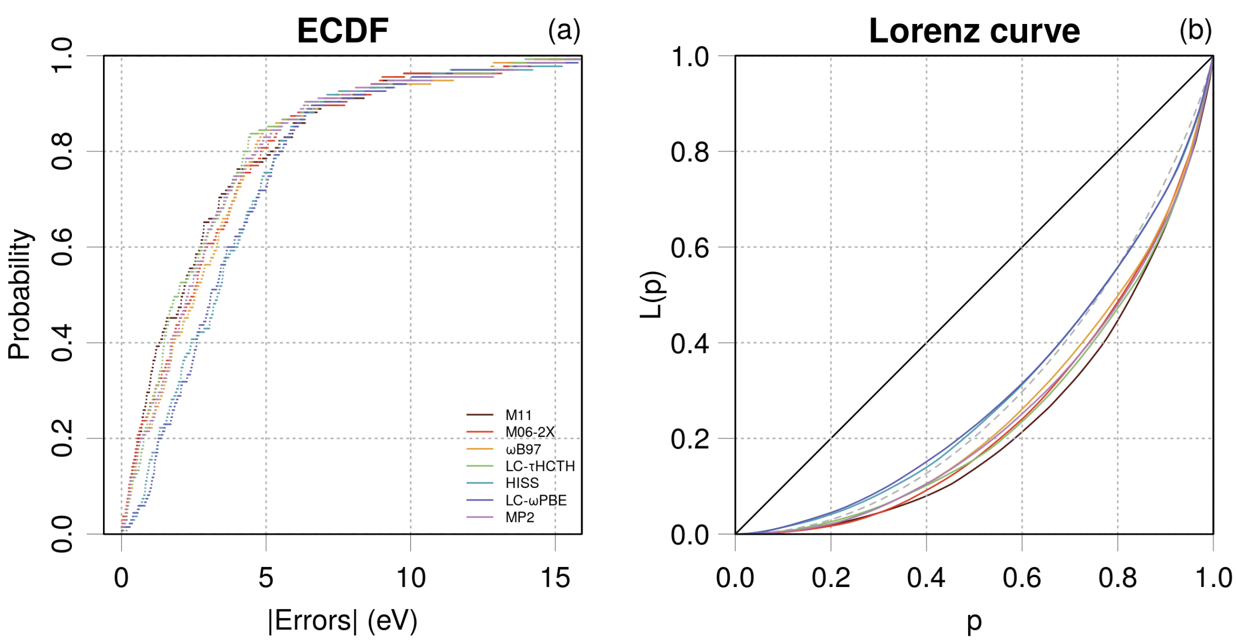

Figure 5: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from THA2015.

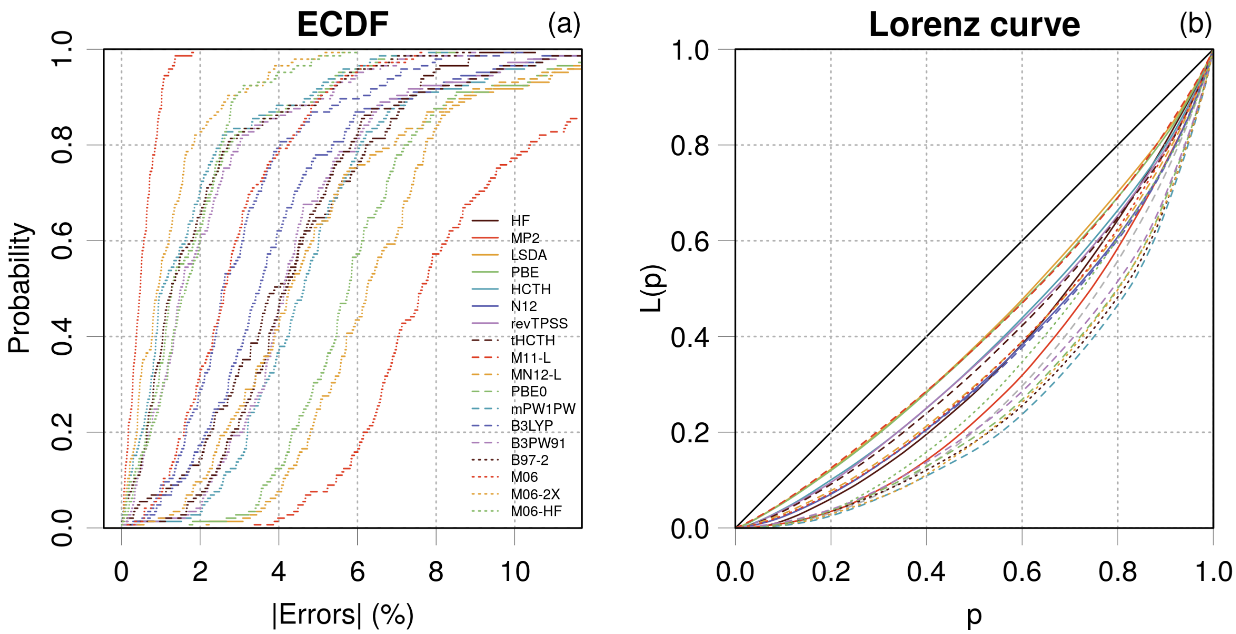

Figure 6: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from WU2015 (1/2).

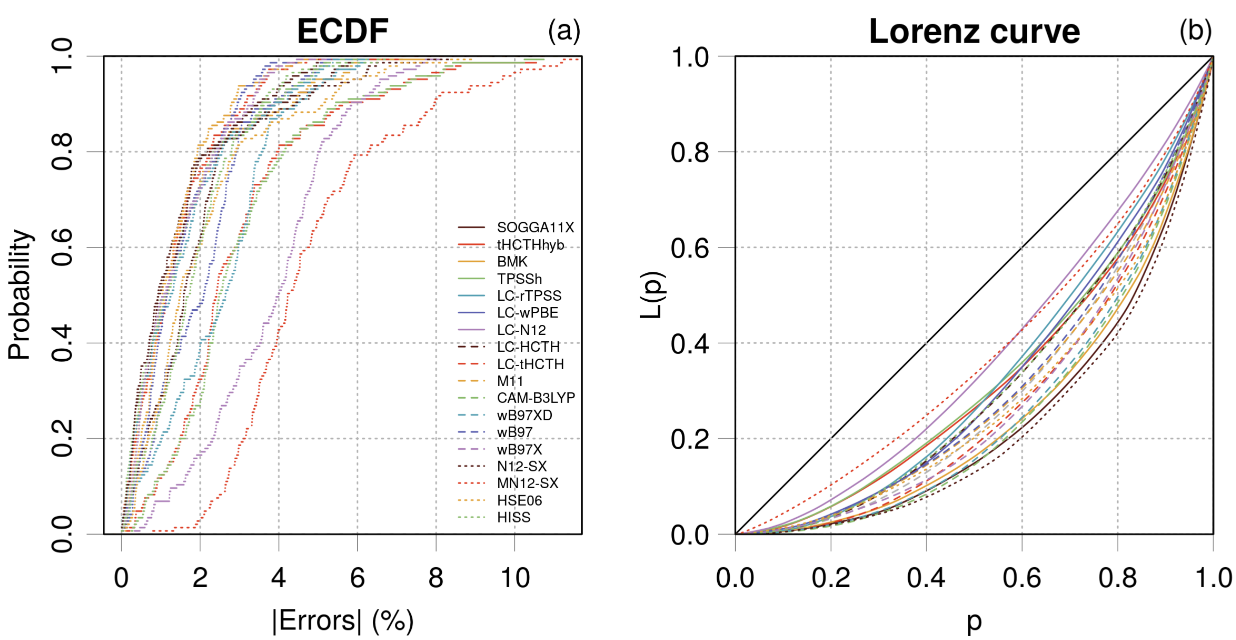

Figure 7: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from WU2015 (2/2).

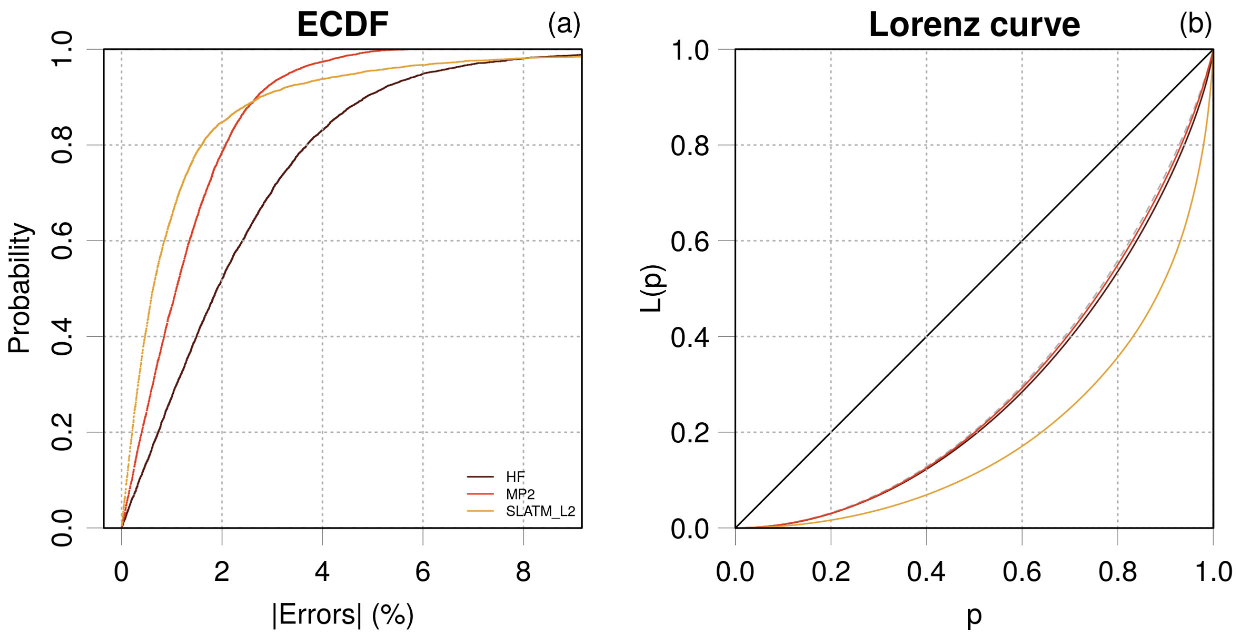

Figure 8: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from ZAS2019.

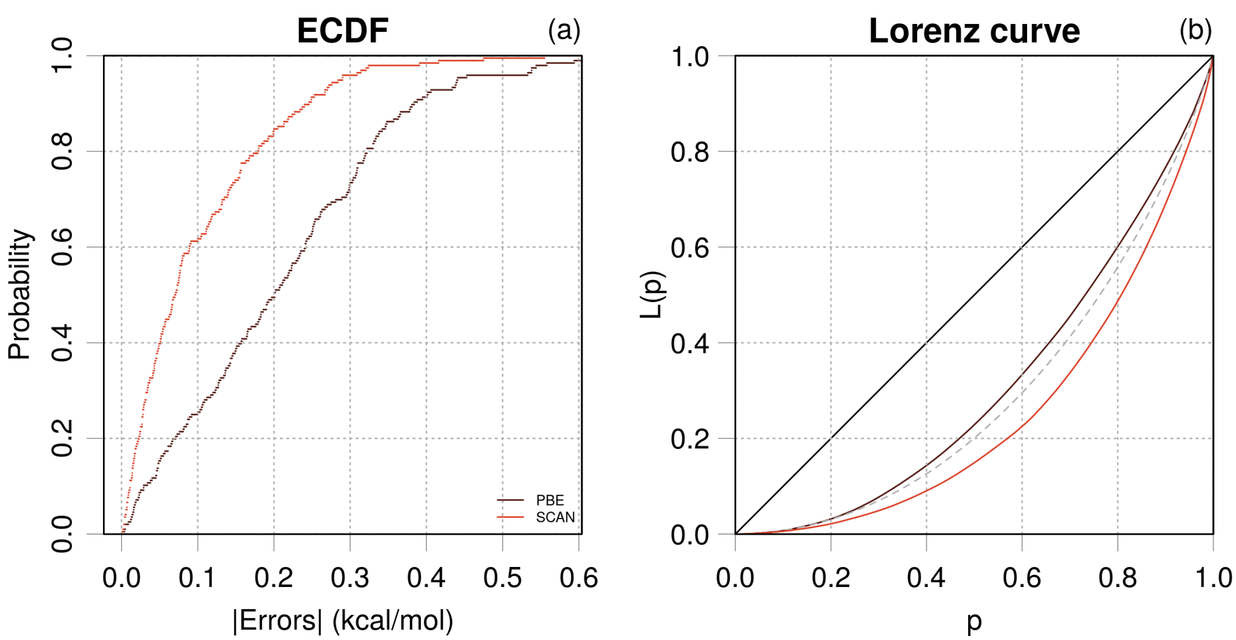

Figure 9: Empirical cumulative distribution functions (ECDF) and Lorenz curves for absolute error sets from ZHA2018.

References

[1] P. Borlido, T. Aull, A. W. Huran, F. Tran, M. A. Marques, and S. Botti. Large-scale benchmark of exchange–correlation functionals for the determination of electronic band gaps of

solids. *J. Chem. Theory Comput.*, 15:5069–5079, 2019. doi:10.1021/acs.jctc.9b00322.

[2] B. Narayanan, P. C. Redfern, R. S. Assary, and L. A. Curtiss. Accurate quantum chemical energies for 133000 organic molecules. *Chem. Sci.*, 10:7449–7455, 2019. doi:10.1039/c9sc02834j.

[3] P. Pernot and A. Savin. Probabilistic performance estimators for computational chemistry methods: the empirical cumulative distribution function of absolute errors. *J. Chem. Phys.*, 148:241707, 2018. doi:10.1063/1.5016248.

[4] P. S. Schmidt and K. S. Thygesen. Benchmark database of transition metal surface and adsorption energies from many-body perturbation theory. *J. Phys. Chem. C*, 122:4381–4390, 2018. doi:10.1021/acs.jpcc.7b12258.

[5] A. J. Thakkar and T. Wu. How well do static electronic dipole polarizabilities from gas-phase experiments compare with density functional and MP2 computations? *J. Chem. Phys.*, 143:144302, 2015. doi:10.1063/1.4932594.

[6] T. Wu, Y. N. Kalugina, and A. J. Thakkar. Choosing a density functional for static molecular polarizabilities. *Chem. Phys. Lett.*, 635:257–261, 2015. doi:10.1016/j.cplett.2015.07.003.

[7] P. Zaspel, B. Huang, H. Harbrecht, and O. A. von Lilienfeld. Boosting quantum machine learning models with a multilevel combination technique: Pople diagrams revisited. *J. Chem. Theory Comput.*, 15(3):1546–1559, 2019. doi:10.1021/acs.jctc.8b00832.

[8] Y. Zhang, D. A. Kitchaev, J. Yang, T. Chen, S. T. Dacek, R. A. Sarmiento-Perez, M. A. L. Marques, H. Peng, G. Ceder, J. P. Perdew, and J. Sun. Efficient first-principles prediction of solid stability: Towards chemical accuracy. *npj Comput. Mater.*, 4:9, 2018. doi:10.1038/s41524-018-0065-z.

[9] F. E. Harrell and C. Davis. A new distribution-free quantile estimator. *Biometrika*, 69:635–640, 1982. doi:10.2307/2335999.

[10] D. R. Bickel. Robust estimators of the mode and skewness of continuous data. *Comput. Stat. Data Anal.*, 39:153–163, 2002. URL: http://www.sciencedirect.com/science/article/pii/S0167947301000573, doi:10.1016/S0167-9473(01)00057-3.

[11] R. A. Groeneveld and G. Meeden. Measuring skewness and kurtosis. *The Statistician*, 33:391–399, 1984. URL: http://www.jstor.org/stable/2987742, doi:10.2307/2987742.

[12] M. Bonato. Robust estimation of skewness and kurtosis in distributions with infinite higher moments. *Finance Research Letters*, 8:77–87, 2011. doi:10.1016/j.frl.2010.12.001.