Supporting Information for "ParAMS: Parameter Optimization for Atomistic and Molecular Simulations"

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Table S1: Composition of the reference data published by MH,[48] split by the computational tasks Single Point (SP) and Geometry Optimization (GO). For each of the two sets, the upper part describes the chemical systems, while the lower breaks down the individual entries in the training and validation sets. Note that some entries might be a function of multiple chemical systems, meaning that the sum of SP+GO is not necessarily equal to the total number of entries for that row (*cf.* Sec. 3.2).

Training Set	**SP**	**GO**	**Total**
Number of systems	222	9	231
Mean system size (atoms)	6.6	11.4	6.8
Std. dev. (atoms)	2.9	7.7	3.3
Total number of entries	4620	317	4875
Energies	219	62	219
Forces	4401	0	4401
Atomic distances	0	94	94
Angles	0	85	85
Dihedrals	0	76	76
Validation Set			
Number of systems	200	24	224
Mean system size (atoms)	24.0	12.7	22.8
Std. dev. (atoms)	0.0	5.9	4.0
Total number of entries	199	771	970
Energies	199	0	199
Forces			0
Atomic distances	0	281	281
Angles	0	257	257
Dihedrals	0	233	233

Table S2: Summary of relevant ParAMS settings used for the re-parameterization of Mue2016.

Setting	**Value**
Number of optimizations	9
Number of parameters to optimize	35
Lower / upper parameter bounds	$\boldsymbol{x}_0 \pm 0.2\lvert\boldsymbol{x}_0\rvert$
Optimization timeout	24 hours
CMA-ES population size	36
CMA-ES sigma	0.3
Loss function	sum of squared errors
Early stopping patience	6000 evaluations
Constraints	$r_0^\sigma \geq r_0^\pi$ and $r_0^\pi \geq r_0^{\pi\pi}$

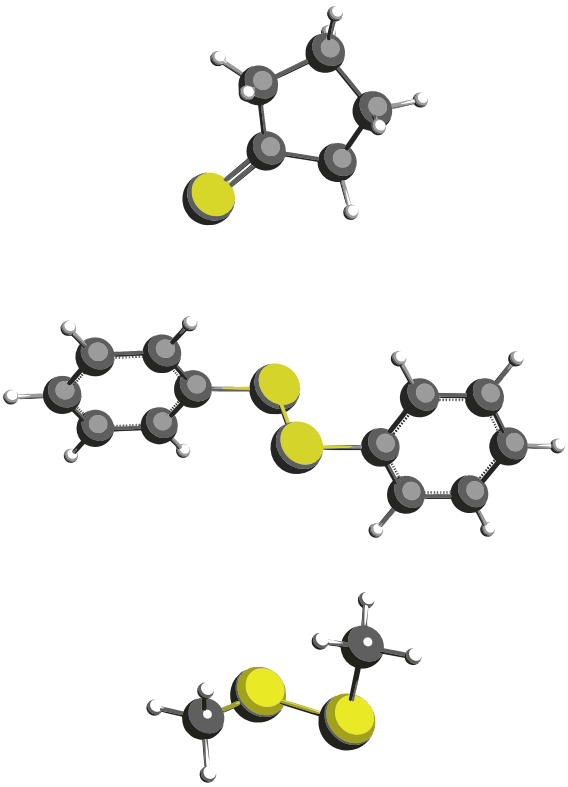

Figure S1: From top to bottom: Example structures of cyclopentathione, diphenyl disulfide and dimethyl disulfide, containing S (yellow), C (black), and H (white), included in the data provided by MH. The fitted properties include bond distances, angles, relative energies and atomic forces.