Uncertainty quantification confirms unreliable extrapolation toward high pressures for united-atom Mie λ-6 force field

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Dear Editor,

We would greatly appreciate if you would consider the following paper for publication in *The Journal of Chemical Physics*. In this work, we demonstrate that the popular state-of-the-art united-atom Mie λ-6 force field is not capable of predicting both vapor-liquid equilibria and high pressure properties. First, we present a case study of the existing united-atom force fields for normal and branched alkanes. Second, we perform Bayesian inference to quantify the uncertainty in the force field parameters to determine whether a suitable parameter set exists to predict each property of interest. The methodological novelty is how we perform this computationally expensive Bayesian analysis by efficiently reweighting sampled configurations. The primary importance of this study is that it provides molecular insight regarding the steepness of the repulsive barrier for nonbonded interactions. As developing theoretically sound force fields (beyond the traditional Lennard-Jones 12-6 potential) is a necessary and growing endeavor in applied chemical physics, we believe this manuscript fits the aims and scope of *The Journal of Chemical Physics.* We are willing to make any changes that may be required to make the manuscript publishable in *The Journal of Chemical Physics*.

Sincerely,

Richard Messerly