Efficient Implementation for Unitary Coupled Cluster State Preparation for Near-Term Quantum Computers

J. Wayne Mullinax, [1,] Norm M. Tubman[2]

1. KBR, Inc. NASA Ames Research Center, Intelligent Systems Division
2. NASA Ames Research Center, Intelligent Systems Division

Introduction

- Quantum simulation of molecular electronic structure represents a potential leap in computational quantum chemistry and a promising application to demonstrate quantum advantage.

- The current state of quantum hardware technology limits applications of chemistry and materials science.

- We aim to address the gap between what can be done on classical and quantum hardware and provide insight into the unitary coupled cluster ansatz for molecules with DZ quality basis sets with up to 64 spin-orbitals.

HPC Resources
- Sparse circuit solver
- Approximate but robust optimization
- Find convergence region to avoid barren plateaus
- Benchmarking

→

Quantum Resources
- Refine and improve ansatz
- Quantum advantage

Outline

- UCC Ansatz
- Computational Details
- Results & Discussion
  - Small molecule benchmark
  - NH_3 analysis
- Summary

UCC Ansatz

$$|\Psi_{\text{UCC}}\rangle = e^{\hat{\sigma}}|\Psi_0\rangle$$

$$\hat{\sigma} = \hat{T} - \hat{T}^\dagger$$

$$\hat{T} = \sum_i^{\text{occ}}\sum_a^{\text{vir}} \theta_i^a \hat{a}_a^\dagger \hat{a}_i + \sum_{ij}^{\text{occ}}\sum_{ab}^{\text{vir}} \theta_{ij}^{ab} \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_j \hat{a}_i + \cdots$$

$$|\Psi_{\text{UCC}}\rangle = \prod_{ij\cdots}^{\text{occ}}\prod_{ab\cdots}^{\text{vir}} \exp\left[\theta_{ij\cdots}^{ab\cdots}\left(\hat{a}_{ij\cdots}^{ab\cdots} - \hat{a}_{ab\cdots}^{ij\cdots}\right)\right]|\Psi_0\rangle$$

$$U_{i_1\cdots i_n}^{a_i\cdots a_n} = \exp\left[\theta_{ij\cdots}^{ab\cdots}\left(\hat{a}_{ij\cdots}^{ab\cdots} - \hat{a}_{ab\cdots}^{ij\cdots}\right)\right]$$
$$= 1 + \sin\theta_{ij\cdots}^{ab\cdots}\left(\hat{a}_{ij\cdots}^{ab\cdots} - \hat{a}_{ab\cdots}^{ij\cdots}\right)$$
$$+ \left(\cos\theta_{ij\cdots}^{ab\cdots} - 1\right) f\left(\{\hat{n}_p\}\right)$$

Chen, J.; Chen, H.-P.; Freericks, J. K. *J. Chem. Theory Comput.* **2021**, *17*, 841-847.

- All single excitations are included and are to the left of the double excitations.
- The double excitations are ordered based on the absolute value of the MP2 parameters (right to left).
- Truncate the UCC ansatz by removing the double operators with the smallest absolute value of the MP2 parameters.
- Initial parameters are taken as the MP2 values.

Computational Methods

- Basis Sets include STO-3G, cc-pVDZ (FC), cc-pCVDZ (AE)

- Experimental geometries taken from the Computational Chemistry Comparison and Benchmark DataBase (CCCBDB)

- Molecular integrals and MP2 parameters taken from PySCF

- Sparse circuit simulators

  - After evaluating each UCC factor, test if the number of determinants in the wave function is greater than N_{CUT}

  - If this is the case, only keep the N_{WF} determinants with the largest absolute value of the corresponding amplitude

- The highest energy virtual orbitals are removed so that there is a maximum of 64 spin-orbitals.

Benchmarks

- STO-3G: No truncation of the wave function or ansatz

- cc-pVDZ (FC) and cc-pCVDZ (AE)

    - Only 5,000 double operators and all single operators

    - N_{WF} = 5,000

    - N_{CUT} = 8,000

- Error = UCCSD − CCSD(T)

Problem size for CH_2O

Basis	Parameters	Determinants
STO-3G	1,424	245,025
cc-pVDZ (FC)	34,398	10^{11}
cc-pCVDZ (AE)	52,704	10^{14}
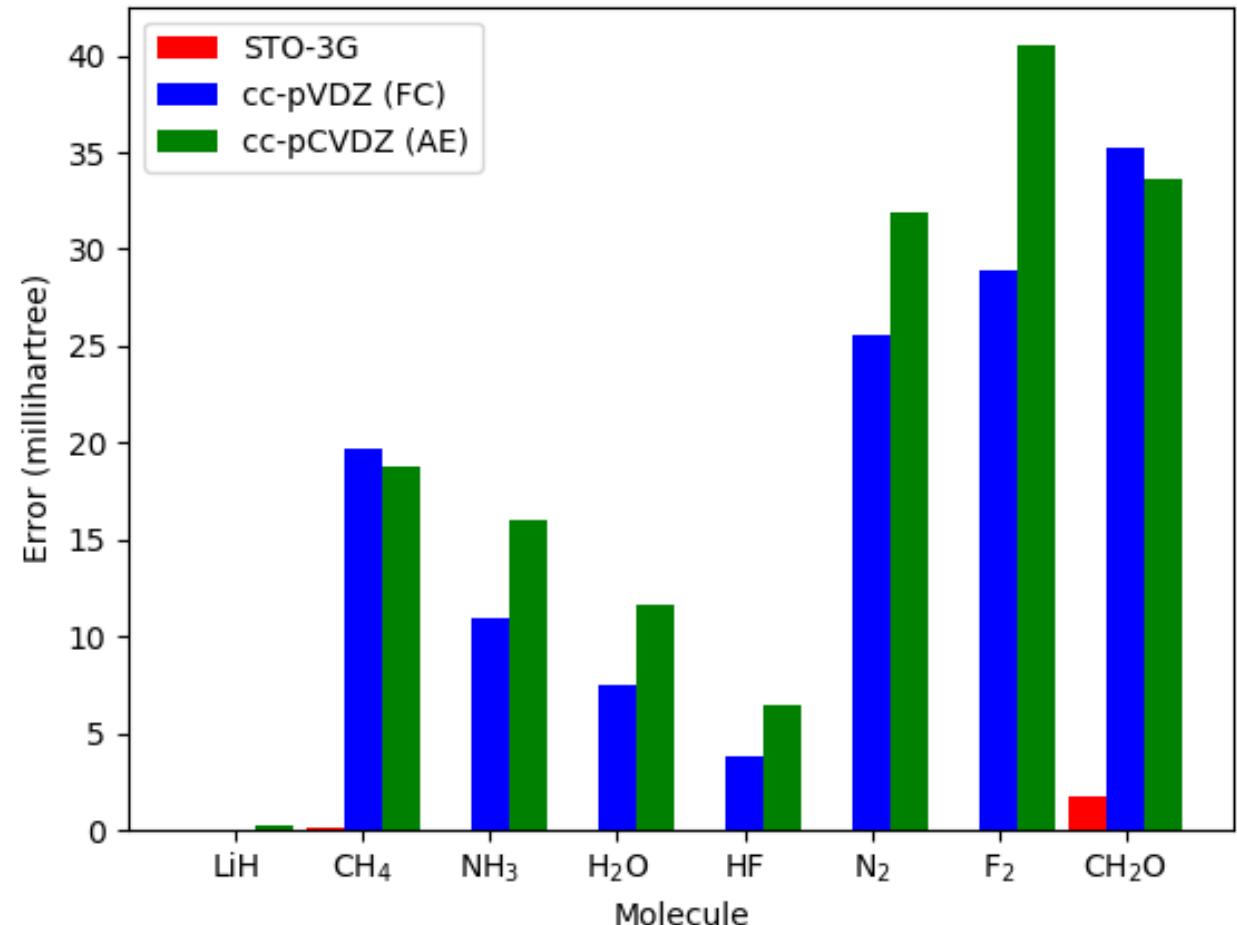
NH$_3$: Convergence
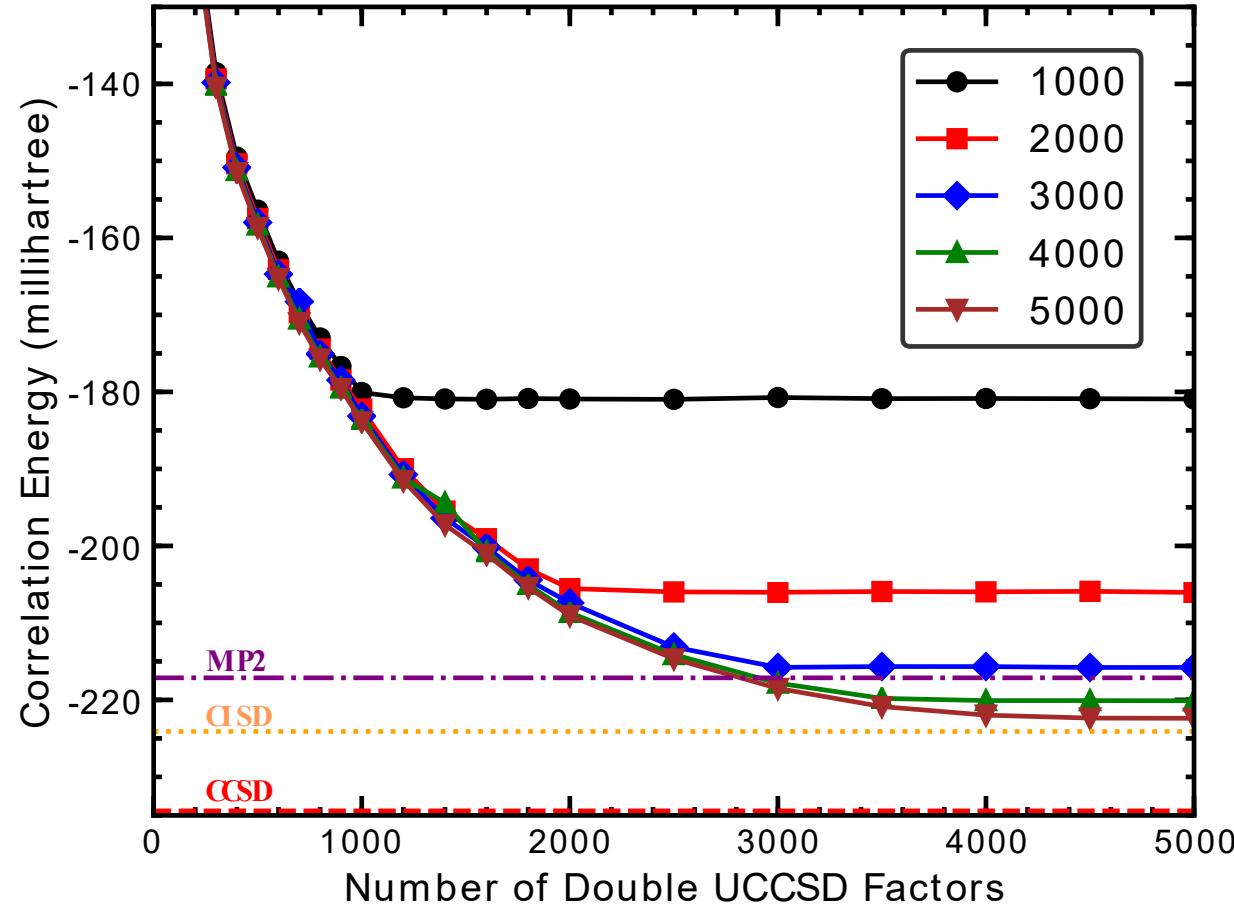
- NH$_3$ experimental geometry
- cc-pCVDZ (AE)
- 64 spin-orbitals
- The legend corresponds to N_{WF}
- N_{CUT} = 8,000
- All singles included
- Selection of doubles is based on the absolute value of the initial MP2 parameters
- Problem size without truncation
  - 10^{11} determinants
 - 25,515 parameters
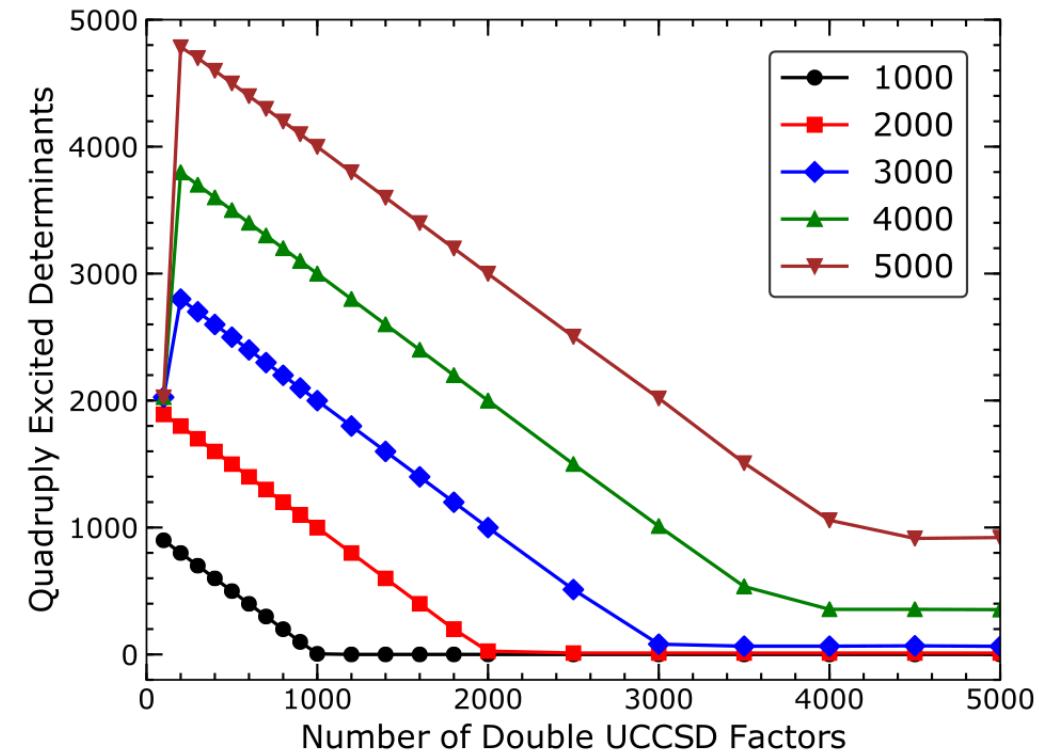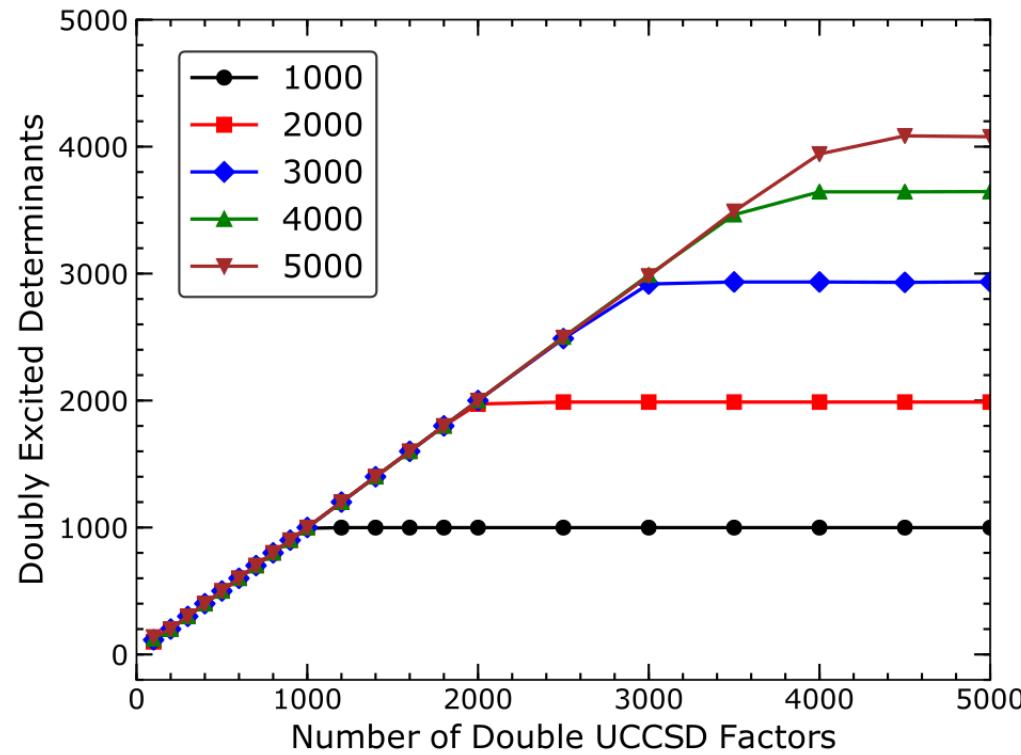
NH$_3$: Wavefunction
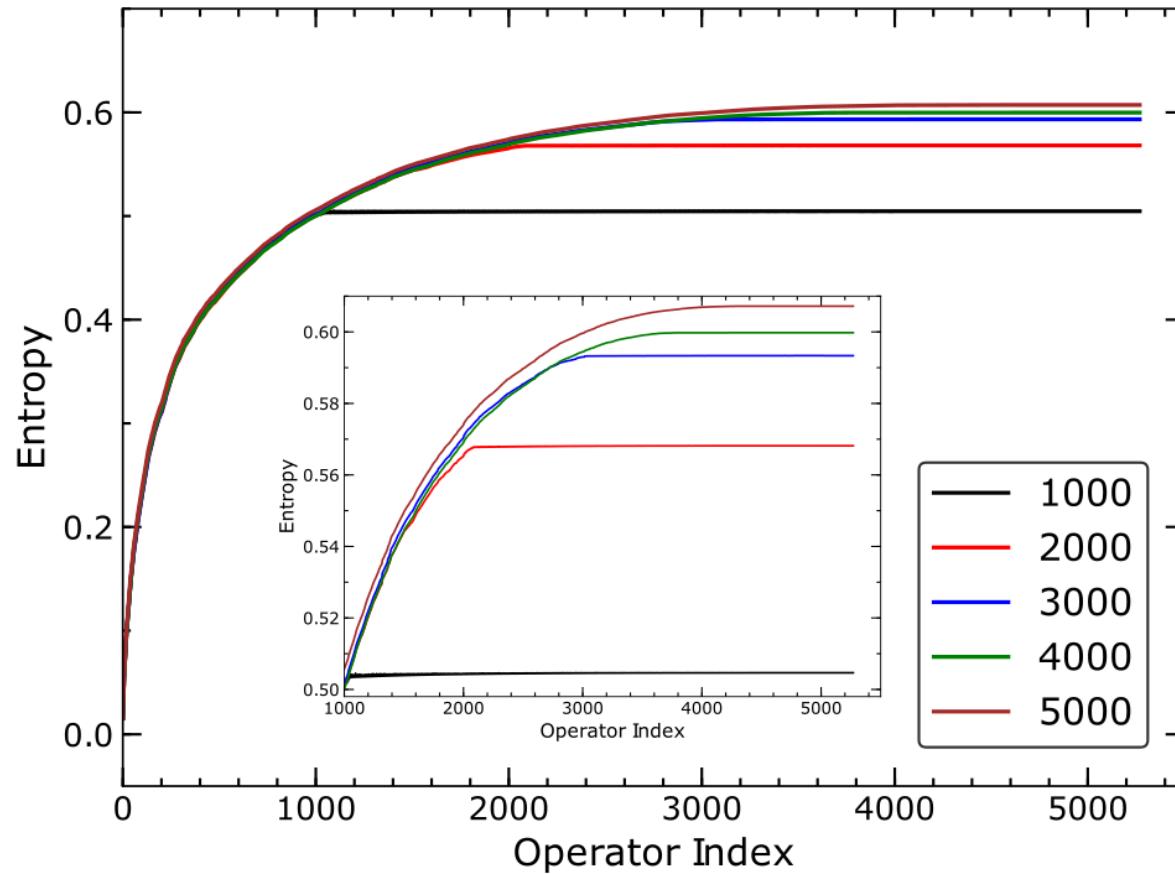

$$S = -\sum_{I}^{N_{det}} |c_I|^2 \, ln|c_I|^2$$

- 5,000 doubles operators in the ansatz
- Double operators applied first based on magnitude of the initial MP2 parameters
- Singles operators are index greater than 5,000
- Legend refers to N_{WF}
- $N_{CUT} = 8,000$

NH$_3$: Robust Parameters
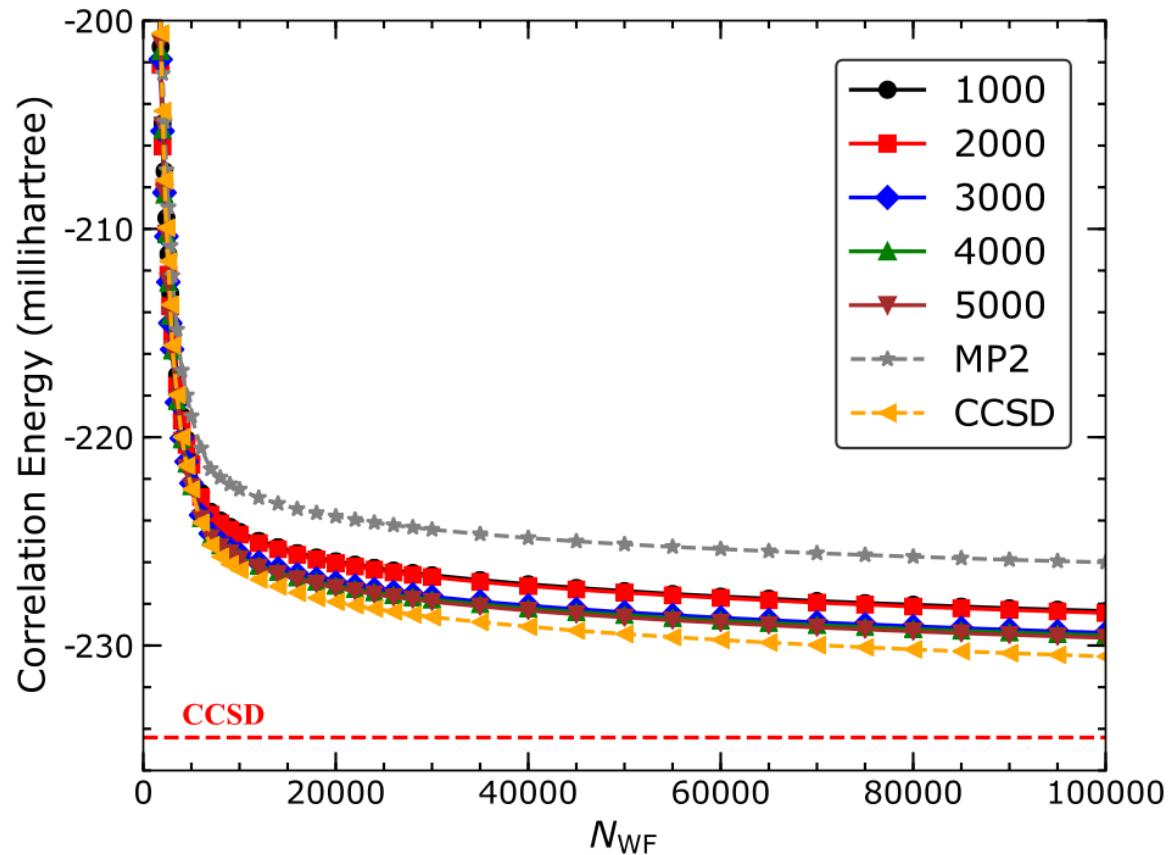
- Optimized parameters taken from optimization that included 5,000 doubles operators

- $N_{CUT} = N_{WF} + 1$

- Correlation energy computed by inserting the optimized parameters into the UCCSD ansatz but allowing the wave function to grow larger
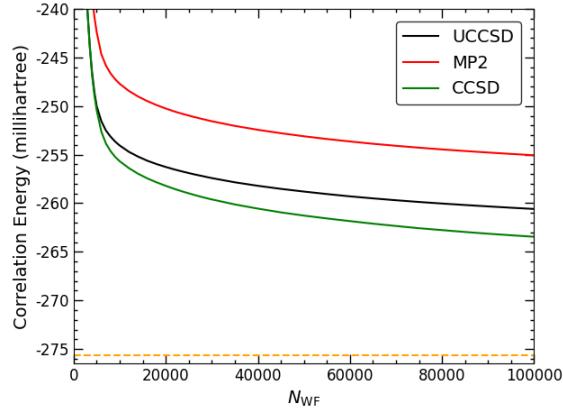
CH$_2$O
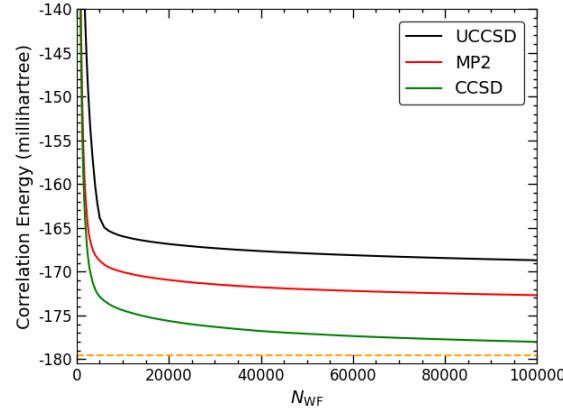
CH$_4$
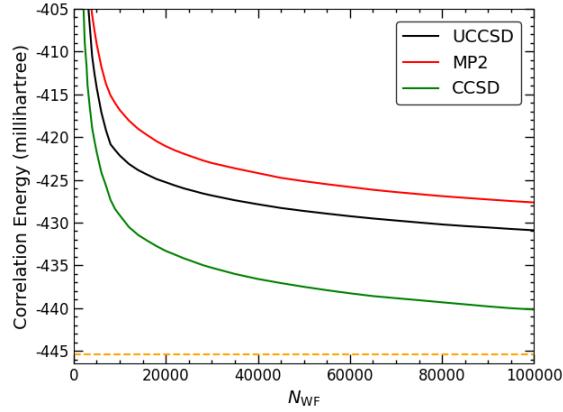
F$_2$
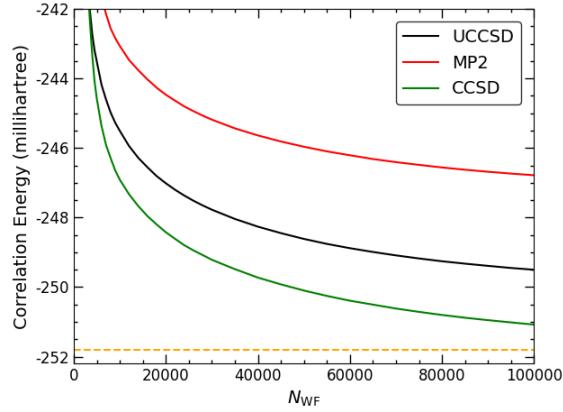
H$_2$O
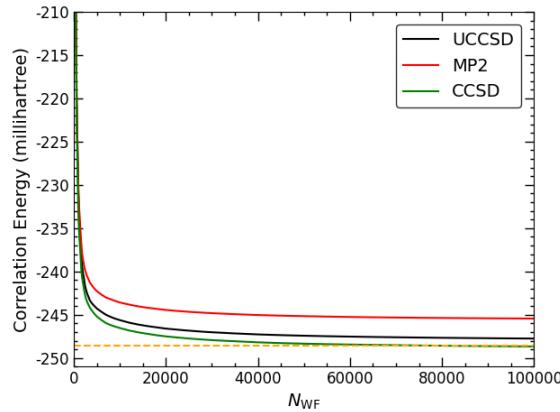
HF
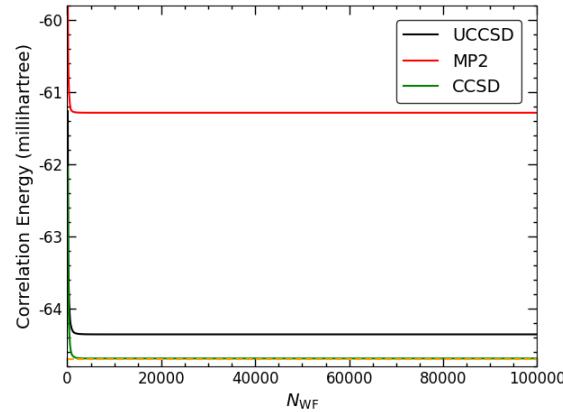
LiH
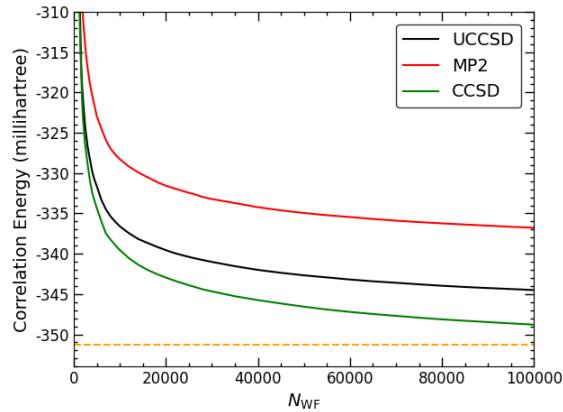
N$_2$
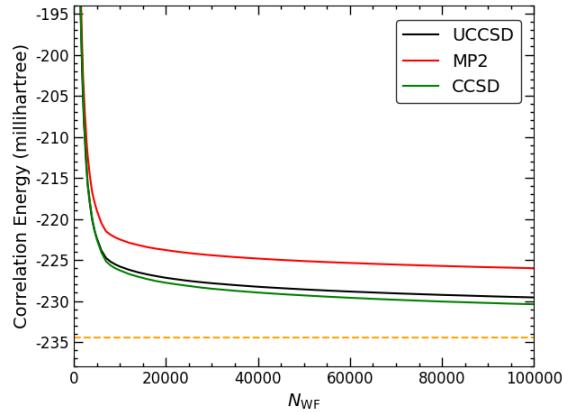
NH$_3$

Summary

- Implementation of new approach for analyzing VQE results employing UCC ansatz for up to 64 qubits with double-zeta quality basis sets

- Benchmark calculations of 8 molecules indicate larger error for larger basis sets

- Analysis of NH_3 with cc-pCVDZ basis indicates that most correlation energy recovered with only a fraction of wave function required

- Our results indicate a limit on the correlation energy recovered by the UCC ansatz for large basis sets

Acknowledgements

This work was funded by the Transformational Tools and Technologies (TTT) project.