Supporting information

Details of HCI calculations

Table 1: Parameters used in the HCI calculations used for generating trial states. The second column shows the orbitals used for obtaining the HCI states. Internal rotations were set to true in the HCISCF calculations. Correlation space refers to the size of the space (using the orbitals in the second column) for the final HCI calculation with the ϵ_1 shown in the last column.

Species and basis set	Orbitals	Correlation space	ϵ_1
H_{50} sto-6g	RHF	(50e, 50o)	10^{-4}
H_{10} cc-pVDZ	CASSCF(10e, 10o)	(10e, 10o)	10^{-6}
CrO Trail-Needs dz	HCISCF(10e, 13o) $\epsilon_1 = 10^{-5}$	(20e, 18o)	10^{-5}
FeO Trail-Needs dz	HCISCF(12e, 13o) $\epsilon_1 = 10^{-5}$	(22e, 18o)	10^{-5}
MnO Trail-Needs dz	HCISCF(11e, 13o) $\epsilon_1 = 10^{-5}$	(21e, 18o)	10^{-5}
TiO Trail-Needs dz	HCISCF(8e, 13o) $\epsilon_1 = 10^{-5}$	(18e, 18o)	10^{-5}
NH_3 6-31g	RHF	(10e, 15o)	10^{-6}
CO aug-cc-pVQZ	CASSCF(10e,8o)	(14e, 50o)	5×10^{-5}
BF aug-cc-pVQZ	CASSCF(10e,8o)	(14e, 50o)	5×10^{-5}
H_2O aug-cc-pVQZ	CASSCF(8e, 6o)	(10e, 50o)	1×10^{-5}

Geometries

Section 4.2 Transition metal oxides:
Bond lengths for energies shown in Figure 3:
FeO: 1.616 Å
CrO: 1.621 Å
MnO: 1.648 Å
TiO: 1.623 Å

Section 4.3 Dipole moments:
NH_3: N-H bond length: 1.07 Å, H-N-H angle: 100.08°
CO: C-O bond length: 1.1282 Å
BF: B-F bond length: 1.2669 Å
H_2O: O-H bond length: 0.958 Å, H-O-H angle: 104.4776°

Dipole moments

Figures 1 and 2 show the convergence of the dipole moment estimators with the number of configurations in the trial state for BF and H_2O, respectively.

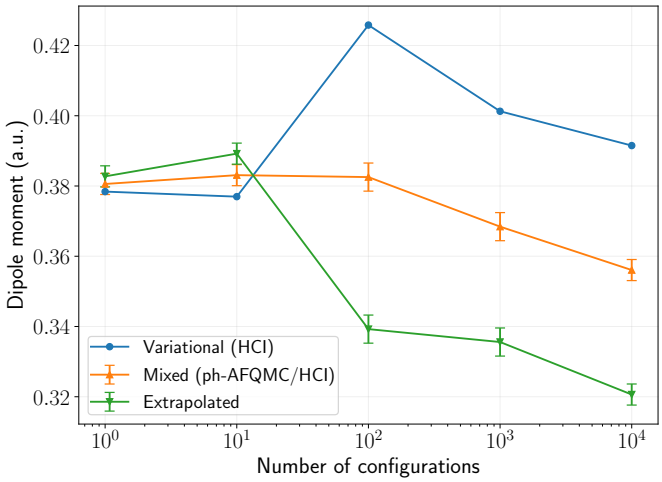

Figure 1: Convergence of different estimators for the dipole moment of BF using the aug-cc-pVQZ basis. Variational and mixed estimators are calculated using HCI and ph-AFQMC/HCI, respectively. Trial states are obtained by truncating an active space HCI wave function.

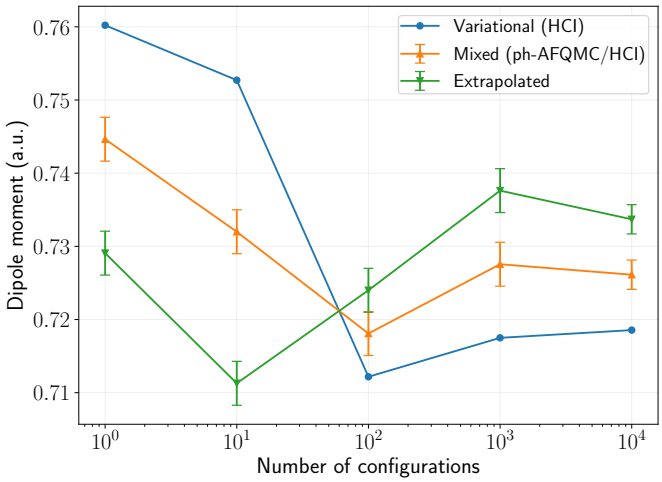

Figure 2: Convergence of different estimators for the dipole moment of H_2O using the aug-cc-pVQZ basis. Variational and mixed estimators are calculated using HCI and ph-AFQMC/HCI, respectively. Trial states are obtained by truncating an active space HCI wave function.