Table 1: Ground states, DMC energies (BFD-VTZ/sm666) in E_h, and spin-orbit (SO) corrections (in eV) of the different atomic species.

Element	Ground State	Optimization level	Energy	SO correction
Zn	^{1}S	Jas+MO	-227.0565(5)	n/a
Fe	^{5}D	Jas+MO	-123.8126(4)	-0.050
Cr	^{7}S	Jas+MO	-86.9010(4)	n/a
O	^{3}P	Jas+MO	-15.8938(1)	-0.010
H	^{2}S	/	-0.5000	n/a
S	^{3}P	Jas+MO	-10.1314(1)	-0.024

Table 2: FeO VMC and DMC energies in E_h at various optimization levels, using different starting orbitals and BFD-VTZ/sm666.

Ansatz	Orbitals	Optimization level	VMC energy	DMC energy
Single det	HF	Jas	-139.7003(5)	-139.8099(6)
	B3LYP	Jas	-139.7326(4)	-139.8394(6)
	opt	Jas+MO	-139.7499(4)	-139.8445(6)
[12,9]-CAS	CAS	Jas	-139.7369(4)	-139.8239(6)
	CAS	Jas+CI	-139.7552(4)	-139.8421(6)
	opt	Jas+MO+CI	-139.7708(3)	-139.8550(6)

Table 3: FeH VMC and DMC energies in E_h at various optimization levels, using different starting orbitals and BFD-VTZ/sm666.

Ansatz	Orbitals	Optimization level	VMC energy	DMC energy
Single det	HF	Jas	-124.2815(2)	-124.3443(5)
	B3LYP	Jas	-124.2923(2)	-124.3519(5)
	opt	Jas+MO	-124.2948(2)	-124.3519(5)
[9,7]-CAS	CAS	Jas	-124.2940(2)	-124.3548(5)
	CAS	Jas+CI	-124.3030(2)	-124.3647(5)
	opt	Jas+MO+CI	-124.3252(2)	-124.3802(5)

Table 4: CrS VMC and DMC energies in E_h at various optimization levels, using different starting orbitals and BFD-VTZ/sm666.

Ansatz	Orbitals	Optimization level	VMC energy	DMC energy
Single det	HF	Jas	-97.0284(2)	-97.1041(5)
	B3LYP	Jas	-97.0543(2)	-97.1304(5)
	opt	Jas+MO	-97.0570(2)	-97.1306(5)
CAS	RAS2	Jas	-97.0655(2)	-97.1318(5)
	RAS2	Jas+CI	-97.0778(2)	-97.1406(5)
	opt	Jas+MO+CI	-97.0822(3)	-97.1426(4)