Supplementary Information (SI) for Journal of Materials Chemistry C.

Double-hybrid density functionals applied to a large set of INVEST systems: validating the (SOS1-)PBE-DH-INVEST expressions

P. Maiz-Pastor, A.J. Pérez-Jiménez,
and J. C. Sancho-García*

Department of Physical Chemistry,
University of Alicante,
E-03080 Alicante, Spain

May 27, 2025

*E-mail: jc.sancho@ua.es

Table S1. Molecules constituting the NAH159 dataset.

001-azulene-2nh2	028-cyclazine-1cn-lumo	055-heptazine-1cf3-lumo
002-azulene-2nh2-4me	029-cyclazine-1nh2-homo	056-heptazine-1cn-lumo
003-azulene-2oh-2me	030-cyclazine-1nme2-homo	057-heptazine-1nh2
004-azulene-2oh-2nh2	031-cyclazine-1oh-homo	058-heptazine-1nme2
005-azulene-2oh-alt	032-cyclazine-1ome-homo	059-heptazine-1oh
006-azulene-2ome-2nh2	033-cyclazine-2cf3-side	060-heptazine-1ome
007-azulene-2ome-alt	034-cyclazine-2cf3-tip	061-heptazine-2cn-lumo
008-azulene-4cf3	035-cyclazine-2cn-lumo	062-heptazine-2nh2
009-azulene-4f-lumo	036-cyclazine-2cn-side	063-heptazine-2nme2
010-azulene-4me-lumo	037-cyclazine-2cn-tip	064-heptazine-2oh
011-azupyrene-2cf3	038-cyclazine-2nh2	065-heptazine-2ome
012-azupyrene-2nh2	039-cyclazine-2nh2-homo	066-heptazine-3cn-lumo
013-azupyrene-2oh	040-cyclazine-2nh2-homo-alt	067-heptazine-3nh2
014-azupyrene-2oh-cf3	041-cyclazine-2oh	068-heptazine-3nme2
015-azupyrene-2ome	042-cyclazine-2oh-2cf3	069-heptazine-3oh
016-azupyrene-2ome-cf3	043-cyclazine-2oh-homo	070-heptazine-3ome
017-azupyrene-4cn	044-cyclazine-2oh-homo-alt	071-isopyrene-1cf3
018-azupyrene-4nh2	045-cyclazine-2ome	072-isopyrene-1cn-1
019-azupyrene-4oh	046-cyclazine-2ome-homo	073-isopyrene-1cn-1-2nh2-3-9
020-azupyrene-4oh-2cf3	047-cyclazine-2ome-homo-alt	074-isopyrene-1cn-2
021-azupyrene-4oh-4me	048-cyclazine-3cn	075-isopyrene-1cn-3
022-azupyrene-4oh-6f	049-cyclazine-3cn-lumo	076-isopyrene-1cn-6-2nh2-2-10
023-azupyrene-4ome	050-cyclazine-3nh2	077-isopyrene-1cn-6-2nh2-3-9
024-cyclazine	051-cyclazine-3nme2	078-isopyrene-2cf3
025-cyclazine-1cf3	052-cyclazine-3oh	079-isopyrene-2cf3-lumo
026-cyclazine-1cf3-lumo	053-cyclazine-3ome	080-isopyrene-2cn-1-6
027-cyclazine-1cn	054-heptazine	081-isopyrene-2cn-2-10

Table S1. Molecules constituting the NAH159 dataset (cont.)

082-isopyrene-2cn-2-7	109-mol15-2nh2	136-pentaazaphenalene-1cf3
083-isopyrene-2cn-lumo	110-mol15-2oh	137-pentaazaphenalene-1cf3-lumo
084-isopyrene-2nh2	111-mol15-2ome	138-pentaazaphenalene-1cn
085-isopyrene-2nh2-2-10	112-mol15-4nh2	139-pentaazaphenalene-1cn-lumo
086-isopyrene-2nh2-homo	113-mol15-4oh	140-pentaazaphenalene-1nh2
087-isopyrene-2oh	114-mol62	141-pentaazaphenalene-1nme2
088-isopyrene-2oh-homo	115-mol62-2cf3	142-pentaazaphenalene-1oh
089-isopyrene-2ome	116-mol62-4nh2	143-pentaazaphenalene-1ome
090-isopyrene-2ome-homo	117-mol62-4oh	144-pentaazaphenalene-2cf3
091-isopyrene-4cn-2-5-7-10	118-mol76	145-pentaazaphenalene-2cf3-lumo
092-isopyrene-4cn-3-4-8-9	119-mol76-4nh2	146-pentaazaphenalene-2cn
093-isopyrene-4me	120-mol77	147-pentaazaphenalene-2cn-lumo
094-isopyrene-4nh2-2-5-7-10	121-mol77-2oh	148-pentaazaphenalene-2nh2
095-isopyrene-4oh	122-mol8	149-pentaazaphenalene-2nh2-homo
096-isopyrene-4oh-2cn	123-mol8-2cf3	150-pentaazaphenalene-2nme2
097-mol136	124-mol8-2cn	151-pentaazaphenalene-2oh
098-mol136-2nh2	125-mol8-2nh2	152-pentaazaphenalene-2oh-homo
099-mol136-2oh	126-mol8-2oh	153-pentaazaphenalene-2ome
100-mol143	127-mol8-2ome	154-pentaazaphenalene-2ome-homo
101-mol143-2oh	128-mol8-4oh	155-pentaazaphenalene-3cn-lumo
102-mol15	129-mol8-4ome	156-pentaazaphenalene-3nh2
103-mol151	130-mol86	157-pentaazaphenalene-3nme2
104-mol151-2cf3	131-mol86-3nh2	158-pentaazaphenalene-3oh
105-mol151-3nh2	132-mol86-3oh	159-pentaazaphenalene-3ome
106-mol151-3oh	133-mol98	
107-mol15-2cf3	134-mol98-3oh	
108-mol15-2cn	135-pentaazaphenalene	

Table S2. Vertical excitation energies (all in eV) calculated with the aug-cc-pVTZ basis set.

	$S_1 \leftarrow S_0$			$T_1 \leftarrow S_0$		
Molecule	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
001	2.777	3.360	2.878	2.419	2.099	2.491
002	2.546	3.093	2.630	2.434	2.032	2.491
003	2.435	3.029	2.523	2.451	2.464	2.459
004	2.984	3.665	3.110	2.675	2.520	2.798
005	2.638	3.266	2.733	2.672	2.705	2.687
006	2.919	3.633	3.044	2.549	2.496	2.703
007	2.539	3.187	2.635	2.571	2.639	2.599
008	2.388	3.135	2.523	2.452	2.632	2.501
009	2.930	3.501	3.010	2.829	2.356	2.823
010	2.462	3.100	2.570	2.501	2.559	2.533
011	2.003	2.585	2.099	2.064	2.232	2.104
012	2.263	2.922	2.368	2.310	2.415	2.380
013	2.196	2.824	2.289	2.274	2.411	2.332
014	2.243	2.900	2.337	2.318	2.504	2.392
015	2.179	2.784	2.264	2.253	2.370	2.302
016	2.232	2.868	2.319	2.301	2.467	2.372
017	1.913	2.519	1.999	1.995	2.159	2.036
018	2.714	3.573	2.820	2.705	2.988	2.791
019	2.523	3.275	2.609	2.554	2.735	2.632
020	2.769	3.639	2.872	2.762	3.007	2.856
021	2.406	3.159	2.490	2.352	2.643	2.470
022	2.346	3.158	2.441	2.220	2.524	2.333
023	2.468	3.189	2.545	2.481	2.663	2.562
024	1.069	1.664	1.182	1.198	1.358	1.275
025	1.196	1.826	1.315	1.320	1.503	1.408
026	1.096	1.652	1.204	1.159	1.248	1.229
027	1.252	1.895	1.378	1.341	1.526	1.444
028	0.959	1.481	1.052	1.047	1.111	1.100
029	1.332	1.821	1.442	1.250	1.245	1.322
030	1.219	1.761	1.331	1.235	1.311	1.316
031	1.346	1.831	1.456	1.240	1.221	1.311
032	1.409	1.899	1.519	1.295	1.278	1.367
033	1.271	1.936	1.390	1.407	1.621	1.499
034	1.321	1.989	1.448	1.445	1.654	1.547
035	0.897	1.361	0.978	0.962	0.967	1.001
036	1.250	1.938	1.373	1.363	1.595	1.460
037	1.319	2.011	1.452	1.426	1.666	1.538
038	1.591	2.358	1.727	1.725	1.982	1.841
039	1.013	1.491	1.120	1.009	1.026	1.074
040	1.007	1.476	1.111	1.015	1.010	1.077

Table S2. Vertical excitation energies (all in eV) calculated with the aug-cc-pVTZ basis set (cont.)

	$S_1 \leftarrow S_0$			$T_1 \leftarrow S_0$		
Molecule	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
041	1.521	2.247	1.647	1.664	1.906	1.763
042	1.787	2.568	1.918	1.910	2.199	2.020
043	1.113	1.601	1.223	1.069	1.073	1.135
044	1.092	1.582	1.203	1.059	1.054	1.125
045	1.484	2.202	1.606	1.607	1.862	1.709
046	0.985	1.487	1.099	0.977	1.014	1.052
047	1.018	1.499	1.127	1.011	1.004	1.074
048	1.414	2.134	1.544	1.495	1.753	1.617
049	0.957	1.368	1.041	0.948	0.870	0.980
050	1.828	2.694	1.977	2.033	2.466	2.165
051	1.922	2.767	2.050	2.117	2.541	2.236
052	1.758	2.558	1.890	1.962	2.309	2.068
053	1.837	2.593	1.956	1.981	2.270	2.082
054	2.790	4.065	2.931	3.023	3.693	3.161
055	2.751	4.018	2.886	2.967	3.616	3.105
056	2.694	3.921	2.817	2.884	3.470	3.012
057	3.153	4.467	3.300	3.324	3.933	3.467
058	3.215	4.512	3.354	3.357	3.939	3.491
059	3.077	4.388	3.221	3.282	3.922	3.428
060	3.078	4.383	3.219	3.271	3.906	3.418
061	2.617	3.812	2.729	2.801	3.376	2.918
062	3.497	4.866	3.653	3.617	4.164	3.758
063	3.584	4.932	3.728	3.652	4.155	3.768
064	3.369	4.721	3.518	3.551	4.145	3.697
065	3.366	4.706	3.508	3.555	4.121	3.690
066	2.552	3.726	2.658	2.747	3.324	2.857
067	3.873	5.339	4.038	4.192	5.058	4.376
068	3.989	5.437	4.135	4.312	4.880	4.431
069	3.708	5.122	3.859	4.022	4.821	4.209
070	3.673	5.085	3.821	3.958	4.794	4.151
071	2.048	2.688	2.099	2.186	2.384	2.179
072	2.035	2.691	2.084	2.176	2.397	2.180
073	2.153	2.130	1.583	2.266	2.130	1.583
074	1.987	2.509	2.009	2.038	2.182	2.029
075	1.925	2.452	1.949	2.002	2.127	1.984
076	1.995	2.763	2.071	2.102	2.347	2.216
077	2.118	2.811	2.191	2.157	2.340	2.216
078	2.097	2.845	2.164	2.267	2.612	2.261
079	1.839	2.356	1.865	1.964	2.053	1.912
080	2.058	2.831	2.114	2.204	2.619	2.213

Table S2. Vertical excitation energies (all in eV) calculated with the aug-cc-pVTZ basis set (cont.)

	$S_1 \leftarrow S_0$			$T_1 \leftarrow S_0$		
Molecule	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
081	1.957	2.455	1.974	1.987	2.136	2.004
082	1.946	2.436	1.957	1.943	2.072	1.940
083	1.855	2.363	1.877	1.969	2.047	1.919
084	2.073	2.679	2.122	2.176	2.285	2.157
085	1.911	2.516	1.974	1.986	2.062	2.006
086	1.533	1.863	1.544	1.502	1.412	1.441
087	2.053	2.622	2.092	2.172	2.282	2.137
088	1.596	2.000	1.607	1.590	1.567	1.533
089	1.957	2.531	2.000	2.085	2.208	2.050
090	1.597	2.028	1.619	1.586	1.585	1.549
091	1.915	2.395	1.925	1.800	2.146	1.852
092	1.702	2.170	1.713	1.816	1.865	1.752
093	1.936	2.520	1.968	2.072	2.239	2.026
094	1.939	2.599	2.006	2.001	1.885	2.039
095	2.155	2.717	2.181	2.287	2.418	2.235
096	2.233	3.033	2.289	2.331	2.808	2.415
097	2.201	2.749	2.300	2.204	2.193	2.305
098	2.583	3.234	2.691	2.195	1.984	2.312
099	2.636	3.279	2.743	2.410	2.071	2.494
100	2.149	2.728	2.254	2.156	2.169	2.173
101	2.515	3.122	2.613	2.271	1.908	2.354
102	1.460	2.063	1.572	1.478	1.576	1.556
103	0.942	1.475	1.047	0.957	1.032	1.022
104	1.029	1.621	1.152	1.049	1.170	1.138
105	1.399	2.188	1.533	1.486	1.800	1.624
106	1.318	2.043	1.436	1.408	1.663	1.515
107	1.597	2.278	1.722	1.628	1.798	1.734
108	1.494	2.174	1.621	1.540	1.700	1.725
109	1.812	2.526	1.939	1.899	2.012	2.078
110	1.730	2.383	1.845	1.801	1.881	1.935
111	1.605	2.229	1.716	1.658	1.735	1.764
112	2.257	3.077	2.385	2.336	2.633	2.544
113	2.107	2.850	2.219	2.218	2.366	2.401
114	1.321	1.741	1.368	1.250	1.129	1.244
115	1.263	1.982	1.355	1.372	1.475	1.605
116	1.560	2.240	1.630	1.676	1.561	1.812
117	1.497	2.104	1.551	1.625	1.587	1.796
118	1.450	2.084	1.590	1.400	1.434	1.577
119	2.020	2.817	2.206	1.932	1.884	2.140
120	2.133	2.816	2.263	2.130	1.778	2.215

Table S2. Vertical excitation energies (all in eV) calculated with the aug-cc-pVTZ basis set (cont.)

	$S_1 \leftarrow S_0$			$T_1 \leftarrow S_0$		
Molecule	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	LR-CC2	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
121	2.371	3.095	2.509	2.207	2.068	2.317
122	1.764	2.454	1.854	1.792	1.947	1.910
123	1.888	2.709	1.999	1.939	2.164	2.248
124	1.806	2.603	1.907	1.862	2.125	1.959
125	2.018	2.740	2.118	2.089	2.249	2.169
126	1.939	2.605	2.024	2.015	2.143	2.064
127	1.848	2.462	1.913	1.934	2.028	1.951
128	2.166	2.886	2.240	2.280	2.579	2.382
129	1.989	2.694	2.066	2.085	2.439	2.338
130	1.866	2.352	1.942	1.809	1.821	1.917
131	2.296	3.059	2.448	2.228	2.169	2.391
132	2.225	2.921	2.336	2.191	2.247	2.418
133	1.545	2.255	1.646	1.560	1.805	1.616
134	1.982	2.773	2.092	1.648	1.472	1.707
135	2.252	3.250	2.403	2.380	2.671	2.550
136	2.281	3.331	2.433	2.429	2.788	2.604
137	2.180	3.163	2.330	2.278	2.493	2.433
138	2.240	3.297	2.388	2.377	2.746	2.549
139	2.121	3.062	2.265	2.184	2.320	2.311
140	2.566	3.666	2.711	2.722	3.250	2.873
141	2.591	3.673	2.729	2.735	3.245	2.882
142	2.475	3.551	2.618	2.644	3.138	2.802
143	2.480	3.548	2.621	2.640	3.127	2.799
144	2.317	3.417	2.468	2.483	2.922	2.658
145	2.208	3.181	2.347	2.321	2.620	2.482
146	2.223	3.324	2.366	2.366	2.810	2.524
147	2.084	2.987	2.194	2.180	2.475	2.305
148	2.858	3.887	3.047	2.786	2.860	2.864
149	1.902	2.698	2.053	1.821	1.742	1.945
150	2.915	3.921	3.090	2.794	2.862	2.862
151	2.794	3.808	2.972	2.772	2.853	2.862
152	1.983	2.859	2.137	1.958	1.982	2.113
153	2.739	3.761	2.915	2.740	2.827	2.828
154	2.068	3.010	2.222	2.120	2.288	2.291
155	1.998	2.872	2.111	2.091	2.280	2.224
156	3.169	4.350	3.358	3.319	3.679	3.465
157	3.244	4.399	3.419	3.356	3.692	3.474
158	3.048	4.181	3.220	3.213	3.521	3.363
159	3.045	4.163	3.210	3.194	3.491	3.353

Table S3. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set.

	$S_1 \leftarrow S_0$		$T_1 \leftarrow S_0$	
Molecule	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
001	3.360	2.892	2.100	2.488
002	3.088	2.639	2.032	2.489
003	3.022	2.537	2.461	2.469
004	3.661	3.125	2.516	2.799
005	3.263	2.754	2.706	2.704
006	3.630	3.063	2.493	2.705
007	3.185	2.656	2.641	2.616
008	3.135	2.525	2.636	2.501
009	3.484	3.028	2.349	2.822
010	3.104	2.590	2.568	2.551
011	2.582	2.102	2.232	2.103
012	2.918	2.379	2.416	2.386
013	2.817	2.300	2.409	2.337
014	2.894	2.347	2.503	2.396
015	2.779	2.275	2.368	2.308
016	2.863	2.330	2.466	2.377
017	2.515	2.003	2.158	2.038
018	3.567	2.843	2.987	2.807
019	3.263	2.630	2.732	2.648
020	3.627	2.892	3.001	2.870
021	3.146	2.509	2.638	2.479
022	3.151	2.453	2.525	2.340
023	3.180	2.567	2.662	2.577
024	1.665	1.201	1.364	1.292
025	1.827	1.331	1.509	1.424
026	1.652	1.224	1.253	1.246
027	1.896	1.393	1.532	1.457
028	1.483	1.073	1.118	1.120
029	1.823	1.457	1.252	1.333
030	1.764	1.347	1.318	1.329
031	1.835	1.470	1.228	1.321
032	1.902	1.535	1.286	1.378
033	1.937	1.407	1.627	1.514
034	1.990	1.464	1.660	1.561
035	1.364	1.002	0.974	1.022
036	1.938	1.388	1.599	1.472
037	2.011	1.466	1.671	1.549
038	2.356	1.748	1.985	1.857
039	1.494	1.133	1.033	1.085
040	1.479	1.126	1.018	1.089

Table S3. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	$S_1 \leftarrow S_0$		$T_1 \leftarrow S_0$	
Molecule	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
041	2.242	1.669	1.908	1.781
042	2.564	1.938	2.200	2.034
043	1.606	1.236	1.084	1.146
044	1.586	1.215	1.064	1.136
045	2.198	1.628	1.864	1.727
046	1.490	1.112	1.022	1.063
047	1.503	1.141	1.013	1.086
048	2.133	1.556	1.757	1.627
049	1.371	1.063	0.877	0.999
050	2.691	2.000	2.470	2.190
051	2.767	2.081	2.549	2.268
052	2.550	1.915	2.308	2.095
053	2.588	1.983	2.271	2.108
054	4.037	2.913	3.676	3.144
055	3.990	2.871	3.599	3.092
056	3.895	2.803	3.453	2.999
057	4.439	3.284	3.917	3.452
058	4.486	3.340	3.923	3.477
059	4.358	3.204	3.905	3.412
060	4.354	3.203	3.889	3.402
061	3.787	2.717	3.359	2.907
062	4.837	3.639	4.148	3.745
063	4.907	3.718	4.139	3.756
064	4.688	3.502	4.129	3.682
065	4.675	3.494	4.104	3.677
066	3.700	2.648	3.308	2.847
067	5.307	4.028	5.042	4.373
068	5.409	4.131	4.859	4.428
069	5.083	3.847	4.798	4.200
070	5.049	3.811	4.773	4.142
071	2.690	2.113	2.390	2.194
072	2.692	2.097	2.402	2.193
074	2.512	2.027	2.189	2.046
075	2.454	1.968	2.133	2.001
076	2.762	2.080	2.347	2.219
077	2.811	2.208	2.344	2.232
078	2.846	2.173	2.617	2.270
079	2.357	1.883	2.058	1.929
080	2.829	2.122	2.621	2.220

Table S3. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	$S_1 \leftarrow S_0$		$T_1 \leftarrow S_0$	
Molecule	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
081	2.458	1.993	2.143	2.022
082	2.440	1.976	2.080	1.958
083	2.366	1.896	2.054	1.938
084	2.681	2.143	2.291	2.175
085	2.518	1.988	2.067	2.017
086	1.865	1.545	1.416	1.439
087	2.624	2.113	2.289	2.157
088	2.003	1.608	1.575	1.531
089	2.533	2.020	2.214	2.069
090	2.030	1.619	1.590	1.546
091	2.398	1.943	2.155	1.868
092	2.173	1.735	1.872	1.773
093	2.522	1.987	2.245	2.045
094	2.603	2.025	1.887	2.041
095	2.719	2.209	2.423	2.262
096	3.029	2.306	2.808	2.429
097	2.751	2.311	2.198	2.311
098	3.234	2.706	1.985	2.310
099	3.278	2.760	2.073	2.493
100	2.730	2.266	2.174	2.182
101	3.121	2.629	1.910	2.353
102	2.060	1.575	1.576	1.555
103	1.472	1.049	1.030	1.019
104	1.618	1.153	1.169	1.134
105	2.183	1.546	1.798	1.632
106	2.034	1.447	1.657	1.523
107	2.275	1.723	1.799	1.731
108	2.170	1.620	1.699	1.715
109	2.522	1.949	2.013	2.079
110	2.376	1.854	1.879	1.935
111	2.223	1.724	1.734	1.764
112	3.071	2.404	2.563	2.550
113	2.838	2.237	2.360	2.403
114	1.740	1.375	1.130	1.247
115	1.978	1.353	1.474	1.601
116	2.235	1.647	1.561	1.815
117	2.097	1.567	1.587	1.801
118	2.085	1.595	1.438	1.575
119	2.816	2.222	1.888	2.143
120	2.814	2.273	1.780	2.214

Table S3. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	$S_1 \leftarrow S_0$		$T_1 \leftarrow S_0$	
Molecule	PBE-DH-INVEST(SCF)	PBE-DH-INVEST	PBE-DH-INVEST(SCF)	PBE-DH-INVEST
121	3.091	2.522	2.071	2.322
122	2.453	1.863	1.950	1.916
123	2.707	2.001	2.165	2.244
124	2.599	1.907	2.124	1.956
125	2.739	2.135	2.252	2.183
126	2.602	2.041	2.145	2.077
127	2.459	1.930	2.030	1.965
128	2.880	2.264	2.577	2.405
129	2.688	2.087	2.437	2.356
130	2.351	1.949	1.823	1.918
131	3.056	2.465	2.168	2.390
132	2.915	2.351	2.245	2.419
133	2.254	1.656	1.805	1.623
134	2.769	2.109	1.469	1.707
135	3.233	2.398	2.672	2.550
136	3.314	2.427	2.787	2.602
137	3.147	2.328	2.493	2.440
138	3.279	2.382	2.744	2.547
139	3.047	2.263	2.322	2.318
140	3.647	2.709	3.240	2.869
141	3.655	2.728	3.236	2.878
142	3.530	2.615	3.128	2.800
143	3.529	2.619	3.118	2.797
144	3.399	2.461	2.918	2.654
145	3.164	2.349	2.618	2.483
146	3.306	2.359	2.806	2.519
147	2.971	2.197	2.472	2.305
148	3.874	3.047	2.865	2.874
149	2.685	2.043	1.749	1.944
150	3.911	3.095	2.867	2.872
151	3.792	2.971	2.858	2.872
152	2.847	2.126	1.992	2.111
153	3.745	2.914	2.831	2.837
154	2.999	2.215	2.300	2.291
155	2.857	2.114	2.280	2.229
156	4.333	3.363	3.680	3.484
157	4.386	3.431	3.696	3.496
158	4.157	3.224	3.518	3.380
159	4.142	3.215	3.490	3.370

Table S4. Oscillator strengths (all in a.u.) calculated with different basis sets.

Molecule	aug-cc-pVTZ	aug-cc-pVDZ
001	0.00168	0.00168
002	0.00009	0.00011
003	0.01106	0.01128
004	0.01215	0.01194
005	0.01083	0.01107
006	0.00565	0.00556
007	0.01360	0.01385
008	0.00716	0.00720
009	0.00276	0.00308
010	0.01481	0.01493
011	0.00000	0.00000
012	0.01277	0.01279
013	0.00447	0.00442
014	0.00413	0.00411
015	0.00450	0.00449
016	0.00440	0.00441
017	0.00000	0.00000
018	0.00000	0.00000
019	0.00000	0.00000
020	0.00000	0.00000
021	0.00000	0.00000
022	0.00000	0.00000
023	0.00000	0.00000
024	0.00000	0.00000
025	0.00020	0.00020
026	0.00040	0.00042
027	0.00052	0.00051
028	0.00141	0.00144
029	0.00123	0.00126
030	0.00026	0.00026
031	0.00107	0.00110
032	0.00062	0.00063
033	0.00038	0.00039
034	0.00020	0.00019
035	0.00088	0.00090
036	0.00602	0.00610
037	0.00033	0.00035
038	0.00560	0.00566
039	0.00011	0.00011
040	0.00202	0.00205

Table S4. Oscillator strengths (all in a.u.) calculated with different basis sets (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ
041	0.00267	0.00267
042	0.00173	0.00176
043	0.00072	0.00076
044	0.00073	0.00073
045	0.00352	0.00354
046	0.00029	0.00030
047	0.00192	0.00192
048	0.00000	0.00000
049	0.00000	0.00000
050	0.00000	0.00000
051	0.00000	0.00000
052	0.00000	0.00000
053	0.00000	0.00000
054	0.00000	0.00000
055	0.00052	0.00052
056	0.00252	0.00248
057	0.01583	0.01560
058	0.01982	0.01979
059	0.00899	0.00872
060	0.00909	0.00887
061	0.00172	0.00170
062	0.03132	0.03064
063	0.05338	0.05280
064	0.01976	0.01912
065	0.02114	0.02047
066	0.00000	0.00000
067	0.00000	0.00000
068	0.00000	0.00000
069	0.00062	0.00060
070	0.00068	0.00066
071	0.00439	0.00443
072	0.00389	0.00385
073	0.00000	0.00026
074	0.00026	0.00016
075	0.00016	0.01556
076	0.01498	0.02885
077	0.02862	0.00000
078	0.00000	0.00007
079	0.00006	0.00000
080	0.00000	0.00001

Table S4. Oscillator strengths (all in a.u.) calculated with different basis sets (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ
081	0.00001	0.00000
082	0.00000	0.00001
083	0.00001	0.00959
084	0.00948	0.02866
085	0.02816	0.00000
086	0.00000	0.00225
087	0.00224	0.00019
088	0.00019	0.00230
089	0.00226	0.00061
090	0.00061	0.00000
091	0.00000	0.00000
092	0.00000	0.00000
093	0.00000	0.00000
094	0.00000	0.00086
095	0.00085	0.00115
096	0.00113	0.02447
097	0.02427	0.00521
098	0.00519	0.00465
099	0.00454	0.01094
100	0.01084	0.00220
101	0.00213	0.00299
102	0.00298	0.00411
103	0.00407	0.00454
104	0.00450	0.00614
105	0.00594	0.00324
106	0.00311	0.00291
107	0.00291	0.00003
108	0.00002	0.00005
109	0.00004	0.00008
110	0.00007	0.00014
111	0.00012	0.02734
112	0.02670	0.00652
113	0.00634	0.00807
114	0.00798	0.00348
115	0.00345	0.00443
116	0.00443	0.00246
117	0.00246	0.01618
118	0.01609	0.07168
119	0.07094	0.00350
120	0.00345	0.03149

Table S4. Oscillator strengths (all in a.u.) calculated with different basis sets (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ
121	0.03257	0.00300
122	0.00297	0.00732
123	0.00722	0.01631
124	0.01630	0.00377
125	0.00366	0.00134
126	0.00129	0.00275
127	0.00265	0.00065
128	0.00066	0.00256
129	0.00256	0.00345
130	0.00345	0.02765
131	0.02749	0.01366
132	0.01372	0.01191
133	0.01175	0.00438
134	0.00419	0.01229
135	0.01293	0.01171
136	0.01226	0.01859
137	0.01929	0.01737
138	0.01808	0.02500
139	0.02589	0.00029
140	0.00039	0.00008
141	0.00013	0.00213
142	0.00233	0.00190
143	0.00210	0.01066
144	0.01112	0.00982
145	0.01038	0.02245
146	0.02325	0.00797
147	0.00849	0.10112
148	0.10302	0.03322
149	0.03462	0.14413
150	0.14537	0.06942
151	0.07151	0.02154
152	0.02272	0.06998
153	0.07215	0.02070
154	0.02175	0.01642
155	0.01716	0.05229
156	0.05395	0.07450
157	0.07602	0.03239
158	0.03380	0.03451
159	0.03592	0.03592

Table S5. Total CPU time (h) with different basis sets at the PBE-DH-INVEST level.

Molecule	aug-cc-pVTZ	aug-cc-pVDZ	Relative time
001	0.567	0.117	0.206
002	2.483	0.433	0.174
003	1.000	0.217	0.217
004	0.833	0.183	0.220
005	0.483	0.117	0.241
006	1.650	0.333	0.202
007	1.033	0.217	0.210
008	5.050	1.217	0.241
009	0.517	0.117	0.226
010	1.333	0.283	0.213
011	3.567	0.967	0.271
012	2.433	0.483	0.199
013	2.333	0.400	0.171
014	3.983	0.800	0.201
015	3.233	0.683	0.211
016	7.767	1.233	0.159
017	4.667	0.933	0.200
018	3.950	0.767	0.194
019	4.450	0.533	0.120
020	9.083	1.650	0.182
021	10.13	1.583	0.156
022	7.600	1.100	0.145
023	11.35	1.383	0.122
024	0.633	0.183	0.289
025	1.383	0.367	0.265
026	1.417	0.383	0.271
027	0.983	0.267	0.271
028	0.900	0.250	0.278
029	0.900	0.250	0.278
030	1.783	0.417	0.234
031	0.800	0.233	0.292
032	1.150	0.300	0.261
033	2.583	0.633	0.245
034	2.650	0.633	0.239
035	1.233	0.350	0.284
036	1.300	0.317	0.244
037	1.283	0.333	0.260
038	1.233	0.283	0.230
039	1.233	0.333	0.270
040	1.283	0.333	0.260

Table S5. Total CPU time (h) with different basis sets at the PBE-DH-INVEST level (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ	Relative time
041	0.950	0.267	0.281
042	3.317	0.717	0.216
043	1.033	0.283	0.274
044	1.033	0.300	0.290
045	1.767	0.417	0.236
046	1.900	0.450	0.237
047	1.917	0.450	0.235
048	1.650	0.433	0.263
049	1.683	0.433	0.257
050	1.733	0.367	0.212
051	6.950	0.383	0.055
052	1.150	0.333	0.290
053	2.583	0.567	0.219
054	0.383	0.100	0.261
055	0.883	0.250	0.283
056	0.567	0.167	0.294
057	0.517	0.167	0.323
058	1.067	0.300	0.281
059	0.517	0.150	0.290
060	0.717	0.200	0.279
061	0.750	0.200	0.267
062	0.717	0.183	0.256
063	2.367	0.600	0.254
064	0.600	0.150	0.250
065	1.217	0.317	0.260
066	0.983	0.300	0.305
067	0.983	0.250	0.254
068	5.033	1.100	0.219
069	0.767	0.200	0.261
070	1.900	0.517	0.272
071	2.750	0.633	0.230
072	1.733	0.483	0.279
073	2.783	0.750	0.269
074	1.733	0.483	0.279
075	1.733	0.500	0.288
076	2.933	0.700	0.239
077	3.300	0.700	0.212
078	5.850	0.967	0.165
079	4.283	0.933	0.218
080	2.733	0.617	0.226

Table S5. Total CPU time (h) with different basis sets at the PBE-DH-INVEST level (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ	Relative time
081	2.217	0.600	0.271
082	2.550	0.600	0.235
083	2.650	0.633	0.239
084	2.267	0.567	0.250
085	2.567	0.550	0.214
086	2.433	0.600	0.247
087	2.350	0.533	0.227
088	2.350	0.583	0.248
089	4.267	0.783	0.184
090	3.750	1.017	0.271
091	4.450	1.067	0.240
092	4.467	1.100	0.246
093	4.917	1.083	0.220
094	3.483	0.933	0.268
095	1.567	0.783	0.500
096	5.933	1.183	0.199
097	3.267	0.933	0.286
098	4.417	1.250	0.283
099	4.067	1.183	0.291
100	2.550	1.000	0.392
101	4.700	1.183	0.252
102	1.333	0.250	0.188
103	3.317	0.767	0.231
104	9.817	1.483	0.151
105	5.300	1.133	0.214
106	4.650	1.000	0.215
107	4.550	0.950	0.209
108	2.183	0.533	0.244
109	2.017	0.400	0.198
110	1.783	0.417	0.234
111	3.483	0.783	0.225
112	3.767	0.750	0.199
113	3.217	0.600	0.187
114	2.067	0.583	0.282
115	5.283	1.233	0.233
116	4.083	0.950	0.233
117	3.183	0.800	0.251
118	2.400	0.617	0.257
119	5.033	1.183	0.235
120	2.600	0.783	0.301

Table S5. Total CPU time (h) with different basis sets at the PBE-DH-INVEST level (cont.)

Molecule	aug-cc-pVTZ	aug-cc-pVDZ	Relative time
121	4.283	0.883	0.206
122	0.517	0.150	0.290
123	2.100	0.433	0.206
124	1.033	0.233	0.226
125	0.917	0.183	0.200
126	0.750	0.167	0.222
127	1.500	0.300	0.200
128	1.133	0.233	0.206
129	3.617	0.717	0.198
130	1.450	0.283	0.195
131	2.667	0.583	0.219
132	2.217	0.467	0.211
133	2.500	0.617	0.247
134	2.933	0.883	0.301
135	0.433	0.100	0.231
136	0.900	0.233	0.259
137	0.867	0.217	0.250
138	0.617	0.150	0.243
139	0.600	0.150	0.250
140	0.617	0.167	0.270
141	1.200	0.300	0.250
142	0.583	0.167	0.286
143	0.783	0.233	0.298
144	1.933	0.467	0.241
145	1.800	0.433	0.241
146	0.900	0.267	0.296
147	0.867	0.250	0.288
148	0.783	0.217	0.277
149	0.800	0.217	0.271
150	2.800	0.533	0.190
151	0.683	0.217	0.317
152	0.733	0.200	0.273
153	1.400	0.317	0.226
154	1.417	0.333	0.235
155	1.250	0.317	0.253
156	1.117	0.283	0.254
157	5.333	0.950	0.178
158	0.867	0.233	0.269
159	2.200	0.467	0.212

Table S6. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set.

	SOS1-PBE-DH-INVEST	
Molecule	$S_1 \leftarrow S_0$	$T_1 \leftarrow S_0$
001	2.790	2.488
002	2.582	2.478
003	2.508	2.560
004	3.009	2.827
005	2.693	2.774
006	2.965	2.747
007	2.610	2.690
008	2.470	2.586
009	2.913	2.812
010	2.534	2.609
011	2.145	2.210
012	2.394	2.455
013	2.323	2.421
014	2.371	2.486
015	2.299	2.389
016	2.354	2.465
017	2.044	2.144
018	2.866	2.916
019	2.644	2.735
020	2.902	2.974
021	2.531	2.589
022	2.493	2.449
023	2.586	2.665
024	1.241	1.428
025	1.374	1.561
026	1.284	1.385
027	1.450	1.596
028	1.120	1.253
029	1.557	1.470
030	1.420	1.465
031	1.580	1.463
032	1.646	1.519
033	1.446	1.651
034	1.509	1.699
035	1.057	1.158
036	1.432	1.609
037	1.522	1.692
038	1.796	1.990
039	1.210	1.217
040	1.193	1.212

Table S6. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	SOS1-PBE-DH-INVEST	
Molecule	$S_1 \leftarrow S_0$	$T_1 \leftarrow S_0$
041	1.709	1.916
042	1.988	2.181
043	1.329	1.288
044	1.307	1.274
045	1.672	1.865
046	1.186	1.194
047	1.214	1.212
048	1.619	1.766
049	1.149	1.141
050	2.053	2.353
051	2.139	2.432
052	1.955	2.246
053	2.039	2.257
054	2.986	3.356
055	2.948	3.303
056	2.886	3.203
057	3.352	3.639
058	3.409	3.660
059	3.272	3.607
060	3.274	3.597
061	2.804	3.115
062	3.704	3.906
063	3.784	3.911
064	3.563	3.857
065	3.559	3.848
066	2.734	3.059
067	4.095	4.590
068	4.202	4.572
069	3.905	4.405
070	3.879	4.359
071	2.115	2.264
072	2.104	2.265
073	2.274	2.418
074	2.033	2.119
075	1.973	2.070
076	2.113	2.299
077	2.213	2.292
078	2.182	2.358
079	1.883	1.995
080	2.146	2.322

Table S6. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	SOS1-PBE-DH-INVEST	
Molecule	$S_1 \leftarrow S_0$	$T_1 \leftarrow S_0$
081	1.999	2.093
082	1.985	2.037
083	1.903	2.006
084	2.147	2.238
085	1.993	2.079
086	1.580	1.511
087	2.110	2.219
088	1.643	1.608
089	2.016	2.134
090	1.662	1.623
091	1.957	1.996
092	1.745	1.844
093	1.982	2.111
094	2.039	2.070
095	2.191	2.318
096	2.319	2.517
097	2.285	2.476
098	2.644	2.332
099	2.718	2.511
100	2.238	2.257
101	2.579	2.365
102	1.727	1.728
103	1.179	1.204
104	1.289	1.323
105	1.683	1.827
106	1.576	1.716
107	1.882	1.910
108	1.764	1.867
109	2.065	2.210
110	1.965	2.076
111	1.827	1.909
112	2.475	2.644
113	2.310	2.518
114	1.418	1.339
115	1.425	1.645
116	1.672	1.829
117	1.592	1.835
118	1.726	1.714
119	2.287	2.184
120	2.332	2.242

Table S6. Vertical excitation energies (all in eV) calculated with the aug-cc-pVDZ basis set (cont.)

	SOS1-PBE-DH-INVEST	
Molecule	$S_1 \leftarrow S_0$	$T_1 \leftarrow S_0$
121	2.560	2.361
122	2.011	2.037
123	2.146	2.314
124	2.040	2.090
125	2.208	2.289
126	2.117	2.189
127	1.979	2.070
128	2.324	2.513
129	2.133	2.372
130	2.043	2.013
131	2.548	2.442
132	2.440	2.484
133	1.679	1.729
134	2.111	1.752
135	2.461	2.680
136	2.488	2.740
137	2.387	2.556
138	2.449	2.686
139	2.321	2.427
140	2.793	3.065
141	2.816	3.073
142	2.689	2.982
143	2.697	2.980
144	2.523	2.804
145	2.424	2.630
146	2.430	2.671
147	2.286	2.473
148	3.068	2.952
149	2.120	2.002
150	3.114	2.949
151	2.996	2.954
152	2.209	2.189
153	2.944	2.920
154	2.292	2.401
155	2.190	2.363
156	3.408	3.605
157	3.478	3.619
158	3.263	3.493
159	3.264	3.481

Technical details of the CPUs used:

vendor-id	:	AuthenticAMD
cpu family	:	23
model	:	49
model name	:	AMD EPYC 7742 64-Core Processor
microcode	:	0x830107a
cpu MHz	:	2673.182
cache size	:	512 KB
siblings	:	64
bogomips	:	4499.98
clflush size	:	64
cache-alignment	:	64

Example input file (main blocks) for PBE-DH-INVEST:

```
! RKS RIJCOSX aug-cc-pVDZ aug-cc-pVDZ/C TightSCF DEFGRID3

%method
Functional PBE
ScalHFX = 0.8434
ScalDFX = 0.1566
ScalGGAC = 0.4000
ScalLDAC = 0.4000
ScalMP2C = 0.6000
end

%mp2
Density relaxed
end

%tddft
Mode riints
Nroots 10
Maxdim 100
TDA true
Triplets true
end

%cis
Dcorr 1
end

*xyzfile 0 1 geom.xyz
```

Example input file (main blocks) for SOS1-PBE-DH-INVEST:

```
! RKS RIJCOSX aug-cc-pVDZ aug-cc-pVDZ/C TightSCF DEFGRID3

%method
Functional PBE
ScalHFX = 0.8434
ScalDFX = 0.1566
ScalGGAC = 0.4000
ScalLDAC = 0.4000
ScalMP2C = 1.0000
end

%mp2
Density relaxed
DoSCS true
pS 0.8000
pT 0.0000
end

%tddft
Mode riints
Nroots 10
Maxdim 100
TDA true
Triplets true
end

%cis
Dcorr 1
DoSCS true
SCSpar 0.0000,0.8000,0.0000,0.8000
end

*xyzfile 0 1 geom.xyz
```