Accelerating Kohn–Sham Response Theory using Density-fitting and the Auxiliary-Density-Matrix Method

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Here we included a more detalied account on the results presented in the paper. We present statistics, mean errors, standard deviations and maximum absolute errors, compared to aug-pcseg-4 reference calculations for three types of calculations: *full, df-J and admm*. Here *full* referes to a calculation combining J-engine for Coulomb and LinK for exchange (i.e. without any approximation), *df-J* to the combination of density-fitting for Coulomb and LinK for exchange, and *admm* to the combination of density-fitting for Coulomb and admm for exchange.

A. Statistical error analysis

We here report mean errors, standard deviations and maximum absolute errors in electronic ground-state energies, excitation energies, isotropic polarizabilties, anisotropic polarizabilties and dipole hyperpolarizabilty for the pcseg-n and aug-pcseg-n basis sets, $n = 1, 2, 3$ compared with aug-pcseg-4 full calculations.

TABLE S1. Errors in CAMB3LYP($\alpha = 0.21$, $\beta = 0.79$, $\mu = 0.45$) electronic ground-state energies per electron for pcseg-n and aug-pcseg-n calculations, with $n = 1, 2, 3$. Mean errors, standard deviations (in paranthesis) and maximum absolute errors (in square brackets) compared to aug-pcseg-4 reference calculations are given in μE$_H$/electron for the M11 benchmark (see article).

Calculation	pcseg-n		
type	1	2	3
full	4055(541)[4927]	458(58)[549]	17(2)[20]
df-J	4052(541)[4924]	456(58)[547]	14(2)[17]
admm	6055(740)[7445]	232(69)[323]	21(6)[31]
	aug-pcseg-n		
	1	2	3
full	3759(408)[4440]	426(50)[506]	15(1)[17]
df-J	3756(408)[4436]	423(50)[504]	11(2)[14]
admm	5627(587)[6816]	197(48)[270]	15(5)[22]

TABLE S2. Errors in CAMB3LYP($\alpha = 0.21$, $\beta = 0.79$, $\mu = 0.45$) excitation energies for pcseg-n and aug-pcseg-n calculations, with $n = 1, 2, 3$. Mean errors, standard deviations (in paranthesis) and maximum absolute errors (in square brackets) compared to aug-pcseg-4 reference calculations are given in μE_H for the M11 benchmark (see article).

Calculation	pcseg-n		
type	1	2	3
full	33042(17985)[77614]	16615(10872)[45147]	7830(6371)[25445]
df-J	33040(17985)[77608]	16614(10874)[45155]	7828(6370)[25433]
admm	35514(20382)[84003]	22813(13721)[54626]	10367(7884)[30670]
	aug-pcseg-n		
	1	2	3
full	1807(3347)[14138]	940(2277)[12124]	57(129)[720]
df-J	1798(3348)[14133]	931(2276)[12107]	48(130)[688]
admm	1254(4549)[16555]	1065(2403)[12081]	200(255)[1376]

TABLE S3. Errors in CAMB3LYP($\alpha = 0.21$, $\beta = 0.79$, $\mu = 0.45$) isotropic polarizabilties for for pcseg-n and aug-pcseg-n calculations, with $n = 1, 2, 3$. Mean errors, standard deviations (in paranthesis) and maximum absolute errors (in square brackets) compared to aug-pcseg-4 reference calculations are given in ma.u. (1 a.u. = 0.148 Å^3) for the M11 benchmark (see article).

Calculation	pcseg-n		
type	1	2	3
full	-6958(2697)[10209]	-2449(905)[3574]	-503(199)[730]
df-J	-6958(2697)[10208]	-2445(903)[3566]	-499(197)[722]
admm	-7323(2882)[10956]	-2983(1125)[4357]	-839(329)[1326]
	aug-pcseg-n		
	1	2	3
full	-314(118)[476]	-66(21)[86]	-3(1)[5]
df-J	-311(117)[472]	-62(20)[79]	1(3)[5]
admm	-1028(446)[1824]	-116(45)[196]	-12(5)[19]

TABLE S4. Errors in CAMB3LYP($\alpha = 0.21$, $\beta = 0.79$, $\mu = 0.45$)/aug-pcseg-n anisotropic polarizabilties calculations, with $n = 1, 2, 3$. Mean errors, standard deviations (in paranthesis) and maximum absolute errors (in square brackets) compared to aug-pcseg-4 reference calculations are given in ma.u. for the M11 benchmark (see article).

Calculation	pcseg-n		
type	1	2	3
full	3558(3356)[9287]	1137(1408)[4130]	247(337)[917]
df-J	3557(3355)[9285]	1131(1407)[4123]	242(338)[911]
admm	3932(3567)[10044]	1180(1517)[4332]	346(467)[1331]
	aug-pcseg-n		
	1	2	3
	-232(353)[829]	-4(31)[54]	-2(3)[7]
full	-239(361)[846]	-9(34)[57]	-8(9)[24]
df-J	378(674)[1612]	-145(165)[429]	-21(25)[71]
admm			

TABLE S5. Errors in the dipole hyperpolarizabilty components along the permanent dipole moment for CAMB3LYP($\alpha = 0.21$, $\beta = 0.79$, $\mu = 0.45$)/aug-pcseg-n calculations, with $n = 1, 2, 3$. Mean errors, standard deviations (in paranthesis) and maximum absolute errors (in square brackets) compared to aug-pcseg-4 reference calculations are given in ma.u. for the M11 benchmark (see article).

Calculation	pcseg-n		
type	1	2	3
full	-7222(19298)[40271]	-4605(12298)[24362]	-55(4163)[9198]
df-J	-7221(19299)[40275]	-4606(12298)[24372]	-57(4149)[9179]
admm	-5455(19408)[43173]	-5618(14669)[28304]	-1366(5665)[13059]
	aug-pcseg-n		
	1	2	3
full	-643(2224)[4359]	-130(420)[1086]	6(50)[105]
df-J	-649(2225)[4326]	-137(424)[1101]	-1(72)[125]
admm	-1104(3908)[9279]	-426(1885)[4713]	-117(147)[427]

I. RESULTS

A. DFT Energy — CAM-B3LYP (α=0.21,β=0.79,μ=0.45)

TABLE S7. acetamide —DFT energy

acetamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-208.926086	-209.053119	-209.068556	-208.939834	-209.054443	-209.068644	-209.069145
df-J	-208.926181	-209.053216	-209.068669	-208.939933	-209.054541	-209.068757	
admm	-208.858863	-209.059756	-209.068350	-208.878307	-209.061809	-209.068580	

TABLE S8. acetone —DFT energy

acetone	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-192.867909	-192.982829	-192.997147	-192.878943	-192.983978	-192.997216	-192.997680
df-J	-192.868006	-192.982930	-192.997269	-192.879040	-192.984081	-192.997340	
admm	-192.799158	-192.989330	-192.997125	-192.815460	-192.992013	-192.997250	

TABLE S9. butadiene —DFT energy

butadiene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-155.711977	-155.801185	-155.812402	-155.715510	-155.802067	-155.812492	-155.812871
df-J	-155.712056	-155.801267	-155.812500	-155.715591	-155.802150	-155.812591	
admm	-155.660347	-155.808565	-155.812398	-155.667754	-155.808830	-155.812590	

TABLE S10. cyclopropene —DFT energy

cyclopropene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-116.410382	-116.477794	-116.485929	-116.413542	-116.478201	-116.485967	-116.486262
df-J	-116.410446	-116.477863	-116.486009	-116.413608	-116.478271	-116.486048	
admm	-116.364198	-116.483130	-116.486028	-116.368202	-116.483217	-116.486045	

TABLE S11. formaldehyde —DFT energy

formaldehyde	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-114.344759	-114.414802	-114.423282	-114.352558	-114.415489	-114.423326	-114.423591
df-J	-114.344806	-114.414846	-114.423333	-114.352607	-114.415534	-114.423377	
admm	-114.304464	-114.418545	-114.423139	-114.314540	-114.419547	-114.423245	

TABLE S12. formamide —DFT energy

formamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-169.667817	-169.772484	-169.785010	-169.679894	-169.773567	-169.785088	-169.785491
df-J	-169.667889	-169.772554	-169.785087	-169.679970	-169.773637	-169.785165	
admm	-169.614519	-169.777744	-169.784751	-169.631217	-169.779006	-169.784970	

TABLE S13. furan —DFT energy

furan	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-229.666379	-229.799628	-229.815961	-229.675394	-229.800553	-229.816053	-229.816618
df-J	-229.666479	-229.799721	-229.816068	-229.675497	-229.800646	-229.816161	
admm	-229.598519	-229.807575	-229.815826	-229.611502	-229.808263	-229.815913	

TABLE S14. ethene —DFT energy

ethene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-78.446218	-78.492829	-78.498442	-78.447724	-78.493127	-78.498482	-78.498683
df-J	-78.446265	-78.492880	-78.498504	-78.447772	-78.493180	-78.498544	
admm	-78.417726	-78.496802	-78.498533	-78.421146	-78.496745	-78.498595	

TABLE S15. imidazole —DFT energy

imidazole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-225.863637	-225.992677	-226.008867	-225.874823	-225.993874	-226.008973	-226.009542
df-J	-225.863735	-225.992765	-226.008966	-225.874926	-225.993962	-226.009073	
admm	-225.796544	-226.000450	-226.008642	-225.811477	-226.001514	-226.008836	

TABLE S16. propanamide —DFT energy

propanamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-248.174980	-248.323419	-248.341646	-248.189631	-248.324882	-248.341745	-248.342347
df-J	-248.175100	-248.323538	-248.341786	-248.189755	-248.325002	-248.341886	
admm	-248.094601	-248.331897	-248.341502	-248.116117	-248.334305	-248.341713	

TABLE S17. pyrrole —DFT energy

pyrrole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-209.824221	-209.943548	-209.958508	-209.831543	-209.944591	-209.958620	-209.959143
df-J	-209.824317	-209.943638	-209.958611	-209.831642	-209.944681	-209.958724	
admm	-209.764887	-209.952293	-209.958401	-209.775938	-209.952304	-209.958557	

B. Excitation energies — CAM-B3LYP (α=0.21,β=0.79,μ=0.45)

TABLE S18: acetamide — Excitation-energy

acetamide	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.216452	0.214288	0.213046	0.213613	0.212750	0.212724	0.212726
	2	0.296755	0.278233	0.270437	0.266008	0.267172	0.267010	0.267013
	3	0.306463	0.293833	0.283983	0.279100	0.278970	0.278788	0.278795
	4	0.332757	0.303146	0.291274	0.287035	0.286172	0.285906	0.285898
	5	0.341918	0.324035	0.313872	0.308122	0.308790	0.308588	0.308587
df-J	1	0.216445	0.214281	0.213035	0.213602	0.212740	0.212713	
	2	0.296752	0.278245	0.270452	0.266004	0.267182	0.267021	
	3	0.306457	0.293833	0.283989	0.279094	0.278979	0.278797	

TABLE S18: acetamide — Excitation-energy

acetamide	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
	4	0.332755	0.303154	0.291278	0.287027	0.286172	0.285905	
	5	0.341903	0.324050	0.313905	0.308098	0.308815	0.308615	
admm	1	0.210793	0.215075	0.213172	0.208102	0.213084	0.212771	
	2	0.305207	0.283892	0.271813	0.264596	0.266967	0.267154	
	3	0.306166	0.298367	0.286906	0.276184	0.279036	0.278956	
	4	0.341583	0.313918	0.294308	0.285180	0.286361	0.286029	
	5	0.346947	0.330100	0.316248	0.307064	0.308758	0.308890	

TABLE S19: acetone — Excitation-energy

acetone	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.166468	0.166772	0.166596	0.167074	0.166524	0.166526	0.166537
	2	0.325477	0.293122	0.280859	0.273978	0.273929	0.273728	0.273735
	3	0.344403	0.333167	0.325765	0.312966	0.310895	0.309849	0.309657
	4	0.349387	0.343987	0.330876	0.314634	0.314018	0.313293	0.313111
	5	0.391204	0.356872	0.334903	0.315787	0.314209	0.313709	0.313590
df-J	1	0.166465	0.166767	0.166589	0.167070	0.166518	0.166520	
	2	0.325479	0.293129	0.280866	0.273969	0.273933	0.273733	
	3	0.344403	0.333168	0.325773	0.312948	0.310891	0.309846	
	4	0.349388	0.343986	0.330894	0.314613	0.314020	0.313309	
	5	0.391197	0.356887	0.334924	0.315777	0.314221	0.313711	
admm	1	0.161279	0.166254	0.166612	0.162036	0.166753	0.166575	
	2	0.335756	0.306279	0.284395	0.271077	0.274144	0.273873	
	3	0.343562	0.336850	0.328228	0.311626	0.311371	0.310109	
	4	0.346991	0.344936	0.337806	0.312253	0.314361	0.313731	
	5	0.397592	0.368216	0.342238	0.314548	0.315296	0.314046	

TABLE S20: butadiene — Excitation-energy

butadiene	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.228177	0.221834	0.219216	0.220008	0.218762	0.218628	0.218626

TABLE S20: butadiene — Excitation-energy

butadiene	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
	2	0.290876	0.267159	0.254729	0.242299	0.242295	0.242116	0.242091
	3	0.296379	0.272796	0.261494	0.253492	0.253762	0.253606	0.253590
	4	0.300141	0.284486	0.271889	0.261021	0.261259	0.261084	0.261017
	5	0.309020	0.287911	0.287038	0.288933	0.278681	0.275515	0.274795
df-J	1	0.228178	0.221828	0.219208	0.220009	0.218755	0.218620	
	2	0.290876	0.267149	0.254721	0.242311	0.242274	0.242094	
	3	0.296385	0.272792	0.261488	0.253496	0.253745	0.253585	
	4	0.300140	0.284474	0.271883	0.261026	0.261240	0.261063	
	5	0.309020	0.287906	0.287032	0.288928	0.278650	0.275483	
admm	1	0.228221	0.223464	0.220184	0.220442	0.219103	0.218642	
	2	0.287163	0.280633	0.257310	0.242455	0.242545	0.242191	
	3	0.304553	0.281065	0.264122	0.254290	0.253758	0.253710	
	4	0.311093	0.288656	0.274494	0.261855	0.261358	0.261184	
	5	0.314705	0.295527	0.287470	0.286632	0.270609	0.276170	

TABLE S21: cyclopropene — Excitation-energy

cyclopropene	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.247516	0.237900	0.235187	0.236517	0.235031	0.234771	0.234759
	2	0.252395	0.247514	0.246489	0.248094	0.246549	0.246343	0.246334
	3	0.282157	0.278604	0.272523	0.260931	0.260812	0.260661	0.260652
	4	0.326965	0.288458	0.277738	0.274841	0.274616	0.274339	0.274311
	5	0.330123	0.298487	0.283499	0.276429	0.275905	0.275702	0.275680
df-J	1	0.247531	0.237920	0.235208	0.236538	0.235051	0.234791	
	2	0.252396	0.247510	0.246483	0.248092	0.246541	0.246336	
	3	0.282172	0.278627	0.272515	0.260931	0.260798	0.260644	
	4	0.326957	0.288457	0.277763	0.274849	0.274594	0.274315	
	5	0.330120	0.298487	0.283491	0.276452	0.275910	0.275708	
admm	1	0.247098	0.240731	0.236166	0.236268	0.235355	0.234856	
	2	0.252208	0.248972	0.246901	0.247884	0.247031	0.246387	
	3	0.280902	0.279939	0.276066	0.260124	0.260920	0.260780	

TABLE S21: cyclopropene — Excitation-energy

cyclopropene	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
	4	0.332121	0.304430	0.278221	0.274857	0.274957	0.274511	
	5	0.345030	0.312175	0.287655	0.276138	0.276175	0.275893	

TABLE S22: formaldehyde — Excitation-energy

formaldehyde	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.144688	0.144550	0.144295	0.144760	0.144257	0.144223	0.144249
	2	0.331960	0.305329	0.292760	0.285139	0.285739	0.285637	0.285666
	3	0.336827	0.345332	0.325595	0.314557	0.313763	0.313248	0.313237
	4	0.354520	0.350531	0.337213	0.322335	0.321787	0.321195	0.321147
	5	0.387658	0.364781	0.342461	0.335267	0.330971	0.329445	0.329246
df-J	1	0.144686	0.144539	0.144282	0.144757	0.144245	0.144210	
	2	0.331962	0.305329	0.292753	0.285121	0.285726	0.285620	
	3	0.336833	0.345329	0.325572	0.314543	0.313736	0.313215	
	4	0.354523	0.350523	0.337193	0.322260	0.321743	0.321143	
	5	0.387661	0.364786	0.342456	0.335223	0.330921	0.329391	
admm	1	0.139412	0.144546	0.144317	0.139832	0.144559	0.144241	
	2	0.330257	0.316506	0.295693	0.282409	0.285651	0.285670	
	3	0.341192	0.347085	0.331791	0.312360	0.313724	0.313339	
	4	0.354032	0.366877	0.342427	0.322353	0.321727	0.321264	
	5	0.385464	0.378608	0.344582	0.336211	0.331435	0.329744	

TABLE S23: formamide — Excitation-energy

formamide	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.211734	0.209560	0.208317	0.208965	0.208036	0.207970	0.207974
	2	0.300732	0.282937	0.275382	0.270809	0.271911	0.271771	0.271772
	3	0.302582	0.290403	0.283352	0.280506	0.279927	0.279659	0.279645
	4	0.334789	0.317914	0.302991	0.295526	0.294849	0.294586	0.294568
	5	0.347358	0.318862	0.308545	0.303383	0.304042	0.303906	0.303909

TABLE S23: formamide — Excitation-energy

formamide	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
df-J	1	0.211724	0.209547	0.208298	0.208949	0.208019	0.207950	
	2	0.300729	0.282946	0.275390	0.270799	0.271916	0.271773	
	3	0.302576	0.290396	0.283345	0.280496	0.279922	0.279650	
	4	0.334779	0.317920	0.302984	0.295512	0.294840	0.294573	
	5	0.347354	0.318863	0.308552	0.303357	0.304041	0.303900	
admm	1	0.205309	0.210186	0.208420	0.203286	0.208421	0.207987	
	2	0.302285	0.288110	0.276502	0.269156	0.271787	0.271888	
	3	0.308919	0.293619	0.285201	0.278641	0.280148	0.279747	
	4	0.341094	0.324366	0.306852	0.294071	0.295003	0.294688	
	5	0.354471	0.329168	0.310830	0.302285	0.304069	0.304031	

TABLE S24: furan — Excitation-energy

furan	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.236388	0.228585	0.226101	0.227530	0.225830	0.225594	0.225568
	2	0.279748	0.264645	0.248417	0.238000	0.237214	0.236979	0.236964
	3	0.296444	0.284464	0.269618	0.258044	0.257521	0.257305	0.257234
	4	0.319313	0.300182	0.282317	0.268375	0.267122	0.266619	0.266448
	5	0.321701	0.312037	0.288169	0.278279	0.269692	0.267300	0.266890
df-J	1	0.236384	0.228577	0.226094	0.227528	0.225823	0.225587	
	2	0.279740	0.264648	0.248419	0.238013	0.237215	0.236984	
	3	0.296445	0.284466	0.269618	0.258044	0.257505	0.257289	
	4	0.319310	0.300193	0.282324	0.268377	0.267107	0.266610	
	5	0.321698	0.312044	0.288152	0.278268	0.269720	0.267325	
admm	1	0.238067	0.230637	0.226907	0.228825	0.226133	0.225601	
	2	0.279647	0.275301	0.252455	0.240131	0.237550	0.237117	
	3	0.308403	0.277859	0.272928	0.260544	0.257872	0.257438	
	4	0.320998	0.299981	0.286926	0.271719	0.267628	0.267014	
	5	0.336326	0.313669	0.295723	0.281295	0.270928	0.267549	

TABLE S25: ethene — Excitation-energy

ethene	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.291706	0.280430	0.277153	0.274469	0.274876	0.274696	0.274703
	2	0.313712	0.292720	0.283153	0.278415	0.276844	0.276630	0.276628
	3	0.321128	0.308131	0.305391	0.298163	0.298327	0.298098	0.298082
	4	0.341524	0.320659	0.309546	0.304844	0.305013	0.304684	0.304634
	5	0.358274	0.331697	0.316347	0.309718	0.307582	0.307415	0.307415
df-J	1	0.291717	0.280424	0.277149	0.274407	0.274831	0.274652	
	2	0.313719	0.292683	0.283123	0.278430	0.276841	0.276625	
	3	0.321116	0.308116	0.305378	0.298064	0.298293	0.298062	
	4	0.341527	0.320657	0.309537	0.304811	0.304969	0.304642	
	5	0.358282	0.331690	0.316337	0.309721	0.307570	0.307403	
admm	1	0.293480	0.283928	0.278335	0.274974	0.275199	0.274734	
	2	0.310294	0.303936	0.284722	0.279704	0.277303	0.276615	
	3	0.335336	0.309677	0.306711	0.300135	0.299096	0.298158	
	4	0.350406	0.328666	0.311715	0.306032	0.305807	0.304584	
	5	0.355722	0.342800	0.319262	0.307729	0.308257	0.307518	

TABLE S26: imidazole — Excitation-energy

imidazole	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.253099	0.244905	0.232655	0.226512	0.226961	0.226810	0.226811
	2	0.265207	0.245162	0.241714	0.242108	0.240773	0.240549	0.240533
	3	0.272462	0.262121	0.261208	0.254161	0.254251	0.254024	0.253971
	4	0.280655	0.272034	0.265826	0.263391	0.261043	0.260958	0.260953
	5	0.310388	0.280360	0.267920	0.265161	0.264496	0.264111	0.263997
df-J	1	0.253093	0.244895	0.232655	0.226518	0.226955	0.226807	
	2	0.265199	0.245165	0.241706	0.242109	0.240768	0.240543	
	3	0.272463	0.262111	0.261197	0.254157	0.254235	0.254009	
	4	0.280646	0.272022	0.265828	0.263384	0.261033	0.260947	
	5	0.310381	0.280365	0.267907	0.265171	0.264490	0.264113	
admm	1	0.254880	0.247215	0.235810	0.228505	0.226920	0.226920	
	2	0.259999	0.254799	0.242907	0.243454	0.240877	0.240563	

TABLE S26: imidazole — Excitation-energy

imidazole	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
	3	0.282426	0.262812	0.261467	0.256813	0.254479	0.254415	
	4	0.285725	0.274462	0.269145	0.257896	0.261664	0.260981	
	5	0.306267	0.294688	0.269379	0.268248	0.264889	0.264450	

TABLE S27: propanamide — Excitation-energy

propanamide	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.218351	0.215399	0.214139	0.214842	0.213894	0.213879	0.213882
	2	0.297166	0.277899	0.269747	0.265539	0.266727	0.266561	0.266565
	3	0.305859	0.293165	0.283701	0.279797	0.279519	0.279339	0.279345
	4	0.334989	0.303791	0.291605	0.287384	0.286630	0.286401	0.286398
	5	0.341451	0.322972	0.312551	0.306780	0.307464	0.307266	0.307266
df-J	1	0.218344	0.215392	0.214129	0.214832	0.213885	0.213869	
	2	0.297163	0.277908	0.269758	0.265535	0.266728	0.266564	
	3	0.305853	0.293163	0.283701	0.279791	0.279515	0.279336	
	4	0.334989	0.303795	0.291603	0.287378	0.286621	0.286392	
	5	0.341436	0.322980	0.312574	0.306757	0.307471	0.307278	
admm	1	0.213106	0.216419	0.214331	0.209810	0.214327	0.213936	
	2	0.305497	0.283709	0.271317	0.264110	0.266498	0.266691	
	3	0.305730	0.297800	0.286700	0.277209	0.279646	0.279474	
	4	0.343537	0.314725	0.294816	0.285538	0.286859	0.286518	
	5	0.346500	0.329086	0.315039	0.305724	0.307398	0.307538	

TABLE S28: pyrrole — Excitation-energy

pyrrole	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	1	0.245515	0.230068	0.217695	0.210802	0.210975	0.210843	0.210841
	2	0.256706	0.236542	0.249017	0.237502	0.237402	0.230621	0.230597
	3	0.288300	0.261686	0.253193	0.239857	0.239302	0.237261	0.237184
	4	0.307027	0.269808	0.258256	0.250451	0.251051	0.239022	0.238927
	5	0.308472	0.271287	0.273805	0.262042	0.258867	0.248835	0.248360

TABLE S28: pyrrole — Excitation-energy

pyrrole	State	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
df-J	1	0.245510	0.230070	0.217692	0.210805	0.210963	0.210836	
	2	0.256709	0.236532	0.249016	0.237495	0.237378	0.230618	
	3	0.288301	0.261686	0.253195	0.239861	0.239286	0.237240	
	4	0.307028	0.269811	0.258251	0.250450	0.251034	0.239009	
	5	0.308474	0.271288	0.273793	0.262029	0.258871	0.248849	
admm	1	0.247426	0.239270	0.220748	0.212206	0.211045	0.210894	
	2	0.268617	0.239763	0.251741	0.239280	0.237532	0.230658	
	3	0.269711	0.272100	0.257524	0.241991	0.239701	0.237317	
	4	0.300049	0.284120	0.261124	0.252315	0.251007	0.239186	
	5	0.312354	0.284653	0.279030	0.264915	0.259662	0.249391	

C. Isotropic polarizability — CAM-B3LYP (α=0.21,β=0.79,μ=0.45)

TABLE S29. acetamide — Isotropic Polarizability

acetamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	29.802693	34.859463	37.181450	37.453367	37.659947	37.725639	37.728816
df-J	29.803013	34.861625	37.183167	37.455265	37.662023	37.727656	
admm	29.465019	34.234052	36.815737	36.768085	37.629653	37.711756	

TABLE S30. acetone — Isotropic Polarizability

acetone	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	34.462989	38.748795	40.378179	40.260839	40.622055	40.685357	40.688462
df-J	34.463201	38.750681	40.379306	40.261529	40.623627	40.686730	
admm	34.268200	38.212588	40.118523	39.935070	40.564059	40.669446	

TABLE S31. butadiene — Isotropic Polarizability

butadiene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	46.405724	52.643320	55.132480	55.262647	55.662270	55.737100	55.739134
df-J	46.405326	52.649725	55.139236	55.267100	55.668514	55.743615	
admm	46.014852	51.866310	54.702070	54.446374	55.543550	55.726547	

TABLE S32. cyclopropene — Isotropic Polarizability

cyclopropene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	28.408885	32.141353	33.500044	33.491522	33.764208	33.836061	33.838580
df-J	28.409074	32.143834	33.502866	33.494020	33.767230	33.839148	
admm	28.060236	31.809425	33.244834	32.893599	33.726468	33.826624	

TABLE S33. formaldehyde — Isotropic Polarizability

formaldehyde	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	12.980648	15.380369	16.878770	17.081821	17.340241	17.401569	17.406570
df-J	12.980532	15.382000	16.880253	17.082767	17.341994	17.403373	
admm	12.839199	15.123231	16.675622	16.717464	17.305683	17.392128	

TABLE S34. formamide — Isotropic Polarizability

formamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	19.683294	23.806254	25.924228	26.456734	26.586850	26.644188	26.648753
df-J	19.683451	23.808507	25.926444	26.459542	26.589541	26.646933	
admm	19.363670	23.309076	25.613483	25.791152	26.558800	26.636055	

TABLE S35. furan — Isotropic Polarizability

furan	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	38.584391	44.475250	46.869637	47.088901	47.377612	47.460906	47.463931
df-J	38.584956	44.480907	46.876177	47.095408	47.384476	47.468003	
admm	37.966784	43.831396	46.406335	45.958802	47.319578	47.452685	

TABLE S36. ethene — Isotropic Polarizability

ethene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	21.334358	25.035257	26.916273	27.218190	27.446241	27.511194	27.511995
df-J	21.334052	25.039027	26.921174	27.221761	27.451258	27.516224	
admm	21.125983	24.628594	26.635806	26.503874	27.386287	27.503594	

TABLE S37. imidazole — Isotropic Polarizability

imidazole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	38.197294	44.339926	46.786254	47.105762	47.361325	47.422604	47.424837
df-J	38.198249	44.345607	46.792171	47.112476	47.367742	47.429148	
admm	37.506095	43.543170	46.299023	45.982428	47.295130	47.415252	

TABLE S38. propanamide — Isotropic Polarizability

propanamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	40.305875	46.209325	48.581237	48.655382	48.923047	48.995868	48.999084
df-J	40.306215	46.211696	48.582955	48.657101	48.925211	48.997877	
admm	39.923338	45.452739	48.199559	47.946398	48.886416	48.979793	

TABLE S39. pyrrole — Isotropic Polarizability

pyrrole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	42.767437	49.402845	52.246206	52.581051	52.896806	52.973324	52.976535
df-J	42.768067	49.410096	52.254191	52.589181	52.905131	52.981796	
admm	42.020032	48.619303	51.650051	51.152708	52.816570	52.965399	

D. Anisotropic polarizability — CAM-B3LYP (α=0.21,β=0.79,μ=0.45)

TABLE S40. acetamide — Anisotropic Polarizability

acetamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	13.419961	12.651303	12.713931	12.673582	12.666536	12.674291	12.675498
df-J	13.419588	12.647947	12.712177	12.671900	12.664420	12.672316	
admm	13.685889	12.573912	12.662813	12.967843	12.645385	12.661227	

TABLE S41. acetone — Anisotropic Polarizability

acetone	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	10.693481	11.178961	11.243468	11.114783	11.107189	11.117860	11.118941
df-J	10.693043	11.178043	11.243042	11.114754	11.107231	11.117773	
admm	10.808465	11.021630	11.197167	11.394136	11.073078	11.115532	

TABLE S42. butadiene — Anisotropic Polarizability

butadiene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	56.282301	51.125601	47.912697	46.588405	47.049789	46.994652	46.995531
df-J	56.280804	51.118037	47.906945	46.579159	47.042537	46.987175	
admm	57.039641	51.327319	48.326099	48.607239	46.809423	46.983496	

TABLE S43. cyclopropene — Anisotropic Polarizability

cyclopropene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	13.265360	11.444577	9.924664	9.460482	9.582459	9.565589	9.567414
df-J	13.265428	11.445158	9.924610	9.460351	9.582341	9.565025	
admm	13.577582	11.613056	10.150852	10.072391	9.499867	9.559119	

TABLE S44. formaldehyde — Anisotropic Polarizability

formaldehyde	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	9.908739	9.731496	8.507488	8.259425	8.048617	8.012154	8.008569
df-J	9.908500	9.729681	8.507160	8.259203	8.048109	8.011680	
admm	10.118221	9.614578	8.637029	9.226318	8.060417	8.009027	

TABLE S45. formamide — Anisotropic Polarizability

formamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	14.390150	13.409728	13.150210	12.964776	12.844560	12.843626	12.843126
df-J	14.389919	13.405701	13.148040	12.962328	12.841655	12.840867	
admm	14.685051	13.354852	13.161923	13.526632	12.842662	12.827256	

TABLE S46. furan — Anisotropic Polarizability

furan	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	27.562130	21.866259	20.862780	20.226148	20.901208	20.935612	20.941325
df-J	27.559964	21.850863	20.848514	20.208680	20.887081	20.920900	
admm	28.180006	22.055110	21.003350	20.702413	20.548119	20.879999	

TABLE S47. ethene — Anisotropic Polarizability

ethene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	20.089015	16.241834	13.546400	12.549648	12.714159	12.689724	12.692741
df-J	20.088245	16.237328	13.541052	12.540999	12.706891	12.682393	
admm	20.499878	16.532797	13.849297	14.002798	12.526970	12.696430	

TABLE S48. imidazole — Anisotropic Polarizability

imidazole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	26.152756	22.254395	21.985358	21.059585	21.846402	21.883360	21.888413
df-J	26.149662	22.238985	21.972488	21.042506	21.833151	21.869839	
admm	26.771575	22.168899	22.023731	21.461074	21.483066	21.831181	

TABLE S49. propanamide — Anisotropic Polarizability

propanamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	15.508142	15.124741	15.674421	15.671802	15.778492	15.812833	15.815265
df-J	15.507827	15.121868	15.672395	15.670402	15.776936	15.811177	
admm	15.694809	15.066560	15.560469	15.417176	15.750577	15.799502	

TABLE S50. pyrrole — Anisotropic Polarizability

pyrrole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	31.389085	24.577973	23.405825	22.606358	23.373792	23.406540	23.413282
df-J	31.386807	24.558718	23.387797	22.583957	23.356402	23.388965	
admm	32.076945	24.794861	23.542018	23.115646	22.984627	23.342168	

E. Beta parallel — CAM-B3LYP (α=0.21,β=0.79,μ=0.45)

TABLE S51. acetamide — Beta parallel

acetamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-36.672672	-32.041067	-24.010715	-24.443056	-22.486146	-22.120666	-22.092090
df-J	-36.670349	-32.041730	-24.004313	-24.441341	-22.494782	-22.126645	
admm	-33.677763	-34.900040	-26.040801	-24.378820	-23.089756	-22.447193	

TABLE S52. acetone — Beta parallel

acetone	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-57.054566	-53.488414	-38.324128	-29.569145	-29.308151	-29.129156	-29.126174
df-J	-57.058543	-53.498666	-38.304765	-29.524937	-29.284500	-29.104305	
admm	-54.448583	-57.055207	-42.185171	-29.396897	-29.407673	-29.383175	

TABLE S53. butadiene — Beta parallel

butadiene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-0.000130	-0.000056	0.000000	0.000000	0.000000	0.000000	0.000000
df-J	-0.000043	-0.000021	0.000000	0.000000	0.000000	0.000000	
admm	-0.000014	-0.000033	0.000000	0.000000	0.000000	0.000000	

TABLE S54. cyclopropene — Beta parallel

cyclopropene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	19.477633	1.934554	-13.225419	-23.126419	-21.099899	-20.788346	-20.793403
df-J	19.481859	1.918175	-13.291327	-23.237285	-21.216705	-20.904860	
admm	22.379399	7.510609	-11.016866	-19.795392	-23.455560	-20.888559	

TABLE S55. formaldehyde — Beta parallel

formaldehyde	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-65.092127	-59.418554	-47.625502	-45.937005	-42.131683	-41.510998	-41.577742
df-J	-65.090282	-59.439021	-47.619942	-45.903612	-42.106402	-41.483403	
admm	-62.801904	-59.212776	-50.415002	-50.856500	-42.976118	-41.710016	

TABLE S56. formamide — Beta parallel

formamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-35.073130	-33.716813	-27.217841	-31.749130	-29.128264	-28.147018	-28.042218
df-J	-35.070002	-33.688500	-27.218286	-31.773751	-29.143277	-28.163025	
admm	-30.527289	-34.826394	-29.277902	-33.024583	-29.493302	-28.468869	

TABLE S57. furan — Beta parallel

furan	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-17.297419	-17.687269	-23.361587	-23.979076	-27.129249	-27.860574	-27.851834
df-J	-17.293895	-17.682614	-23.360406	-24.006291	-27.153883	-27.885337	
admm	-15.187312	-16.536204	-22.459311	-20.087019	-23.138613	-27.981172	

TABLE S58. ethene — Beta parallel

ethene	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-0.000005	0.000000	-0.000005	0.000000	0.000000	0.000000	0.000000
df-J	-0.000037	-0.000000	0.000000	0.000000	0.000000	0.000000	
admm	-0.000012	0.000042	0.000000	0.000000	0.000000	0.000000	

TABLE S59. imidazole — Beta parallel

imidazole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-35.454719	-16.534850	2.125554	3.737302	1.702597	1.632315	1.562271
df-J	-35.451778	-16.532146	2.135928	3.740391	1.716556	1.644275	
admm	-33.607567	-21.992542	-2.559878	1.374820	2.108812	1.512979	

TABLE S60. propanamide — Beta parallel

propanamide	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-31.705695	-25.890451	-19.524724	-20.991407	-21.194861	-21.226517	-21.209861
df-J	-31.700404	-25.884689	-19.529172	-21.002276	-21.208285	-21.240083	
admm	-28.565217	-28.368504	-21.322300	-21.730513	-21.645606	-21.232088	

TABLE S61. pyrrole — Beta parallel

pyrrole	pcseg-1	pcseg-2	pcseg-3	aug-pcseg-1	aug-pcseg-2	aug-pcseg-3	aug-pcseg-4
full	-1.622995	7.757819	16.673577	14.511045	15.386112	15.396816	15.303400
df-J	-1.629149	7.748175	16.675586	14.534059	15.416108	15.428524	
admm	-2.856450	4.140626	15.057175	10.821016	12.161091	15.364307	

TABLE S6. Average component wall timings in seconds per SCF/response interation for the acetone molecule. Here reg-J refers to J-engine based Coulomb, df-J to density-fitted Coulomb, LinK to linear-scaling exchange, ADMM to ADMM exchange, XC to exchange-correation and solver to the matrix operations performed in either the SCF or response solver.

		aug-pcseg-2	aug-pcseg-3
SCF	reg-J	6.1	100.2
	df-J	0.2	0.9
	LinK	36.4	465.6
	ADMM	6.3	47.8
	XC	3.7	10.7
	solver	3.1	13.6
Excitation energy	reg-J	6.1	227.9
	df-J	0.8	3.2
	LinK	59.0	620.1
	ADMM[a]	23.0	148.5
	XC	14.8	42.8
	solver	5.9	30.3
Polarizabilities	reg-J	6.5	109.0
	df-J	0.2	0.9
	LinK	33.7	492.3
	ADMM	6.5	42.3
	XC	5.5	16.0
	solver	2.2	10.9
Hyperpolarizabilities	reg-J	6.6	109.0
	df-J	0.2	0.9
	LinK	33.8	499.2
	ADMM	6.5	42.4
	XC	5.5	16.0
	solver	2.2	10.7

[a] The ADMM excitation vectors have been calculated separately for each of the five exciation energies. Timings reported include the sum of all vectors