SI Tables

Table S1. Protonation states calculated using the Mn-depleted PSII structure (PDB codes 5MX2) [2].

residue	protonation [H^+]
ligand	
D1-Asp170	1.0
D1-Glu189	1.0
D1-His332	1.0
D1-Glu333	1.0
D1-Asp342	0.0 [a]
D1-Ala344 [b]	1.0
CP43-Glu354	1.0
H-bond partner	
D1-His337	1.0

[a] D1-Asp342 was ionized but it accepted H-bonds from protonated D1-Glu189 and protonated D1-His337.

[b] The carboxy terminus of the D1 protein.

Table S2. Atomic partial charges of Chl*a* and Pheo*a*. –, not applicable.

	Chl*a*$^{\bullet+}$	**Chl*a***	**Chl*a*$^{\bullet-}$**	**Pheo*a***	**Pheo*a*$^{\bullet-}$**
MG	0.878	0.821	0.747	–	–
CHA	0.086	0.071	0.081	0.109	0.081
CHB	-0.386	-0.365	-0.350	-0.337	-0.360
HHB	0.161	0.144	0.129	0.144	0.135
CHC	-0.255	-0.221	-0.247	-0.161	-0.205
HHC	0.146	0.138	0.131	0.127	0.118
CHD	-0.359	-0.280	-0.326	-0.220	-0.296
HHD	0.189	0.173	0.169	0.165	0.165
NA	-0.345	-0.311	-0.302	-0.241	-0.247
C1A	0.043	-0.008	-0.062	-0.081	-0.133
C2A	0.081	0.062	0.074	0.217	0.255
H2A	0.061	0.050	0.040	0.023	0.011
C3A	0.061	0.080	0.076	0.174	0.190
H3A	0.072	0.050	0.033	0.006	-0.021
C4A	0.318	0.233	0.180	0.153	0.100
CMA	-0.419	-0.457	-0.479	-0.418	-0.373
HMA1	0.128	0.126	0.123	0.105	0.082
HMA2	0.128	0.126	0.123	0.105	0.082
HMA3	0.128	0.126	0.123	0.105	0.082
CAA	-0.051	0.028	0.074	-0.037	0.025
HAA1	0.040	0.003	-0.024	0.040	0.019
HAA2	0.040	0.003	-0.024	0.040	0.019
CBA	-0.296	-0.302	-0.301	-0.548	-0.578
HBA1	0.069	0.073	0.077	0.144	0.136
HBA2	0.069	0.073	0.077	0.144	0.136
CGA	0.787	0.788	0.787	0.856	0.875
O1A	-0.512	-0.526	-0.538	-0.529	-0.536
O2A	-0.404	-0.407	-0.407	-0.413	-0.418
NB	-0.436	-0.371	-0.285	-0.115	-0.044
HNB	–	–	–	0.189	0.170
C1B	0.223	0.131	0.014	0.087	0.007
C2B	0.132	0.151	0.156	0.123	0.115
C3B	-0.071	-0.088	-0.156	-0.063	-0.130
C4B	0.248	0.135	0.086	0.039	-0.001
CMB	-0.376	-0.350	-0.278	-0.311	-0.248
HMB1	0.128	0.105	0.071	0.100	0.070
HMB2	0.128	0.105	0.071	0.100	0.070
HMB3	0.128	0.105	0.071	0.100	0.070
CAB	-0.077	-0.045	0.006	-0.051	0.000
HAB	0.122	0.105	0.087	0.112	0.096
CBB	-0.331	-0.391	-0.475	-0.387	-0.474
HBB1	0.174	0.163	0.147	0.164	0.148

HBB2	0.174	0.163	0.147	0.164	0.148
NC	-0.458	-0.381	-0.348	-0.292	-0.298
C1C	0.193	0.091	0.040	0.065	0.040
C2C	0.140	0.141	0.147	0.138	0.137
C3C	-0.221	-0.234	-0.229	-0.246	-0.248
C4C	0.360	0.221	0.183	0.201	0.195
CMC	-0.356	-0.328	-0.331	-0.329	-0.320
HMC1	0.125	0.100	0.087	0.100	0.085
HMC2	0.125	0.100	0.087	0.100	0.085
HMC3	0.125	0.100	0.087	0.100	0.085
CAC	0.062	0.143	0.206	0.157	0.210
HAC1	0.039	0.000	-0.036	-0.006	-0.036
HAC2	0.039	0.000	-0.036	-0.006	-0.036
CBC	-0.186	-0.197	-0.182	-0.178	-0.185
HBC1	0.064	0.053	0.036	0.046	0.037
HBC2	0.064	0.053	0.036	0.046	0.037
HBC3	0.064	0.053	0.036	0.046	0.037
ND	-0.470	-0.395	-0.339	-0.027	0.009
HND	–	–	–	0.091	0.072
C1D	0.288	0.143	0.094	0.005	-0.013
C2D	0.114	0.113	0.089	0.171	0.118
C3D	-0.196	-0.218	-0.269	-0.272	-0.301
C4D	0.188	0.124	0.088	0.037	0.028
CMD	-0.345	-0.300	-0.233	-0.320	-0.254
HMD1	0.129	0.098	0.061	0.105	0.071
HMD2	0.129	0.098	0.061	0.105	0.071
HMD3	0.129	0.098	0.061	0.105	0.071
CAD	0.629	0.627	0.618	0.661	0.641
OBD	-0.424	-0.473	-0.535	-0.481	-0.542
CBD	-0.624	-0.630	-0.653	-0.618	-0.642
HBD	0.217	0.207	0.197	0.213	0.207
CGD	0.840	0.859	0.870	0.763	0.786
O1D	-0.529	-0.536	-0.547	-0.508	-0.536
O2D	-0.379	-0.398	-0.407	-0.338	-0.342
CED	0.032	0.068	0.106	0.019	0.064
HED1	0.076	0.055	0.032	0.067	0.044
HED2	0.076	0.055	0.032	0.067	0.044
HED3	0.076	0.055	0.032	0.067	0.044
C1	0.083	0.094	0.109	0.103	0.102
H1	0.079	0.066	0.052	0.060	0.046
H2	0.079	0.066	0.052	0.060	0.046

Table S3. Atomic partial charges of BChl*a* and BPheo*a*. –, not applicable.

	BChl*a*$^{\bullet+}$	**BChl*a***	**BChl*a*$^{\bullet-}$**	**BPheo*a***	**BPheo*a*$^{\bullet-}$**
MG	0.872	0.816	0.763	–	–
CHA	0.100	0.089	0.097	0.150	0.117
CHB	-0.410	-0.378	-0.408	-0.400	-0.448
HHB	0.157	0.144	0.135	0.151	0.143
CHC	-0.444	-0.389	-0.429	-0.387	-0.426
HHC	0.201	0.180	0.172	0.180	0.170
CHD	-0.445	-0.404	-0.396	-0.364	-0.412
HHD	0.185	0.169	0.151	0.168	0.158
NA	-0.243	-0.203	-0.218	-0.230	-0.265
C1A	-0.025	-0.085	-0.130	-0.047	-0.056
C2A	-0.066	-0.043	-0.051	-0.051	-0.078
H2A	0.100	0.087	0.083	0.080	0.082
C3A	0.247	0.273	0.295	0.232	0.264
H3A	0.019	-0.007	-0.029	-0.009	-0.034
C4A	0.189	0.090	0.060	0.178	0.164
CMA	-0.429	-0.412	-0.401	-0.384	-0.367
HMA1	0.126	0.108	0.095	0.100	0.085
HMA2	0.126	0.108	0.095	0.100	0.085
HMA3	0.126	0.108	0.095	0.100	0.085
CAA	-0.042	-0.016	0.094	0.021	0.099
HAA1	0.069	0.058	0.025	0.044	0.023
HAA2	0.069	0.058	0.025	0.044	0.023
CBA	-0.410	-0.437	-0.488	-0.442	-0.497
HBA1	0.132	0.130	0.132	0.129	0.133
HBA2	0.132	0.130	0.132	0.129	0.133
CGA	0.755	0.735	0.721	0.743	0.738
O1A	-0.506	-0.479	-0.473	-0.486	-0.473
O2A	-0.379	-0.382	-0.379	-0.385	-0.389
NB	-0.524	-0.453	-0.419	-0.232	-0.200
HNB	–	–	–	0.209	0.205
C1B	0.299	0.193	0.152	0.161	0.129
C2B	0.164	0.174	0.140	0.161	0.116
C3B	-0.374	-0.432	-0.510	-0.428	-0.499
C4B	0.438	0.350	0.355	0.298	0.292
CMB	-0.352	-0.319	-0.253	-0.299	-0.222
HMB1	0.122	0.096	0.063	0.095	0.059
HMB2	0.122	0.096	0.063	0.095	0.059
HMB3	0.122	0.096	0.063	0.095	0.059
CAB	0.686	0.715	0.735	0.726	0.741
CBB	-0.509	-0.518	-0.524	-0.532	-0.535
HBB1	0.150	0.136	0.121	0.141	0.124
HBB2	0.150	0.136	0.121	0.141	0.124

HBB3	0.150	0.136	0.121	0.141	0.124
OBB	-0.469	-0.508	-0.563	-0.504	-0.561
NC	-0.307	-0.235	-0.244	-0.213	-0.243
C1C	0.190	0.058	0.035	0.102	0.071
C2C	0.198	0.282	0.293	0.265	0.314
H2C	0.026	-0.010	-0.017	-0.008	-0.029
C3C	-0.033	-0.016	-0.055	-0.073	-0.086
H3C	0.064	0.037	0.034	0.046	0.030
C4C	0.232	0.116	0.081	0.148	0.141
CMC	-0.303	-0.295	-0.291	-0.297	-0.278
HMC1	0.090	0.072	0.062	0.073	0.056
HMC2	0.090	0.072	0.062	0.073	0.056
HMC3	0.090	0.072	0.062	0.073	0.056
CAC	-0.028	-0.015	0.006	0.020	0.032
HAC1	0.036	0.025	0.023	0.016	0.012
HAC2	0.036	0.025	0.023	0.016	0.012
CBC	-0.185	-0.180	-0.196	-0.190	-0.190
HBC1	0.063	0.047	0.040	0.049	0.036
HBC2	0.063	0.047	0.040	0.049	0.036
HBC3	0.063	0.047	0.040	0.049	0.036
ND	-0.513	-0.462	-0.403	-0.092	-0.060
HND	–	–	–	0.104	0.089
C1D	0.372	0.261	0.177	0.140	0.108
C2D	0.077	0.080	0.062	0.090	0.053
C3D	-0.181	-0.242	-0.298	-0.233	-0.300
C4D	0.171	0.136	0.116	0.017	0.026
CMD	-0.311	-0.288	-0.228	-0.271	-0.204
HMD1	0.122	0.097	0.062	0.095	0.060
HMD2	0.122	0.097	0.062	0.095	0.060
HMD3	0.122	0.097	0.062	0.095	0.060
CAD	0.612	0.651	0.666	0.667	0.687
OBD	-0.425	-0.481	-0.543	-0.479	-0.544
CBD	-0.589	-0.600	-0.680	-0.636	-0.679
HBD	0.213	0.188	0.196	0.198	0.191
CGD	0.832	0.823	0.852	0.839	0.844
O1D	-0.529	-0.525	-0.538	-0.528	-0.536
O2D	-0.350	-0.370	-0.386	-0.365	-0.381
CED	-0.006	0.063	0.080	0.029	0.068
HED1	0.081	0.051	0.037	0.060	0.040
HED2	0.081	0.051	0.037	0.060	0.040
HED3	0.081	0.051	0.037	0.060	0.040
C1	0.092	0.081	0.055	0.081	0.076
H1A	0.080	0.073	0.072	0.072	0.064
H1B	0.080	0.073	0.072	0.072	0.064

Table S4. Atomic partial charges of ubiquinone (UQ).

	UQ	$UQ^{\bullet-}$
C1	-0.008	-0.039
C1M	-0.240	-0.090
H1MA	0.090	0.018
H1MB	0.090	0.018
H1MC	0.090	0.018
C2	0.430	0.228
O2	-0.401	-0.476
C3	0.115	0.078
O3	-0.237	-0.312
C3M	0.027	0.104
H3MA	0.059	0.010
H3MB	0.059	0.010
H3MC	0.059	0.010
C4	-0.065	0.032
O4	-0.259	-0.310
C4M	0.051	0.131
H4MA	0.048	0.002
H4MB	0.048	0.002
H4MC	0.048	0.002
C5	0.477	0.297
O5	-0.350	-0.464
C6	-0.254	-0.253
C7	0.145	0.254
H7A	0.028	-0.031
H7B	0.028	-0.031
C8	-0.377	-0.373
H8	0.144	0.125
C9	0.249	0.224
C10	-0.314	-0.290
H10A	0.089	0.066
H10B	0.089	0.066
H10C	0.089	0.066
C11	-0.225	-0.224
H11A	0.089	0.066
H11B	0.089	0.066

Table S5. Atomic partial charges of plastoquinone (PLQ).

	PLQ	**PLQ$^{\bullet-}$**
C2	-0.352	-0.402
C3	-0.082	-0.171
C4	0.480	0.324
C5	-0.067	-0.084
C6	-0.103	-0.122
C1	0.558	0.442
C7	0.264	0.448
C8	-0.467	-0.487
C9	0.294	0.243
C10	-0.373	-0.336
C11	-0.268	-0.254
C52	-0.130	-0.099
C53	-0.152	-0.053
O1	-0.437	-0.557
O2	-0.427	-0.575
H2	0.160	0.139
H7A	0.007	-0.061
H7B	0.007	-0.061
H8	0.163	0.165
H10A	0.106	0.081
H10B	0.106	0.081
H10C	0.106	0.081
H11A	0.104	0.081
H11B	0.104	0.081
H52A	0.065	0.023
H52B	0.065	0.023
H52C	0.065	0.023
H53A	0.068	0.009
H53B	0.068	0.009
H53C	0.068	0.009

Table S6. Atomic partial charges of spheroidene.

	spheroidene
CM1	0.145
HM1A	0.015
HM1B	0.015
HM1C	0.015
O1	-0.462
C1	0.655
C2	-0.407
H2A	0.093
H2B	0.093
H2C	0.093
C3	-0.407
H3A	0.093
H3B	0.093
H3C	0.093
C4	-0.314
H4A	0.089
H4B	0.089
C5	0.014
H5	0.095
C6	-0.330
H6	0.137
C7	0.221
C8	-0.265
H8A	0.078
H8B	0.078
H8C	0.078
C9	-0.286
H9	0.136
C10	-0.006
H10	0.116
C11	-0.283
H11	0.141
C12	0.185
C13	-0.245
H13A	0.075
H13B	0.075
H13C	0.075
C14	-0.254
H14	0.124
C15	-0.045
H15	0.132

C16	-0.250
H16	0.149
C17	0.209
C18	-0.336
H18A	0.095
H18B	0.095
H18C	0.095
C19	-0.259
H19	0.130
C20	-0.102
H20	0.121
C21	-0.078
H21	0.125
C22	-0.264
H22	0.127
C23	0.214
C24	-0.243
H24A	0.073
H24B	0.073
H24C	0.073
C25	-0.317
H25	0.141
C26	0.014
H26	0.122
C27	-0.285
H27	0.144
C28	0.061
C29	-0.255
H29A	0.083
H29B	0.083
H29C	0.083
C30	-0.114
H30A	0.050
H30B	0.050
C31	0.215
H31A	0.000
H31B	0.000
C32	-0.399
H32	0.135
C33	0.099
C34	-0.286
H34A	0.088
H34B	0.088
H34C	0.088
C35	-0.132

H35A	0.051
H35B	0.051
C36	0.244
H36A	0.000
H36B	0.000
C37	-0.518
H37	0.159
C38	0.309
C39	-0.368
H39A	0.100
H39B	0.100
H39C	0.100
C40	-0.368
H40A	0.100
H40B	0.100
H40C	0.100

Table S7. Atomic partial charges of sulfoquinovosyl diacylglycerol (SQD).

	SQD$^-$
O6	-0.398
C44	0.077
C45	0.235
C46	0.152
O47	-0.359
C7	0.735
O49	-0.540
C8	-0.302
O48	-0.388
C23	0.788
O10	-0.546
C24	-0.358
C1	0.428
C2	0.114
O2	-0.601
C3	0.202
O3	-0.628
C4	0.041
O4	-0.606
C5	0.312
C6	-0.221
O5	-0.433
S	1.004
O7	-0.610
O8	-0.610
O9	-0.610
H44A	0.062
H44B	0.062
H45	0.020
H46A	0.000
H46B	0.000
H8A	0.117
H8B	0.117
H24A	0.136
H24B	0.136
H1	0.003
H2	0.062
HO	0.398
H3	0.022
HO3	0.412
H4	0.036

HO4	0.399
H5	0.018
H6A	0.061
H6B	0.061

Table S8. Atomic partial charges of heptyl 1-thiohexopyranoside (HTG).

	HTG
C1	0.039
S1	-0.219
C2	0.274
O2	-0.609
C3	0.072
O3	-0.618
C4	0.145
O4	-0.601
C5	0.361
O5	-0.415
C6	0.064
O6	-0.600
C1'	0.008
C2'	-0.015
C3'	0.098
C4'	-0.175
C5'	-0.047
C6'	0.139
C7'	-0.275
H1	0.112
H2	0.062
HO	0.416
H3	0.054
HO3	0.426
H4	0.042
HO4	0.393
H5	-0.038
H6A	0.036
H6B	0.036
HO6	0.410
H1'1	0.062
H1'2	0.062
H2'1	0.002
H2'2	0.002
H3'1	-0.002
H3'2	-0.002
H4'1	0.044
H4'2	0.044
H5'1	0.021
H5'2	0.021
H6'1	-0.009

H6'2	-0.009
H7'1	0.063
H7'2	0.063
H7'3	0.063

Table S9. Atomic partial charges of the Fe complex in PbRC.

Fe complex (PbRC)	
1FE	0.746
Glu-M234	
2CB	-0.081
2HB1	0.036
2HB2	0.036
2CG	0.034
2HG1	0.005
2HG2	0.005
2CD	0.633
2OE1	-0.635
2OE2	-0.635
His-L190	
3CB	-0.258
3HB1	0.126
3HB2	0.126
3CG	0.268
3ND1	-0.300
3HD1	0.340
3CD2	-0.254
3HD2	0.168
3CE1	0.038
3HE1	0.156
3NE2	-0.197
His-M219	
4CB	-0.258
4HB1	0.126
4HB2	0.126
4CG	0.268
4ND1	-0.300
4HD1	0.340
4CD2	-0.254
4HD2	0.168
4CE1	0.038
4HE1	0.156
4NE2	-0.197
His-M266	
5CB	-0.258
5HB1	0.127
5HB2	0.127
5CG	0.268
5ND1	-0.300

5HD1	0.340
5CD2	-0.254
5HD2	0.168
5CE1	0.038
5HE1	0.156
5NE2	-0.197
His-L190	
6CB	-0.258
6HB1	0.127
6HB2	0.127
6CG	0.268
6ND1	-0.300
6HD1	0.340
6CD2	-0.254
6HD2	0.168
6CE1	0.038
6HE1	0.156
6NE2	-0.197

Table S10. Atomic partial charges of the Fe complex in PSII.

Fe complex (PSII)	
1FE	0.695
HCO_3^-	
2C	0.847
2O1	-0.534
2O2	-0.635
2O3	-0.635
2HO	0.385
D2-His214	
3CB	-0.259
3CG	0.266
3ND1	-0.288
3CD2	-0.262
3CE1	0.037
3NE2	-0.182
3HB1	0.131
3HB2	0.131
3HD1	0.342
3HD2	0.174
3HE1	0.153
D1-His215	
4CB	-0.271
4CG	0.328
4ND1	-0.330
4CD2	-0.356
4CE1	0.009
4NE2	-0.090
4HB1	0.130
4HB2	0.130
4HD1	0.347
4HD2	0.188
4HE1	0.181
D1-His272	
5CB	-0.258
5CG	0.209
5ND1	-0.232
5CD2	-0.176
5CE1	-0.029
5NE2	-0.239
5HB1	0.128
5HB2	0.128
5HD1	0.324

5HD2	0.151
5HE1	0.178
D2-His268	
6CB	-0.270
6CG	0.273
6ND1	-0.336
6CD2	-0.229
6CE1	0.114
6NE2	-0.252
6HB1	0.130
6HB2	0.130
6HD1	0.346
6HD2	0.159
6HE1	0.119

Table S11. Atomic partial charges of the Mn_4CaO_5 cluster in S_1.

Mn_4CaO_5 cluster	
1Mn1	1.398
1Mn2	1.768
1Mn3	1.904
1Mn4	1.549
1Ca1	1.743
1O1	-1.010
1O2	-1.148
1O3	-0.979
1O4	-1.014
1O5	-1.071
water ligand	
1OH1	-0.935
1H11	0.482
1H12	0.450
1OH2	-0.958
1H21	0.444
1H22	0.511
1OH3	-0.862
1H31	0.490
1H32	0.496
1OH4	-0.921
1H41	0.458
1H42	0.536
D1-Asp170	
2CB	-0.307
2CG	0.932
2OD1	-0.839
2OD2	-0.795
2HB1	0.091
2HB2	0.091
D1-Glu189	
3CB	-0.292
3CG	-0.041
3CD	0.757
3OE1	-0.696
3OE2	-0.715
3HB1	0.085
3HB2	0.085
3HG1	0.081
3HG2	0.081
D1-His332	

4ND1	-0.294
4CG	0.400
4CB	-0.515
4NE2	-0.165
4CD2	-0.294
4CE1	0.029
4HD1	0.435
4HB1	0.213
4HB2	0.213
4HD2	0.174
4HE1	0.201
D1-Glu333	
5CB	-0.084
5CG	-0.076
5CD	0.784
5OE1	-0.658
5OE2	-0.652
5HB1	0.064
5HB2	0.064
5HG1	0.045
5HG2	0.045
D1-Asp342	
6CB	-0.416
6CG	0.927
6OD1	-0.737
6OD2	-0.780
6HB1	0.192
6HB2	0.192
D1-Ala344	
7N	-0.310
7HN	0.310
7CA	-0.016
7C	0.658
7OT1	-0.634
7CB	-0.313
7OT2	-0.680
7HA	0.054
7HB1	0.099
7HB2	0.099
7HB3	0.099
CP43-Glu354	
8CB	-0.132
8CG	-0.185
8CD	0.821
8OE1	-0.645

8OE2	-0.748
8HB1	0.056
8HB2	0.056
8HG1	0.113
8HG2	0.113
CP43-Arg357	
9CB	-0.238
9CG	-0.033
9CD	0.306
9NE	-0.709
9CZ	0.911
9NH1	-0.838
9NH2	-1.007
9HB1	0.095
9HB2	0.095
9HG1	0.041
9HG2	0.041
9HD1	-0.003
9HD2	-0.003
9HE	0.419
9HH11	0.443
9HH12	0.443
9HH21	0.533
9HH22	0.533

SI Figure legends

Figure S1. E_m for one-electron reduction calculated using a) the cyanobacterial PSI crystal structure (PDB code 1JB0) [3] and b) the plant PSI crystal structure (PDB code 4XK8) [4] in mV.

Figure S2. E_m for one-electron reduction calculated using the crystal structures of PbRC from a) *Rhodobacter sphaeroides* (PDB codes 1M3X) [5] and b) *Thermochromatium tepidum* (PDB codes 1EYS) [6] in mV.

Figure S3. (a) The binding interface of cytochrome c_2 with PbRC [7] and (b) the corresponding region in D1 (yellow) and D2 (blue) of PSII.

Figure S4. E_m for one-electron reduction calculated using a) the 1.87 Å-structure (PDB codes 2J8C) [8] and b) the refined 1.87 Å-structure, where the methyl-keto O and C atoms in P_M, B_L, B_M, H_L, and H_M in the original 1.87 Å-structure were swapped.

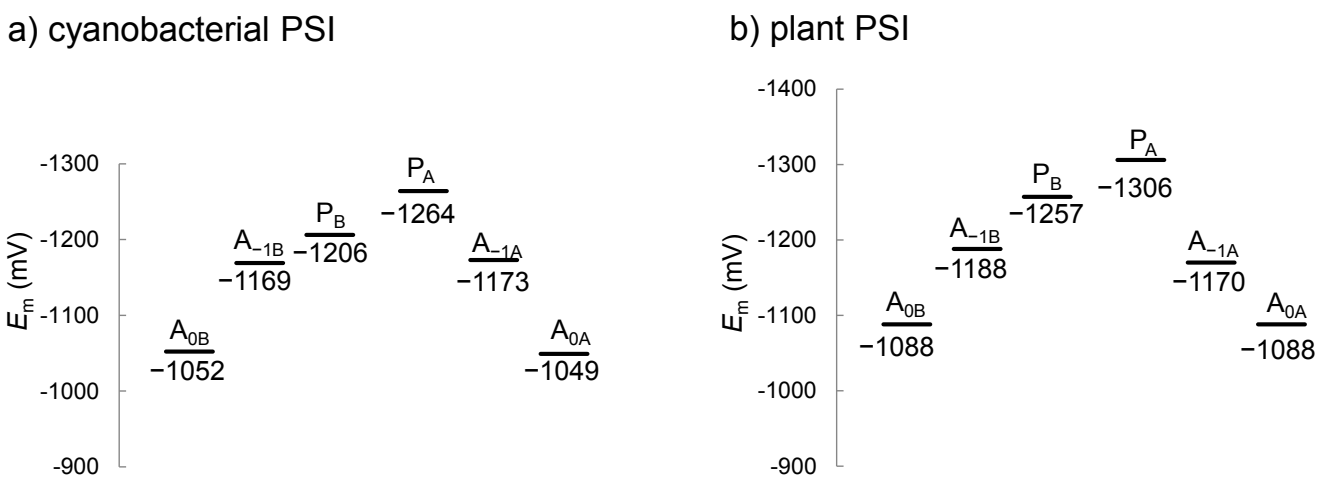
Figure S5. Localization of hydrophobic residues (white sphere) and cofactors, spheroidene (red sphere) and Q_B (cyan sphere), at the B_M binding moiety. Hydrophobic residues shown are Trp-M66, Phe-M67, Phe-M68, Phe-M74, Trp-M75, Phe-M85, Phe-M105, Trp-M115, Phe-M120, Phe-M123, Trp-M157, Phe-M162, Trp-M171, Pro-M176, Tyr-M177, and His-M182.

Figure S6. a) Deletion of the Q_B isoprene side-chain from C_{56} to C_{16} in the 2.01 Å-PbRC crystal structure (PDB code 3I4D) and b) changes in E_m for one-electron reduction (red arrow) in mV. Note that the Q_B isoprene side-chain is comprised of C_{16} in the 1.87 Å-PbRC crystal structure (PDB code 2J8C).

Figure S7. Loop ***a-b*** and helix ***cd*** of D2 in PSII (green) and subunit M in PbRC (cyan). The region between D2-Val55 and Ser66 in PSII (red dotted circle), where D2-His61 [9] is also located, is structurally absent in PbRC.

Figure S8. E_m for one-electron reduction calculated using the Mn-depleted PSII crystal structure (PDB codes 5MX2) [2] in mV.

Figure S9. The amino acid sequences of the L and M subunits in PbRC and the D1 and D2 subunits in PSII. Asp-M184, D1-Asn181/D2-Arg180, and Phe-L181/Tyr-M210 are in bold.



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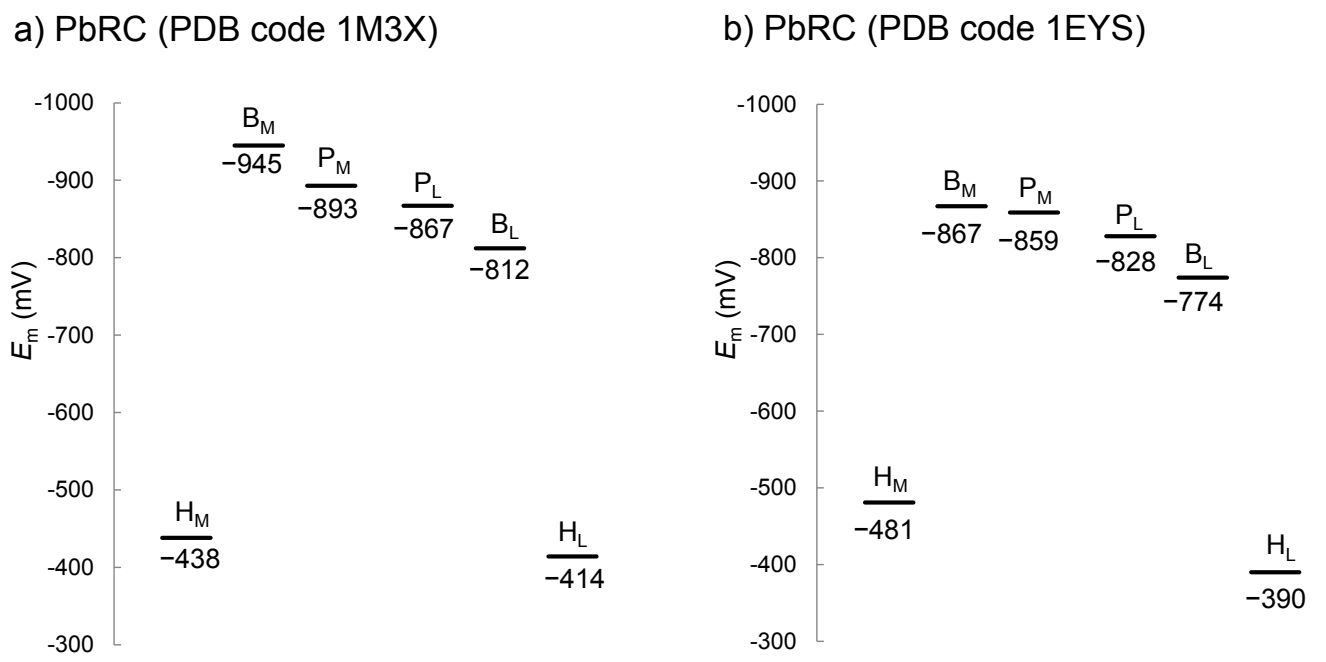

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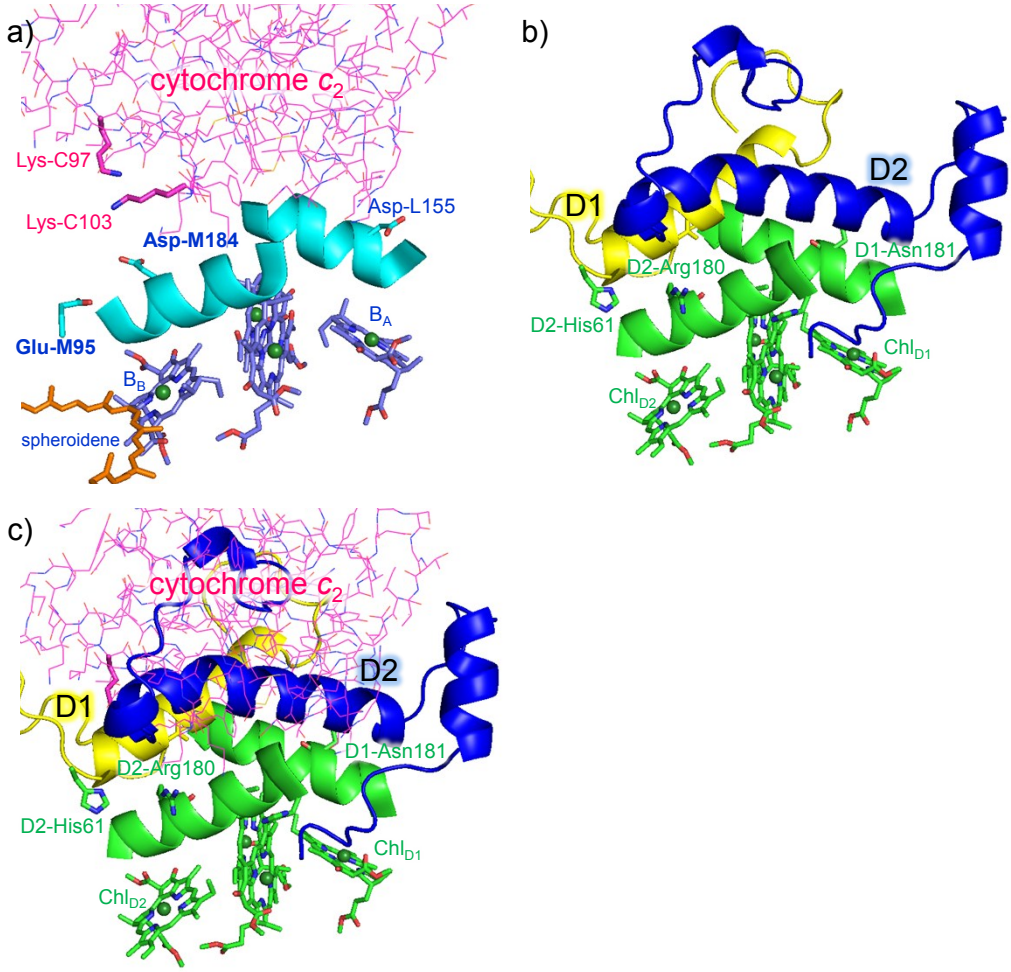
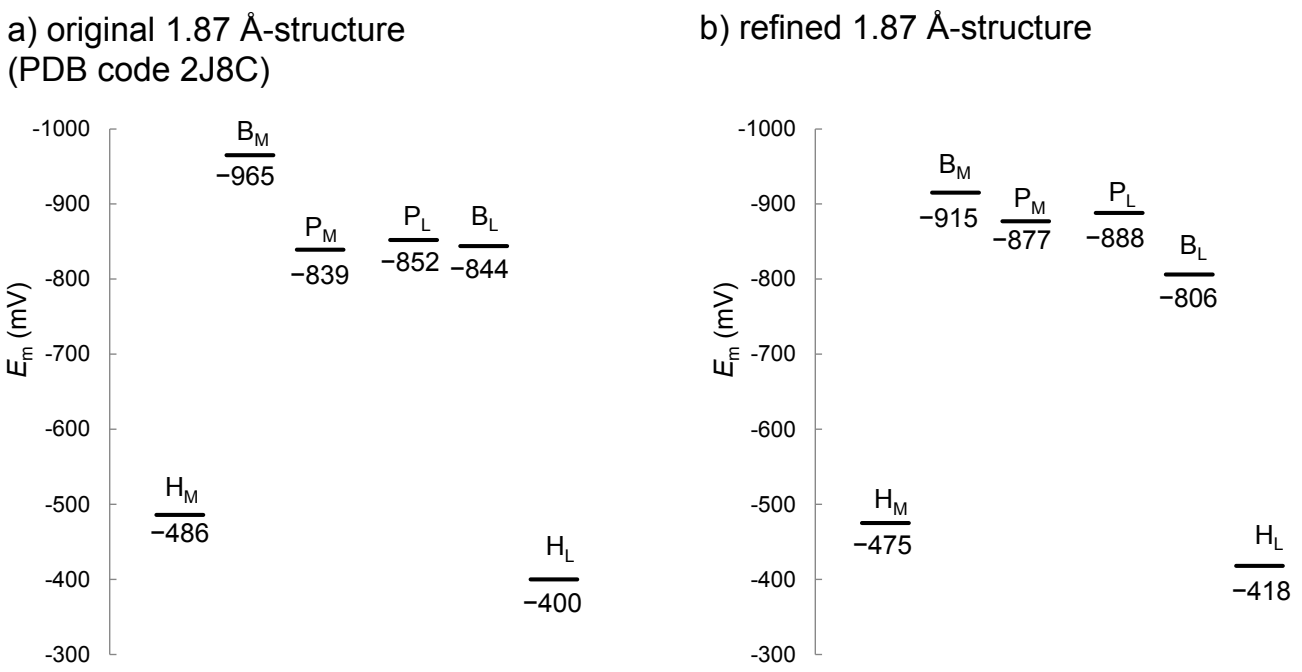


Figure S3. (a) The binding interface of cytochrome c_2 with PbRC (cyan) [7], (b) the corresponding region in D1 (yellow) and D2 (blue) (where cytochrome c_2 would exist) in PSII (green), and (c) comparison between PbRC (cytochrome c_2) and PSII (D1 and D2 proteins).



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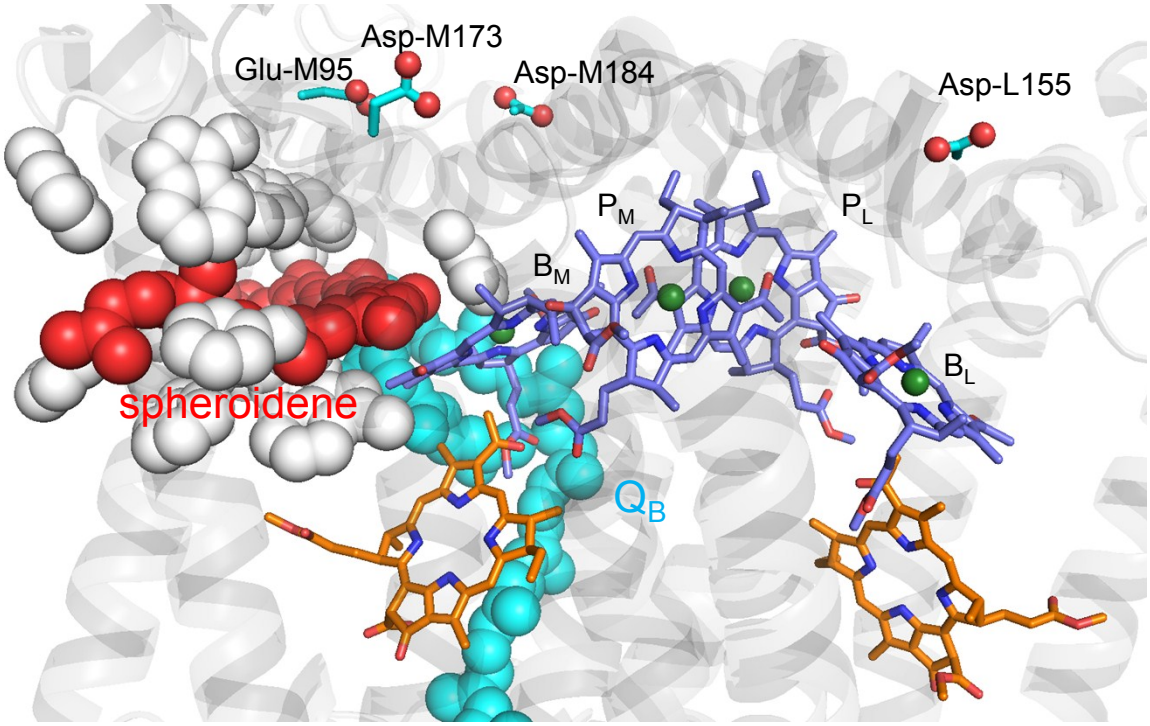

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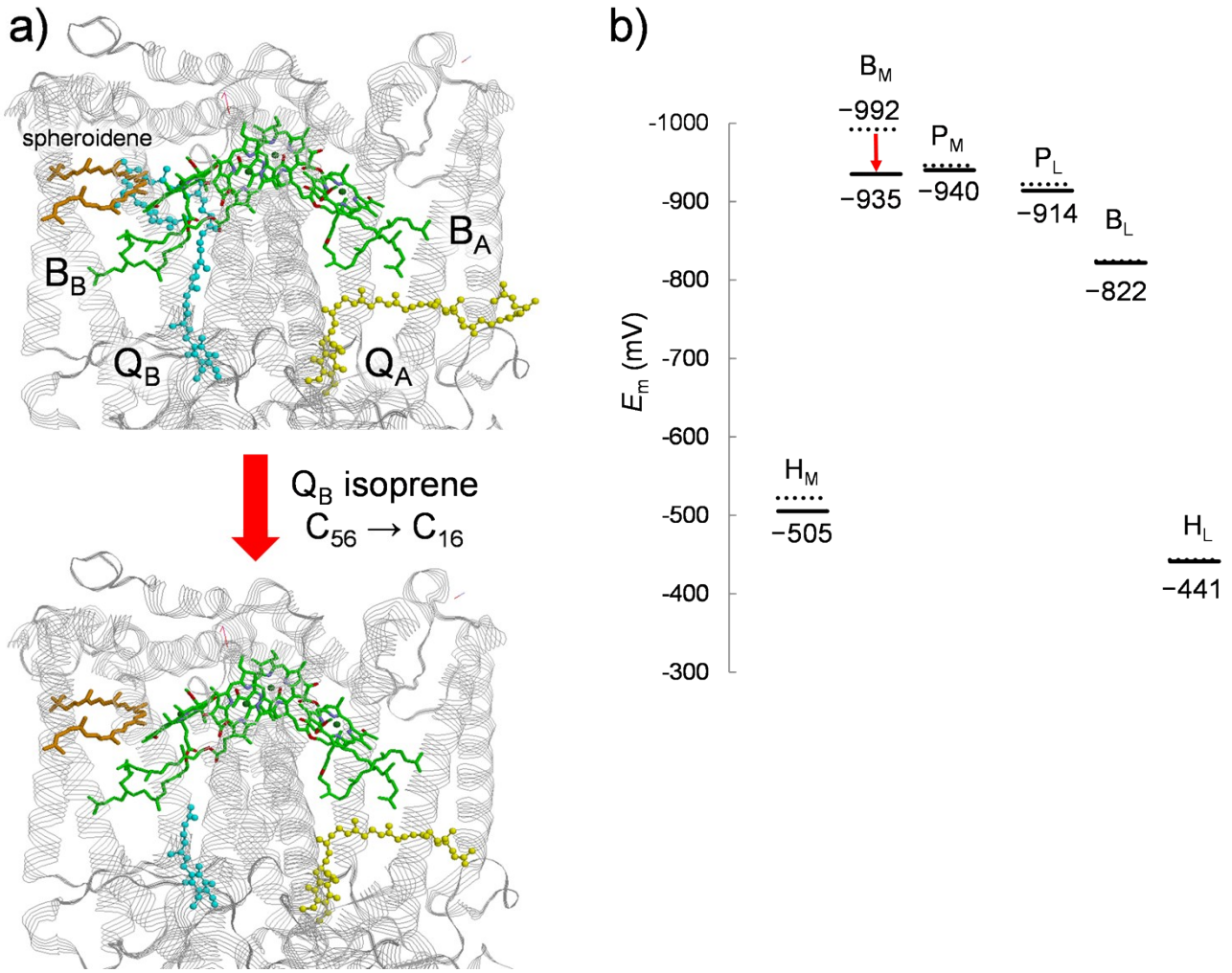

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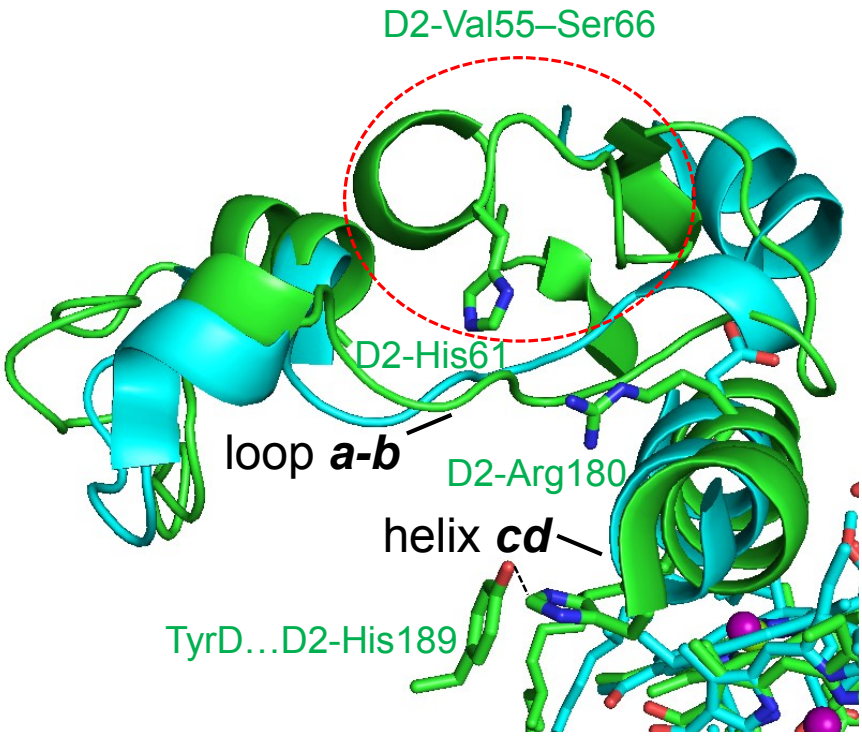

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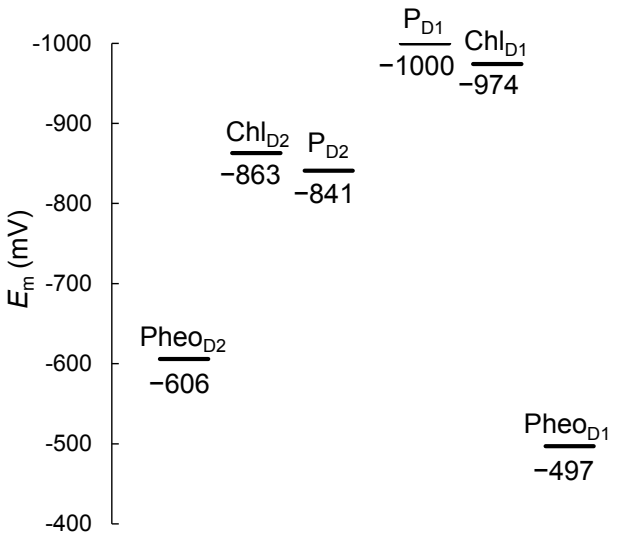

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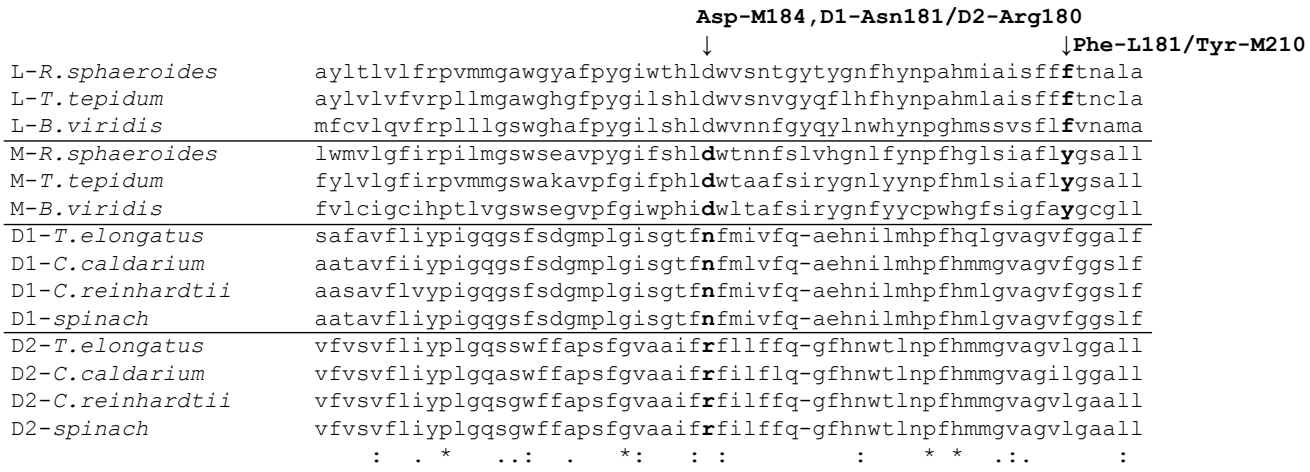

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SI References

1. Y. Umena, K. Kawakami, J.-R. Shen and N. Kamiya, *Nature*, 2011, **473**, 55-60.
2. M. Zhang, M. Bommer, R. Chatterjee, R. Hussein, J. Yano, H. Dau, J. Kern, H. Dobbek and A. Zouni, *eLife*, 2017, **6**, e26933.
3. P. Jordan, P. Fromme, H. T. Witt, O. Klukas, W. Saenger and N. Krauss, *Nature*, 2001, **411**, 909-917.
4. X. Qin, M. Suga, T. Kuang and J. R. Shen, *Science*, 2015, **348**, 989-995.
5. A. Camara-Artigas, D. Brune and J. P. Allen, *Proc. Natl. Acad. Sci. USA*, 2002, **99**, 11055-11060.
6. T. Nogi, I. Fathir, M. Kobayashi, T. Nozawa and K. Miki, *Proc Natl Acad Sci U S A*, 2000, **97**, 13561-13566.
7. H. L. Axelrod, E. C. Abresch, M. Y. Okamura, A. P. Yeh, D. C. Rees and G. Feher, *J. Mol. Biol.*, 2002, **319**, 501-515.
8. J. Koepke, E. M. Krammer, A. R. Klingen, P. Sebban, G. M. Ullmann and G. Fritzsch, *J Mol Biol*, 2007, **371**, 396-409.
9. K. Saito, A. W. Rutherford and H. Ishikita, *Proc Natl Acad Sci U S A*, 2013, **110**, 7690-7695.