Supplement for "A Modified Resonance-Theoretic Framework for Structure-Property Relationships in a Halochromic Oxonol Dye"

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Contents

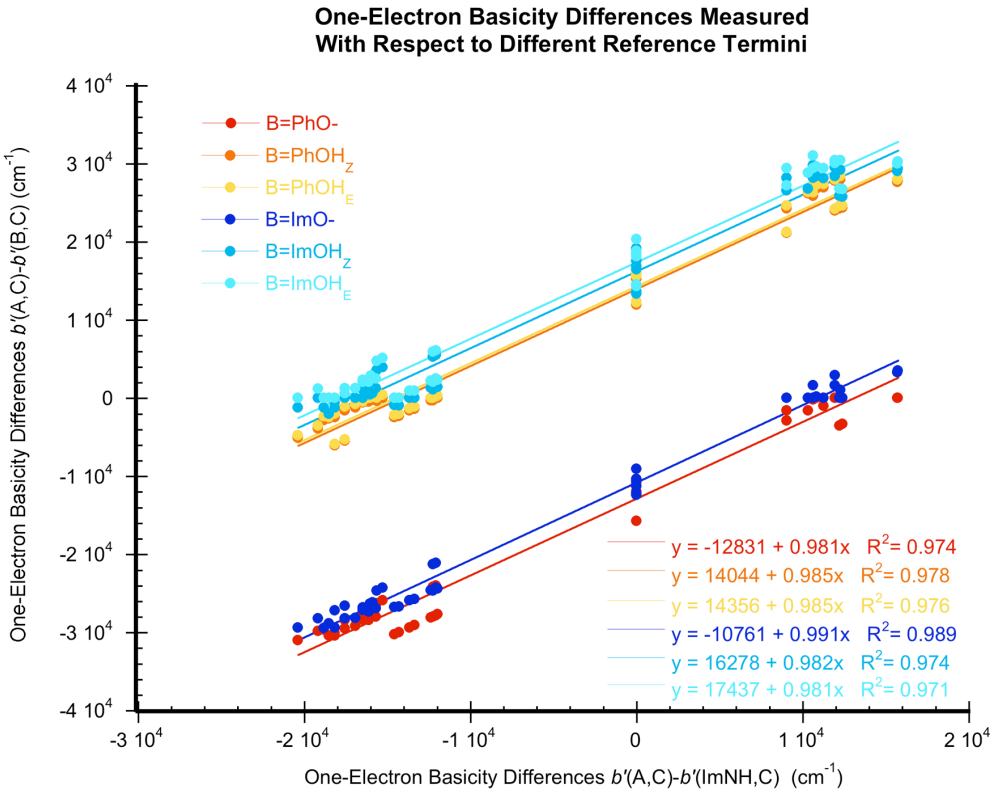


Figure S1.1. One-electron basicity differences measured with respect to different reference terminal groups. One-electron basicities were extracted from orbital energies (diagonal state-averaged Fock matrix elements) of Boys-localized active space orbitals, as described in the main text. Regression lines were fit to each group corresponding to different choices of B. The regression lines are parallel, with slope close to unity. Coefficients of determination (R^2) are close to one, showing that the regression lines represent each group well. The basicity differences can be brought into the same scale (with a common origin) by shifting all the groups according to the *y* intercept of the corresponding regression lines.

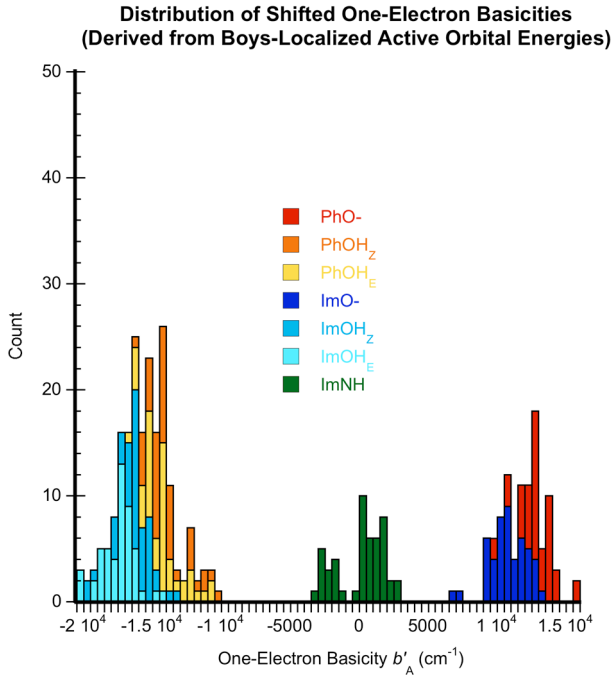


Figure S1.2. Distribution of shifted one-electron basicities for different terminal groups represented in the data set. One-electron basicities were extracted from orbital energies (diagonal state-averaged Fock matrix elements) as described in the main text. The values corresponding to each group are clustered together, suggesting that the data can be adequately represented by the median basicities of the different terminal groups.

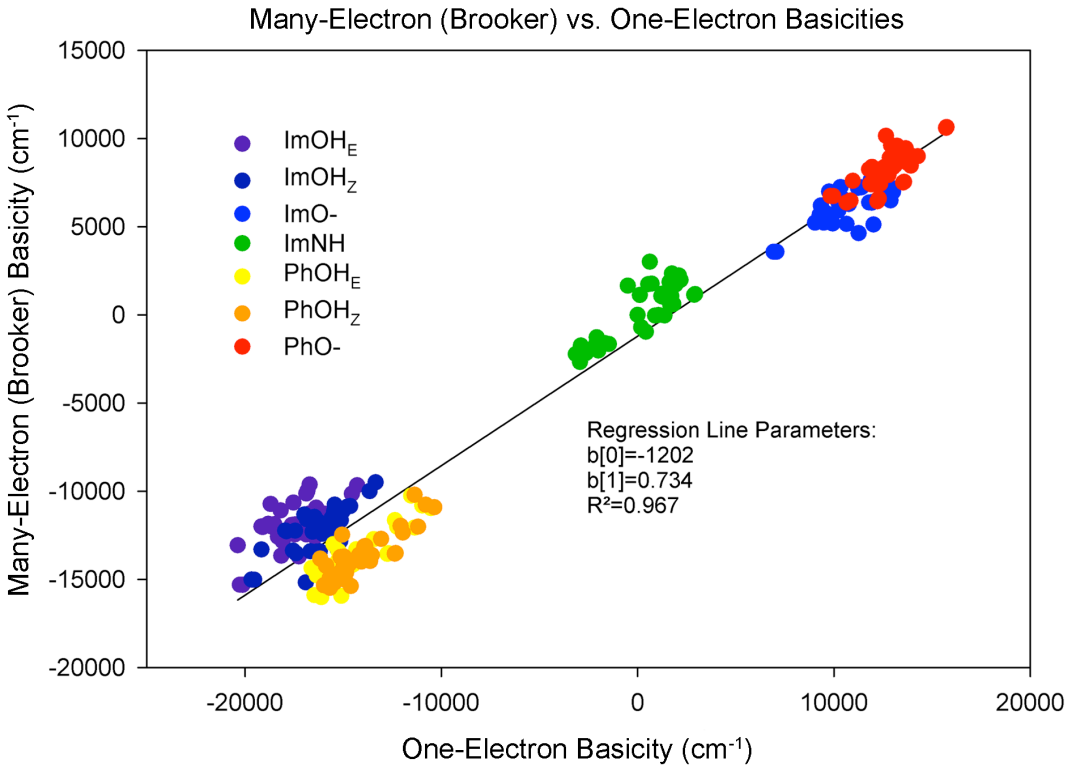


Figure S1.3. This figure shows the correlation between many-electron (Brooker) and one-electron (Boys orbital) basicity scales, after shifting each scale to a common line. Brooker basicities were extracted from calculated excitation energies as described in the main text. One-electron basicities were extracted from orbital energies (diagonal state-averaged Fock matrix elements) of Boys-Localized active space orbitals, as described in the main text. The figure shows that the Brooker basicity does measure a "basicity" in the usual (one-electron) sense.

Table S1.1. Median One-electron basicities (cm^{-1}) for different terminal groups in the data set. Basicities were extracted from Boys Orbital Energies as described in the main text. Median absolute deviations of the distributions are also given.

Group	Median b_A (cm^{-1})	Median Abs. Dev. (cm^{-1})
PhO-	12831	709
$PhOH_Z$	-14071	886
$PhOH_E$	-14417	764
ImO-	10761	1120
$ImOH_Z$	-16278	696
$ImOH_E$	-17437	733
ImNH	424	1242

Data for the Reproduction of Electronic Structure Computations

Summary

This section lists Cartesian coordinates (Å), SA-CASSCF[1] and MS-MRPT2[2] absolute energies (h), MS-MRPT2 mixing coefficients, as well as state-averaged Boys localized and natural active space orbitals. They are listed in one table per page, per dye. All calculations were performed with the Molpro[3] sofware. Geometries are stationary points[4] on the MP2[5] surfaces (not necessarily minima for all cases). All calculations were done with a cc-pvdz[6] basis set. The zeroth order Hamiltonian in the MS-MRPT2 was constructed with the SA-CASSCF canonical orbitals. No level shift was used. One zeroth-order Hamiltonian was used for all states in the MS-MRPT2, and the reference states were allowed to mix during the application of the perturbation.

(1) H.-J. Werner and P. J. Knowles, J. Chem. Phys. 82, 5053 (1985); P. J. Knowles and H.-J. Werner, Chem. Phys. Lett. 115, 259 (1985).

(2) H.-J. Werner, Mol. Phys. 89, 645 (1996); P. Celani and H.-J. Werner, J. Chem. Phys. 112, 5546 (2000).

(3) MOLPRO, version 2009.1, a package of ab initio programs, H.-J. Werner, P. J. Knowles, R. Lindh, F. R. Manby, M. Schütz, P. Celani, T. Korona, A. Mitrushenkov, G. Rauhut, T. B. Adler, R. D. Amos, A. Bernhardsson, A. Berning, D. L. Cooper, M. J. O. Deegan, A. J. Dobbyn, F. Eckert, E. Goll, C. Hampel, G. Hetzer, T. Hrenar, G. Knizia, C. Köppl, Y. Liu, A. W. Lloyd, R. A. Mata, A. J. May, S. J. McNicholas, W. Meyer, M. E. Mura, A. Nicklass, P. Palmieri, K. Pflüger, R. Pitzer, M. Reiher, U. Schumann, H. Stoll, A. J. Stone, R. Tarroni, T. Thorsteinsson, M. Wang, and A. Wolf, , see http://www.molpro.net.

(4) A. El Azhary, G. Rauhut, P. Pulay, and H.-J. Werner, J. Chem. Phys. 108, 5185 (1998).

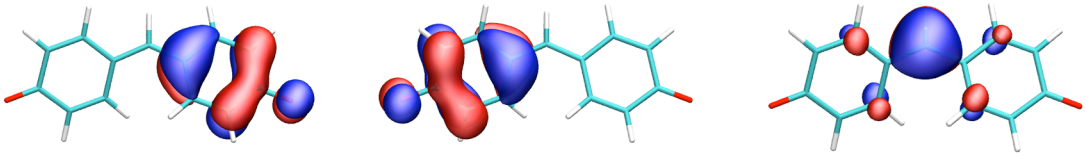
(5) C. Hampel, K. Peterson, and H.-J. Werner, Chem. Phys. Lett. 190, 1 (1992) and references therein.

(6) T.H. Dunning, Jr. J. Chem. Phys. 90, 1007 (1989).

Table S2.1 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (PhO-,PhO-).

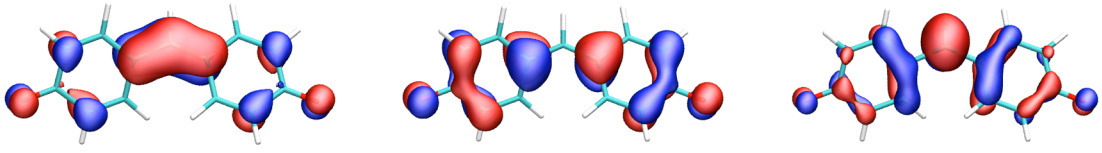
```
ATOMIC COORDINATES
C       -0.00002  -1.13835  -0.00000
C        0.03847  -0.56676   1.29314
C        0.32979   0.81233   1.59153
C        0.32049   1.29047   2.88628
C        0.00913   0.45399   4.05122
C       -0.22615  -0.95458   3.72139
C       -0.19520  -1.42619   2.42613
O       -0.01939   0.89836   5.22096
C       -0.03853  -0.56677  -1.29315
C        0.19518  -1.42620  -2.42613
C        0.22621  -0.95456  -3.72139
C       -0.00909   0.45400  -4.05121
C       -0.32049   1.29048  -2.88627
C       -0.32984   0.81231  -1.59154
O        0.01942   0.89836  -5.22095
H       -0.00000  -2.24242   0.00001
H        0.62965   1.47916   0.77686
H        0.57750   2.33682   3.09401
H       -0.42496  -1.63059   4.56234
H       -0.38328  -2.49154   2.22937
H        0.38331  -2.49154  -2.22937
H        0.42505  -1.63057  -4.56234
H       -0.57750   2.33682  -3.09400
H       -0.62972   1.47914  -0.77687

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -647.315557667736 h
 !MCSCF STATE 2.1 Energy              -647.185849746324 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -649.0228084     1.0000     0.0000
    2    -648.9397750     0.0000     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.4)



Energy: -0.0784 h Occ: 1.48 Energy: -0.0784 h Occ: 1.48 Energy: -0.0253 Occ: 1.04

State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

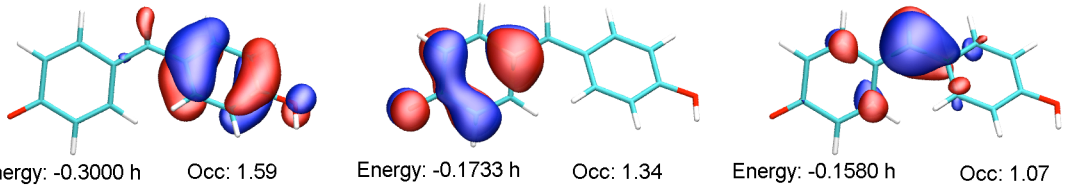


Energy:-0.2202 h Occ: 1.98 Energy:-0.0819 h Occ: 1.49 Energy: 0.1200 h Occ:0.57

Table S2.2 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($PhOH_Z$,PhO-).

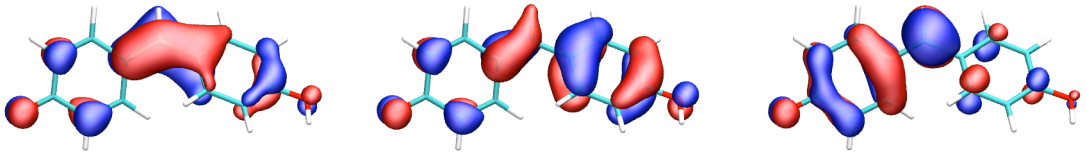
```
ATOMIC COORDINATES
C        0.20783  -1.28233  -0.11317
C        0.13317  -0.67305   1.21645
C        0.64388   0.61653   1.49851
C        0.56350   1.15656   2.78964
C       -0.02603   0.41978   3.83345
C       -0.51140  -0.87707   3.57611
C       -0.41869  -1.41609   2.29012
O       -0.13897   0.88891   5.11021
C        0.06129  -0.66000  -1.33729
C        0.30524  -1.43159  -2.55290
C        0.24301  -0.86081  -3.79093
C       -0.11179   0.56345  -3.97319
C       -0.43929   1.30359  -2.73369
C       -0.35465   0.73019  -1.49665
O       -0.15903   1.09612  -5.08914
H        0.38958  -2.36748  -0.13341
H        1.15251   1.18201   0.71230
H        0.97964   2.15149   2.98994
H       -0.95163  -1.44554   4.40052
H       -0.79805  -2.42690   2.10262
H        0.57724  -2.48853  -2.44079
H        0.45210  -1.43571  -4.69937
H       -0.77771   2.33772  -2.86005
H       -0.65026   1.29524  -0.60747
H1       0.23424   1.78243   5.12834

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -647.859135450908 h
 !MCSCF STATE 2.1 Energy              -647.679492467424 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -649.5611521     0.9998     0.0199
    2    -649.4260530    -0.0199     0.9998
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



Energy: -0.3000 h Occ: 1.59 Energy: -0.1733 h Occ: 1.34 Energy: -0.1580 h Occ: 1.07

State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

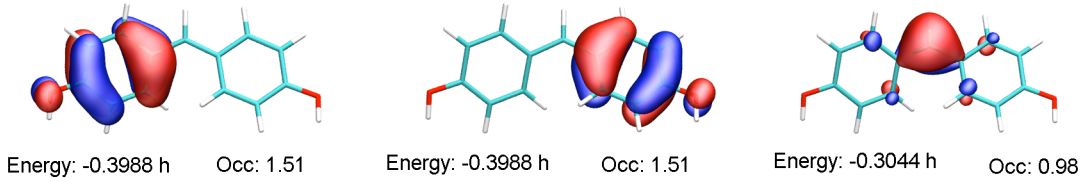


Energy:-0.3663 h Occ: 1.97 Energy:-0.2557 h Occ: 1.49 Energy:-0.0093 h Occ:0.55

Table S2.3 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($PhOH_Z$,$PhOH_Z$).

```
ATOMIC COORDINATES
C        0.00016  -1.23718  -0.00001
C        0.04163  -0.63352   1.28201
C        0.42741   0.72539   1.51429
C        0.42156   1.24753   2.80040
C        0.02326   0.43925   3.89833
C       -0.32209  -0.92109   3.69173
C       -0.28566  -1.45211   2.41285
O       -0.01551   0.88093   5.15842
C       -0.04134  -0.63348  -1.28200
C        0.28564  -1.45211  -2.41292
C        0.32180  -0.92112  -3.69181
C       -0.02342   0.43927  -3.89833
C       -0.42136   1.24761  -2.80031
C       -0.42712   0.72543  -1.51422
O        0.01514   0.88094  -5.15843
H        0.00013  -2.33825  -0.00002
H        0.80870   1.33441   0.69077
H        0.75285   2.27721   2.97584
H       -0.59712  -1.52753   4.55912
H       -0.54875  -2.50333   2.25264
H        0.54868  -2.50336  -2.25274
H        0.59661  -1.52757  -4.55925
H       -0.75264   2.27731  -2.97570
H       -0.80813   1.33452  -0.69061
H1       0.23690   1.81913   5.18841
H1      -0.23715   1.81918  -5.18839

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -648.253519502205 h
 !MCSCF STATE 2.1 Energy              -648.120660752085 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -649.9433234     1.0000     0.0000
    2    -649.8511680     0.0000     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

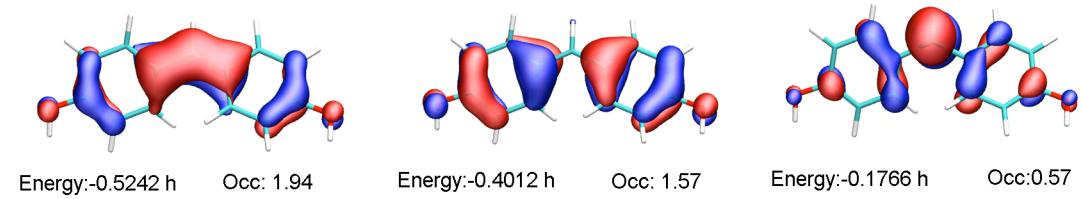
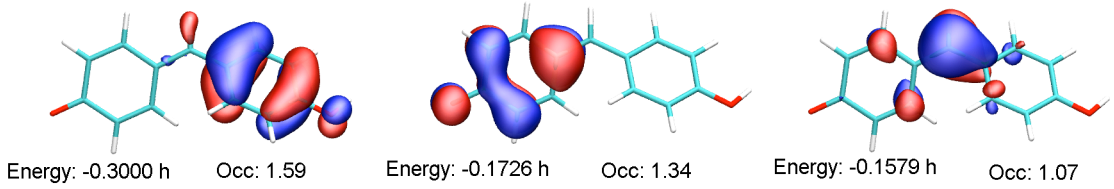


Table S2.4 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($PhOH_E$,PhO-).

```
ATOMIC COORDINATES
C        0.20760  -1.27767  -0.11204
C        0.13540  -0.66570   1.21659
C        0.64887   0.62572   1.49863
C        0.57070   1.16894   2.78542
C       -0.01874   0.43425   3.83121
C       -0.50531  -0.86221   3.57621
C       -0.41479  -1.40650   2.28917
O       -0.06817   1.02387   5.06128
C        0.06281  -0.65757  -1.33747
C        0.30114  -1.43408  -2.55110
C        0.23688  -0.86762  -3.79097
C       -0.11483   0.55693  -3.97717
C       -0.43432   1.30305  -2.73919
C       -0.34743   0.73398  -1.50031
O       -0.16533   1.08515  -5.09509
H        0.38560  -2.36349  -0.13058
H        1.15653   1.18917   0.71030
H        0.97893   2.15953   3.00679
H       -0.95019  -1.44976   4.38846
H       -0.79555  -2.41733   2.10490
H        0.57025  -2.49147  -2.43609
H        0.44113  -1.44645  -4.69802
H       -0.76794   2.33844  -2.86782
H       -0.63654   1.30434  -0.61244
H1      -0.48212   0.39571   5.67107

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -647.859023215532 h
 !MCSCF STATE 2.1 Energy              -647.678924349882 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -649.5610513     0.9998     0.0192
    2    -649.4256986    -0.0192     0.9998
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

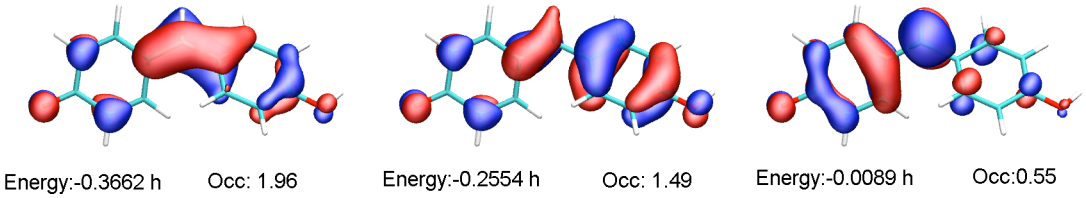


Table S2.5 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($PhOH_E$,$PhOH_Z$).

ATOMIC COORDINATES

C	0.00120	-1.23234	-0.00095
C	0.04076	-0.63131	1.28222
C	0.42108	0.72879	1.51738
C	0.41646	1.24706	2.80490
C	0.02448	0.43393	3.90160
C	-0.31524	-0.92741	3.69216
C	-0.27954	-1.45462	2.41172
O	-0.01424	0.87234	5.16271
C	-0.04433	-0.62564	-1.28159
C	0.28036	-1.44427	-2.41068
C	0.31690	-0.91315	-3.69196
C	-0.02565	0.44768	-3.89776
C	-0.42109	1.25652	-2.79899
C	-0.42847	0.73535	-1.51491
O	-0.03407	1.03291	-5.09852
H	0.00426	-2.33342	-0.00284
H	0.79512	1.34316	0.69461
H	0.74206	2.27829	2.98195
H	-0.58544	-1.53745	4.55853
H	-0.53797	-2.50669	2.24935
H	0.54167	-2.49610	-2.25110
H	0.59402	-1.54398	-4.54379
H	-0.74138	2.28247	-3.00142
H	-0.80649	1.34578	-0.69111
H1	0.23412	1.81159	5.19461
H1	0.22493	0.39082	-5.78103

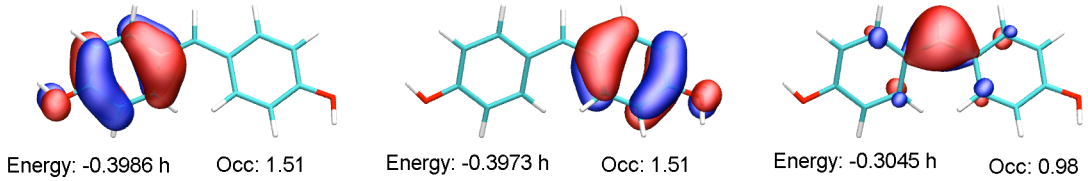
SA-CASSCF RESULTS

!MCSCF STATE 1.1 Energy	-648.254090186788 h
!MCSCF STATE 2.1 Energy	-648.120724938051 h

MS-MRPT2 Results

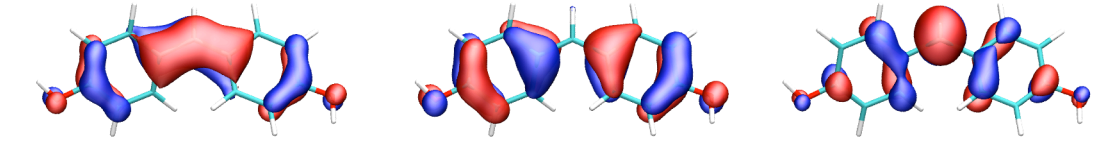
STATE	ENERGY	MIXING COEFFICIENTS	
1	-649.9438682	1.0000	-0.0048
2	-649.8513217	0.0048	1.0000

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.001)



Energy: -0.3986 h Occ: 1.51 Energy: -0.3973 h Occ: 1.51 Energy: -0.3045 h Occ: 0.98

State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.001)



Energy:-0.5238 h Occ: 1.94 Energy:-0.4007 h Occ: 1.49 Energy:-0.1760 h Occ:0.57

Table S2.6 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($PhOH_E$,$PhOH_E$).

ATOMIC COORDINATES

C	0.00096	-1.22465	-0.00006
C	0.04460	-0.62139	1.28219
C	0.41996	0.74141	1.52019
C	0.41272	1.25706	2.80634
C	0.02582	0.44101	3.90296
C	-0.30810	-0.92124	3.69253
C	-0.27126	-1.44675	2.40898
O	0.03273	1.02153	5.10589
C	-0.04420	-0.62149	-1.28225
C	0.27103	-1.44681	-2.40927
C	0.30719	-0.92129	-3.69282
C	-0.02636	0.44110	-3.90296
C	-0.41216	1.25729	-2.80602
C	-0.41883	0.74159	-1.51991
O	-0.03385	1.02169	-5.10582
H	0.00131	-2.32574	-0.00006
H	0.78946	1.35916	0.69813
H	0.72512	2.28488	3.01166
H	-0.57894	-1.55733	4.54245
H	-0.52583	-2.49972	2.24594
H	0.52557	-2.49982	-2.24640
H	0.57727	-1.55742	-4.54295
H	-0.72403	2.28533	-3.01107
H	-0.78744	1.35951	-0.69758
H1	-0.22063	0.37506	5.78638
H1	0.21883	0.37519	-5.78654

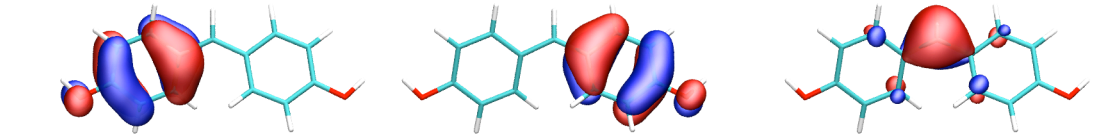
SA-CASSCF RESULTS

!MCSCF STATE 1.1 Energy	-648.254568326509 h
!MCSCF STATE 2.1 Energy	-648.120618262731 h

MS-MRPT2 Results

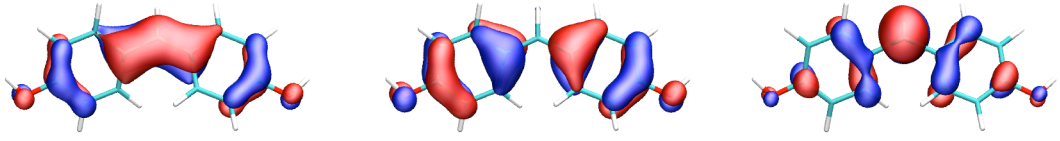
STATE	ENERGY	MIXING COEFFICIENTS	
1	-649.9442359	1.0000	-0.0002
2	-649.8512702	0.0002	1.0000

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



Energy: -0.3972 h Occ: 1.51 Energy: -0.3971 h Occ: 1.51 Energy: -0.3046 h Occ: 0.98

State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

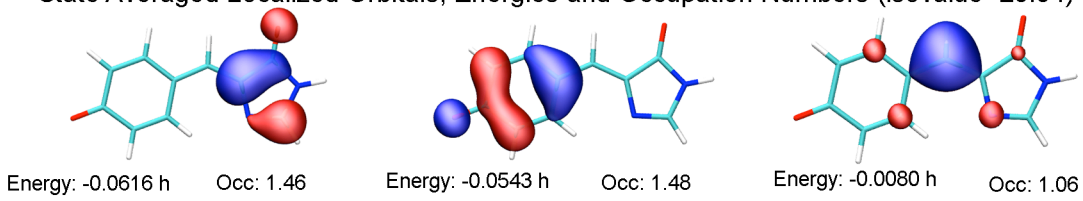


Energy:-0.5233 h Occ: 1.94 Energy:-0.4002 h Occ: 1.49 Energy:-0.1754 h Occ:0.57

Table S2.7 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImO-,PhO-).

```
ATOMIC COORDINATES
N3I      0.00001    0.48726    3.90774
H1N3     0.00003    0.43641    4.91962
C4I     -0.00000    1.62549    3.12295
H1C4     0.00000    2.62655    3.56302
N5I     -0.00001    1.37799    1.82879
C1I     -0.00001   -0.02515    1.73178
C2I      0.00001   -0.65000    3.06283
O1C2     0.00002   -1.82479    3.45338
C1B     -0.00001   -0.78941    0.56328
H1C1    -0.00001   -1.87149    0.77384
C1P     -0.00001   -0.41940   -0.80170
C2P     -0.00001    0.94124   -1.27139
H1C2    -0.00001    1.74061   -0.52307
C3P     -0.00000    1.23034   -2.62133
H1C3     0.00001    2.27263   -2.96684
C4P      0.00001    0.20358   -3.66976
C5P     -0.00001   -1.16718   -3.15394
H1C5    -0.00000   -1.97307   -3.89916
C6P     -0.00001   -1.45082   -1.80239
H1C6    -0.00002   -2.49686   -1.46212
O1C4     0.00002    0.47684   -4.89375

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -641.463008700368 h
 !MCSCF STATE 2.1 Energy              -641.318894628020 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -643.2085856     0.9999     0.0102
    2    -643.1152069    -0.0102     0.9999
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

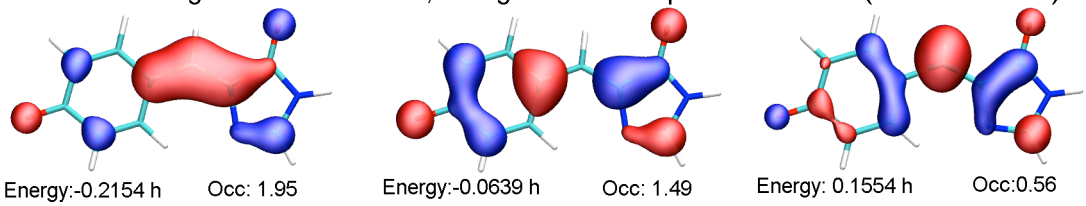
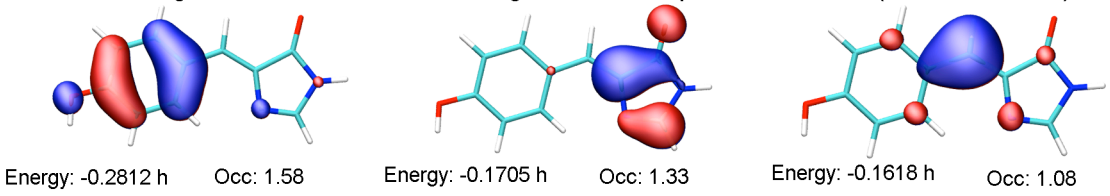


Table S2.8 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImO-,$PhOH_Z$).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01430
C4I      0.00000    1.10353   -0.83297
H1C4    -0.00001    2.11664   -0.42179
N5I      0.00001    0.83753   -2.11716
C1I      0.00002   -0.57180   -2.17912
C2I      0.00002   -1.15989   -0.79904
O1C2     0.00002   -2.31853   -0.41310
C1B      0.00004   -1.36553   -3.29305
H1C1     0.00004   -2.44229   -3.07035
C1P      0.00005   -0.98370   -4.69870
C2P      0.00006    0.36321   -5.14116
H1C2     0.00006    1.16618   -4.39998
C3P      0.00007    0.65716   -6.50881
H1C3     0.00008    1.70412   -6.83809
C4P      0.00008   -0.37552   -7.46804
C5P      0.00006   -1.71674   -7.04225
H1C5     0.00007   -2.50933   -7.79656
C6P      0.00005   -2.01113   -5.67601
H1C6     0.00004   -3.05780   -5.35051
O1C4     0.00009   -0.14229   -8.81406
H1O4     0.00010    0.81802   -8.94153

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.021859733899 h
 !MCSCF STATE 2.1 Energy              -641.836214496059 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -643.7632622     0.9991    -0.0414
    2    -643.6292023     0.0414     0.9991
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

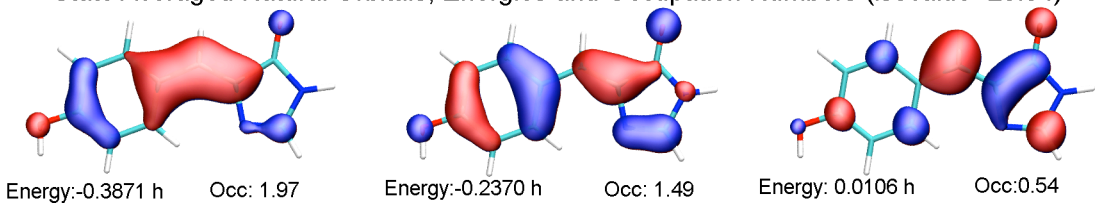
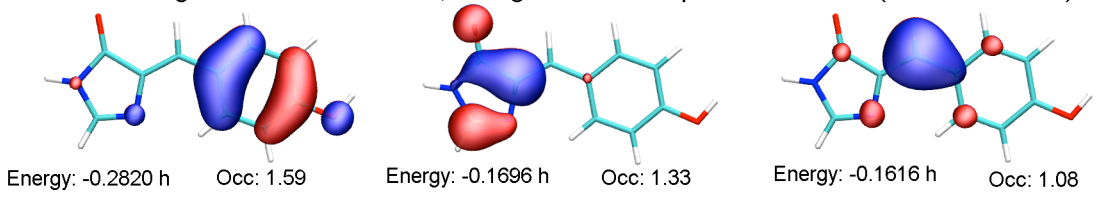


Table S2.9 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImO-,$PhOH_E$).

```
ATOMIC COORDINATES
N3I     -0.00000    0.52046    3.94559
H1N3     0.00000    0.51558    4.95986
C4I     -0.00001    1.62861    3.11826
H1C4    -0.00001    2.63955    3.53473
N5I     -0.00001    1.36943    1.83286
C1I     -0.00000   -0.03932    1.76361
C2I      0.00000   -0.63489    3.14055
O1C2     0.00001   -1.79585    3.51987
C1B      0.00000   -0.82768    0.64594
H1C1     0.00000   -1.90544    0.86398
C1P     -0.00000   -0.43945   -0.75783
C2P      0.00001    0.91108   -1.19757
H1C2     0.00001    1.71059   -0.45266
C3P      0.00001    1.21239   -2.56104
H1C3     0.00001    2.25009   -2.90889
C4P      0.00000    0.18271   -3.52395
C5P     -0.00001   -1.15974   -3.10311
H1C5    -0.00001   -1.96828   -3.84462
C6P     -0.00001   -1.46260   -1.73595
H1C6    -0.00001   -2.51081   -1.41564
O1C4     0.00000    0.55439   -4.83854
H1O4    -0.00001   -0.25558   -5.36993

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.021784397109 h
 !MCSCF STATE 2.1 Energy              -641.835411347333 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -643.7632205     0.9990    -0.0437
    2    -643.6287880     0.0437     0.9990
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

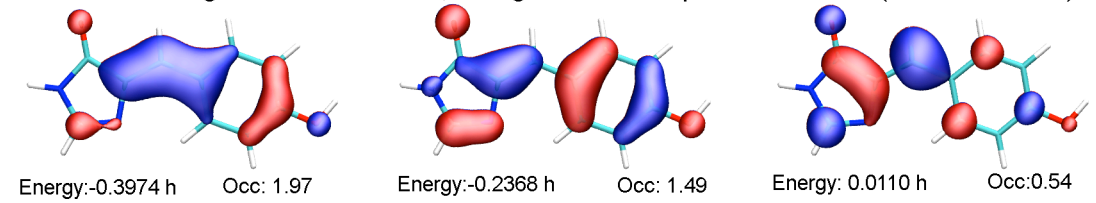
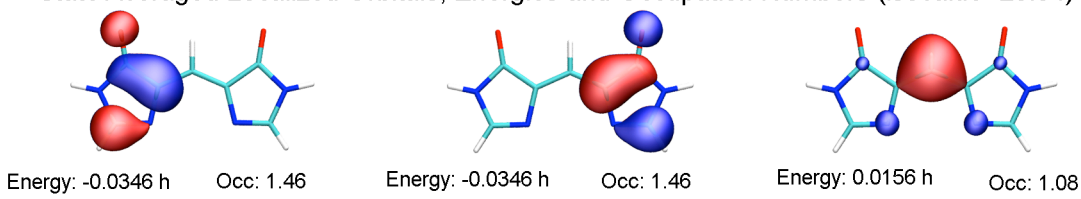


Table S2.10 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImO-,ImO-).

```
ATOMIC COORDINATES
C       -0.00000  -0.50589   0.00000
C       -0.00000   0.05937   1.27850
N       -0.00000   1.42940   1.59851
C       -0.00000   1.46152   2.91526
N       -0.00000   0.20915   3.50628
C       -0.00000  -0.77485   2.48638
O       -0.00000  -1.99905   2.68769
C       -0.00000   0.05937  -1.27849
N        0.00000   1.42940  -1.59851
C        0.00000   1.46152  -2.91526
N        0.00000   0.20915  -3.50628
C        0.00000  -0.77485  -2.48638
O        0.00000  -1.99905  -2.68769
H       -0.00000  -1.60720   0.00000
H       -0.00000   2.37672   3.51505
H        0.00001  -0.00915   4.49544
H        0.00000   2.37672  -3.51506
H        0.00000  -0.00915  -4.49544

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -635.591338886138 h
 !MCSCF STATE 2.1 Energy              -635.435687049858 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -637.3946968     1.0000     0.0000
    2    -637.2884090     0.0000     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

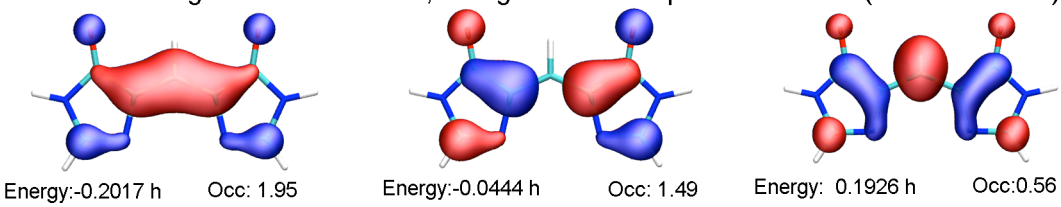
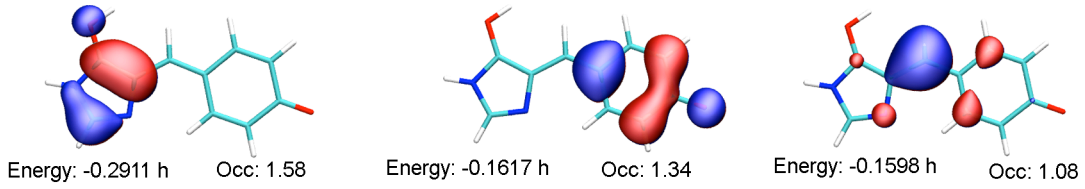


Table S2.11 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_Z$,PhO-).

```
ATOMIC COORDINATES
N3I      0.00000    0.56926    3.83335
H1N3     0.00001    0.52762    4.84801
C4I     -0.00001    1.69335    3.03226
H1C4    -0.00001    2.69975    3.44922
N5I     -0.00001    1.38575    1.74585
C1I     -0.00001   -0.00817    1.68963
C2I      0.00000   -0.51320    3.00205
O1C2     0.00002   -1.75107    3.54892
C1B     -0.00001   -0.81688    0.49728
H1C1    -0.00001   -1.90470    0.67405
C1P     -0.00001   -0.43816   -0.83338
C2P      0.00000    0.94932   -1.28233
H1C2     0.00001    1.73459   -0.52218
C3P      0.00001    1.24918   -2.61615
H1C3     0.00002    2.28894   -2.96185
C4P      0.00000    0.21014   -3.66832
C5P     -0.00001   -1.18677   -3.18432
H1C5    -0.00001   -1.97322   -3.94680
C6P     -0.00001   -1.48200   -1.85111
H1C6    -0.00002   -2.52776   -1.51644
O1C4     0.00001    0.49044   -4.87504
H1O2     0.00002   -2.38624    2.81680

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -641.979531250706 h
 !MCSCF STATE 2.1 Energy              -641.802432719960 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -643.7237918     0.9999     0.0139
    2    -643.5924622    -0.0139     0.9999
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

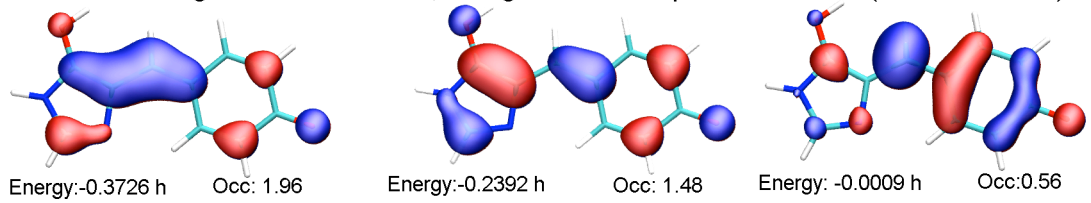
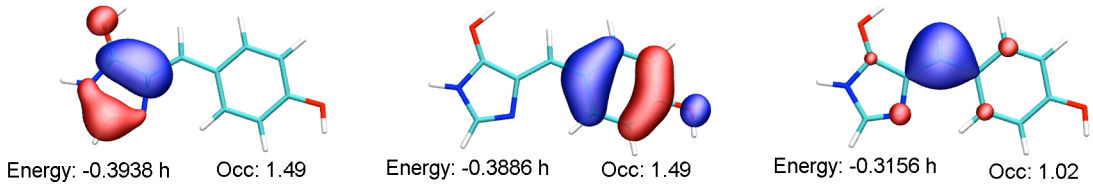


Table S2.12 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_Z$,$PhOH_Z$).

```
ATOMIC COORDINATES
N3I      0.60738    0.00002    3.82154
H1N3     1.35041    0.00010    4.51974
C4I     -0.77148    0.00006    4.04587
H1C4    -1.18213    0.00009    5.05553
N5I     -1.45000    0.00002    2.93066
C1I     -0.49778   -0.00002    1.89132
C2I      0.81349   -0.00001    2.49427
O1C2     2.06113   -0.00003    2.06040
C1B     -1.01249   -0.00004    0.59187
H1C1    -2.10840   -0.00004    0.64820
C1P     -0.50991   -0.00005   -0.74535
C2P      0.84735   -0.00003   -1.17105
H1C2     1.67984   -0.00005   -0.46726
C3P      1.18334   -0.00000   -2.51954
H1C3     2.23565    0.00001   -2.81787
C4P      0.16941    0.00002   -3.51023
C5P     -1.18636    0.00000   -3.11655
H1C5    -1.95707    0.00002   -3.89092
C6P     -1.51178   -0.00003   -1.76791
H1C6    -2.56390   -0.00003   -1.47093
O1C4     0.42051    0.00006   -4.82434
H1O2     2.07011   -0.00006    1.09181
H1O4     1.37804    0.00007   -4.97849

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.368774418160 h
 !MCSCF STATE 2.1 Energy              -642.223179852439 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1036370     0.9956     0.0933
    2    -643.9992598    -0.0933     0.9956
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

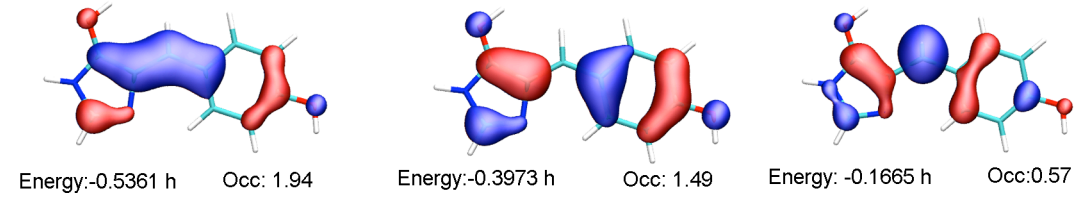
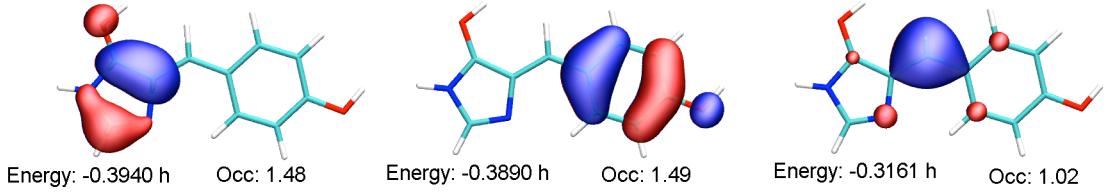


Table S2.13 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_Z$,$PhOH_E$).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01910
C4I      0.00000    1.13792   -0.81926
H1C4    -0.00001    2.14004   -0.38936
N5I      0.00001    0.83925   -2.09551
C1I      0.00001   -0.55814   -2.14955
C2I      0.00001   -1.07754   -0.81775
O1C2     0.00001   -2.35671   -0.47391
C1B      0.00002   -1.36618   -3.29215
H1C1     0.00002   -2.44622   -3.08396
C1P      0.00003   -0.99154   -4.65863
C2P      0.00004    0.36758   -5.10706
H1C2     0.00003    1.17563   -4.37177
C3P      0.00005    0.64803   -6.46539
H1C3     0.00006    1.68893   -6.80867
C4P      0.00006   -0.40584   -7.41804
C5P      0.00005   -1.75647   -6.98818
H1C5     0.00006   -2.54666   -7.74401
C6P      0.00004   -2.04182   -5.63208
H1C6     0.00004   -3.08449   -5.29549
O1C4     0.00008   -0.19762   -8.73948
H1O2     0.00001   -2.47081    0.49131
H1O4     0.00008    0.75745   -8.91966

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.387646550938 h
 !MCSCF STATE 2.1 Energy              -642.241956288559 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1169626     0.9998     0.0206
    2    -644.0149093    -0.0206     0.9998
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

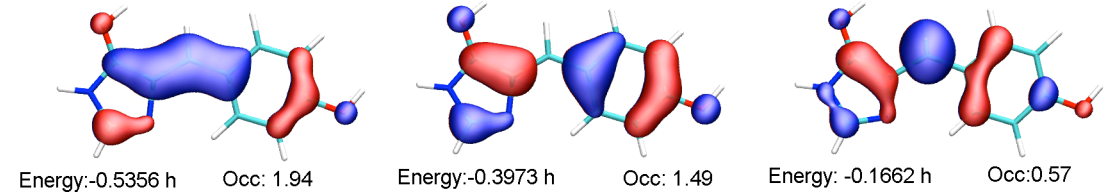
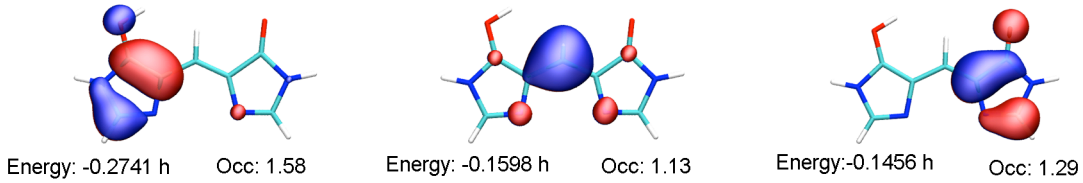


Table S2.14 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_Z$,ImO-).

```
ATOMIC COORDINATES
C       -0.00000  -0.51802  -0.05387
C       -0.00000   0.08235   1.25069
N       -0.00000   1.44783   1.53027
C       -0.00000   1.53285   2.84897
N       -0.00000   0.28986   3.45771
C       -0.00000  -0.63774   2.45755
O       -0.00000  -1.95160   2.78334
C       -0.00000   0.06537  -1.29082
N        0.00000   1.43594  -1.61409
C        0.00000   1.44652  -2.92442
N        0.00000   0.19830  -3.53323
C        0.00000  -0.78000  -2.52724
O        0.00000  -1.99595  -2.67381
H       -0.00000  -1.61864  -0.10410
H       -0.00000   2.45447   3.43030
H        0.00001   0.08072   4.45115
H        0.00000   2.35938  -3.52629
H        0.00000   0.00189  -4.52806
H1      -0.00000  -2.44890   1.95083

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.130281191373 h
 !MCSCF STATE 2.1 Energy              -635.943234809276 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -637.9280597     1.0000    -0.0006
    2    -637.7902508     0.0006     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

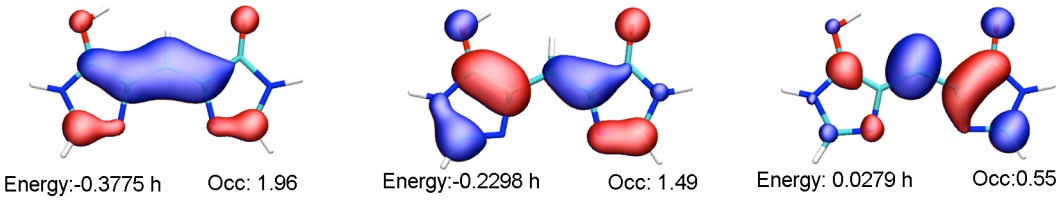
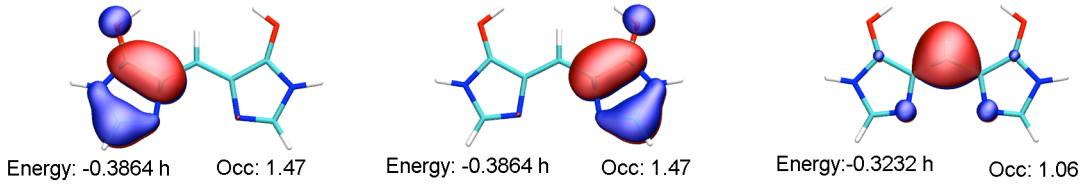


Table S2.15 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_Z$,$ImOH_Z$).

```
ATOMIC COORDINATES
C       -0.00000  -0.57851  -0.00001
C       -0.00000   0.03868   1.25643
N       -0.00000   1.41788   1.48429
C       -0.00000   1.54775   2.78848
N       -0.00000   0.31779   3.45673
C       -0.00000  -0.64675   2.51866
O       -0.00000  -1.92006   2.89741
C       -0.00000   0.03874  -1.25631
N       -0.00000   1.41797  -1.48428
C        0.00000   1.54787  -2.78837
N        0.00000   0.31777  -3.45670
C        0.00000  -0.64670  -2.51870
O        0.00000  -1.92004  -2.89753
H       -0.00000  -1.68022  -0.00016
H       -0.00000   2.48883   3.33925
H        0.00000   0.15530   4.46335
H        0.00000   2.48887  -3.33926
H        0.00000   0.15539  -4.46334
H1      -0.00000  -2.49672   2.11692
H1       0.00000  -2.49696  -2.11717

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.501568311079 h
 !MCSCF STATE 2.1 Energy              -636.349935660346 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.2915165     1.0000    -0.0002
    2    -638.1797991     0.0002     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

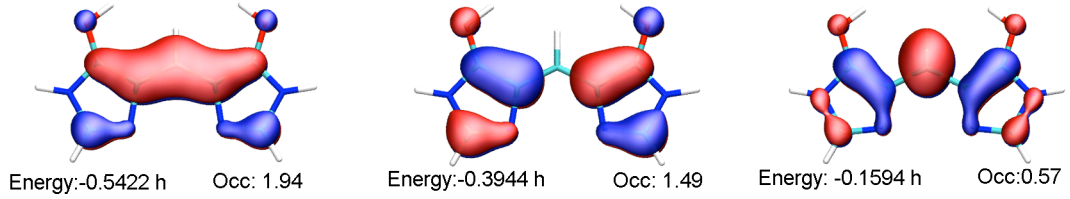
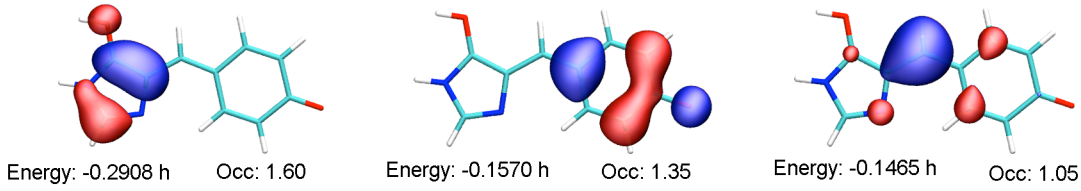


Table S2.16 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,PhO-).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01543
C4I      0.00000    1.11801   -0.81566
H1C4    -0.00001    2.12741   -0.40564
N5I      0.00001    0.79493   -2.09723
C1I      0.00002   -0.59833   -2.13912
C2I      0.00001   -1.09483   -0.82783
O1C2     0.00002   -2.38896   -0.42705
C1B      0.00003   -1.43212   -3.31599
H1C1     0.00003   -2.50938   -3.09687
C1P      0.00005   -1.07019   -4.65035
C2P      0.00007    0.31004   -5.12201
H1C2     0.00006    1.10804   -4.37516
C3P      0.00009    0.58940   -6.46022
H1C3     0.00010    1.62356   -6.82258
C4P      0.00011   -0.46590   -7.49620
C5P      0.00009   -1.85473   -6.99029
H1C5     0.00010   -2.65279   -7.74070
C6P      0.00007   -2.12954   -5.65269
H1C6     0.00006   -3.16894   -5.29983
O1C4     0.00013   -0.20347   -8.70712
H1O2     0.00001   -2.42715    0.53969

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -641.974616969015 h
 !MCSCF STATE 2.1 Energy              -641.794693208253 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -643.7173379     1.0000    -0.0047
    2    -643.5842348     0.0047     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

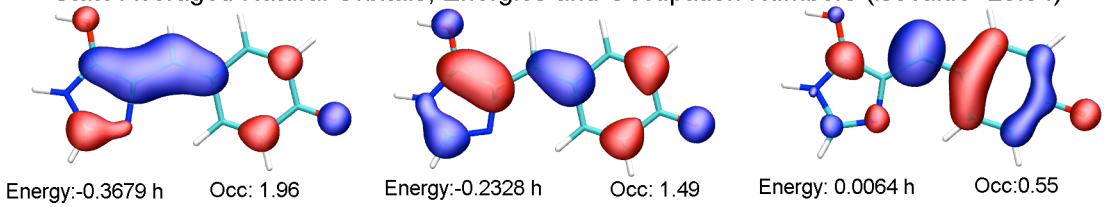
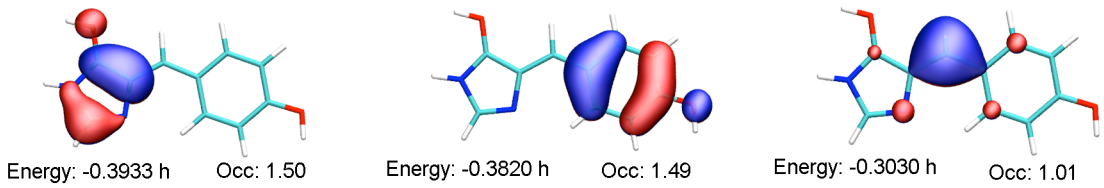


Table S2.17 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,$PhOH_Z$).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01910
C4I      0.00000    1.13792   -0.81926
H1C4    -0.00001    2.14004   -0.38936
N5I      0.00001    0.83925   -2.09551
C1I      0.00001   -0.55814   -2.14955
C2I      0.00001   -1.07754   -0.81775
O1C2     0.00001   -2.35671   -0.47391
C1B      0.00002   -1.36618   -3.29215
H1C1     0.00002   -2.44622   -3.08396
C1P      0.00003   -0.99154   -4.65863
C2P      0.00004    0.36758   -5.10706
H1C2     0.00003    1.17563   -4.37177
C3P      0.00005    0.64803   -6.46539
H1C3     0.00006    1.68893   -6.80867
C4P      0.00006   -0.40584   -7.41804
C5P      0.00005   -1.75647   -6.98818
H1C5     0.00006   -2.54666   -7.74401
C6P      0.00004   -2.04182   -5.63208
H1C6     0.00004   -3.08449   -5.29549
O1C4     0.00008   -0.19762   -8.73948
H1O2     0.00001   -2.47081    0.49131
H1O4     0.00008    0.75745   -8.91966

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.387646550938 h
 !MCSCF STATE 2.1 Energy              -642.241956288559 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1169626     0.9998     0.0206
    2    -644.0149093    -0.0206     0.9998
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

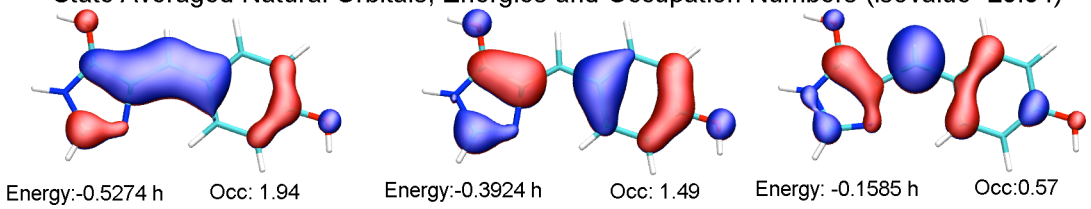
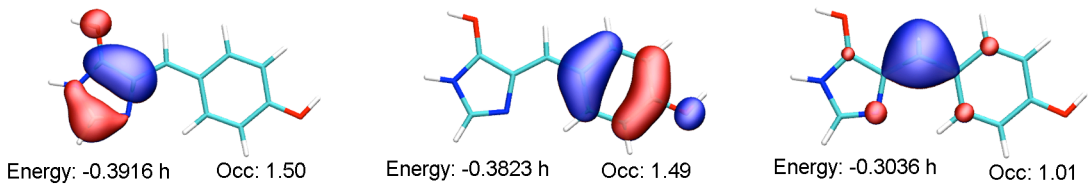


Table S2.18 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,$PhOH_E$).

```
ATOMIC COORDINATES
N3I      0.00000    0.55891    3.87599
H1N3     0.00001    0.55778    4.89510
C4I     -0.00000    1.69818    3.05787
H1C4    -0.00000    2.69978    3.48905
N5I     -0.00000    1.40113    1.78153
C1I     -0.00000    0.00385    1.72563
C2I      0.00000   -0.51735    3.05704
O1C2     0.00001   -1.79705    3.39918
C1B     -0.00000   -0.80201    0.58181
H1C1    -0.00000   -1.88250    0.78765
C1P     -0.00000   -0.42260   -0.78363
C2P      0.00000    0.93812   -1.23288
H1C2     0.00000    1.74580   -0.49748
C3P      0.00000    1.21851   -2.58925
H1C3     0.00001    2.24813   -2.95830
C4P      0.00000    0.16509   -3.54260
C5P     -0.00000   -1.18560   -3.11367
H1C5    -0.00000   -1.99641   -3.85056
C6P     -0.00000   -1.47170   -1.75543
H1C6    -0.00001   -2.51472   -1.41972
O1C4     0.00000    0.52837   -4.83018
H1O2     0.00001   -1.91217    4.36427
H1O4     0.00000   -0.26349   -5.39383

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.387678175014 h
 !MCSCF STATE 2.1 Energy              -642.241332184770 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1170415     0.9997     0.0261
    2    -644.0142759    -0.0261     0.9997
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

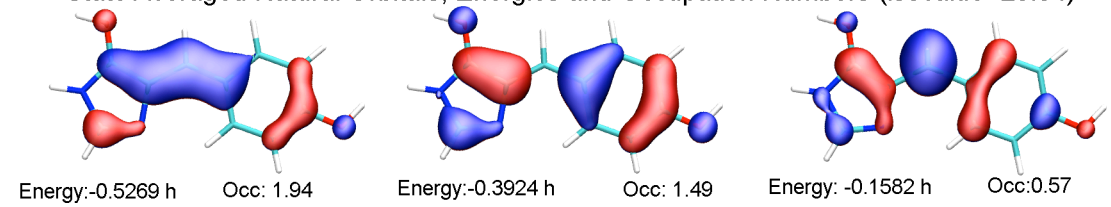
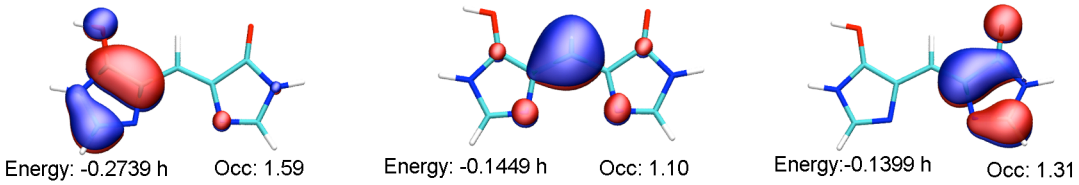


Table S2.19 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,ImO-).

```
ATOMIC COORDINATES
C       -0.00000  -0.53994  -0.05477
C       -0.00000   0.07802   1.24488
N       -0.00000   1.44479   1.51350
C       -0.00000   1.54434   2.83005
N       -0.00000   0.30438   3.45168
C       -0.00000  -0.63488   2.45130
O       -0.00000  -1.97864   2.62986
C       -0.00000   0.04774  -1.28903
N       -0.00000   1.42293  -1.59964
C        0.00000   1.44832  -2.90939
N        0.00000   0.20789  -3.53198
C        0.00000  -0.78378  -2.53655
O        0.00000  -1.99604  -2.70341
H       -0.00000  -1.63867  -0.07128
H       -0.00000   2.46941   3.40613
H        0.00001   0.13306   4.45229
H        0.00000   2.36823  -3.50077
H        0.00000   0.02196  -4.52870
H1      -0.00000  -2.17546   3.57688

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.123553029051 h
 !MCSCF STATE 2.1 Energy              -635.933046478531 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1   -637.9204518     0.9998    -0.0202
    2   -637.7800224     0.0202     0.9998
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

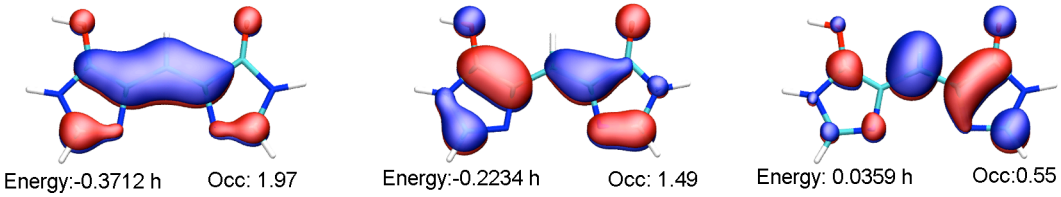
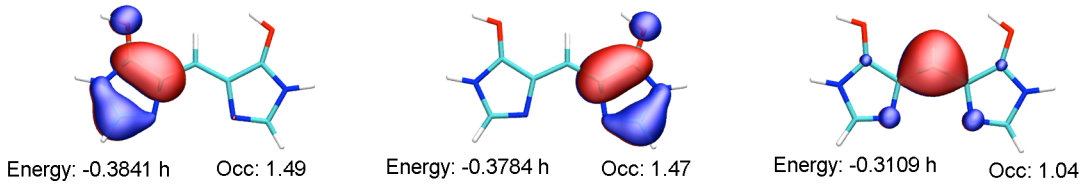


Table S2.20 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,$ImOH_Z$).

```
ATOMIC COORDINATES
C       -0.00000  -0.55169  -0.00352
C       -0.00000   0.05503   1.25430
N       -0.00000   1.43142   1.50177
C       -0.00000   1.54158   2.80748
N       -0.00000   0.30089   3.45851
C       -0.00000  -0.64909   2.50615
O       -0.00000  -1.92851   2.85781
C       -0.00000   0.07037  -1.25845
N       -0.00000   1.44231  -1.51806
C        0.00000   1.53901  -2.82467
N        0.00000   0.28806  -3.46458
C        0.00000  -0.64959  -2.49072
O        0.00000  -1.97124  -2.63304
H       -0.00000  -1.65152  -0.03824
H       -0.00000   2.47378   3.37313
H        0.00001   0.12449   4.46262
H        0.00000   2.46290  -3.40393
H        0.00000   0.13087  -4.47126
H1      -0.00000  -2.48643   2.06259
H1       0.00000  -2.22865  -3.56987

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.503964030180 h
 !MCSCF STATE 2.1 Energy              -636.351127229697 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.2924931     1.0000    0.0025
    2    -638.1796859    -0.0025    1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

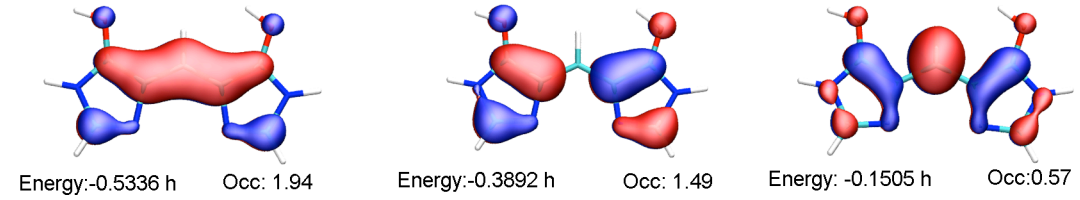
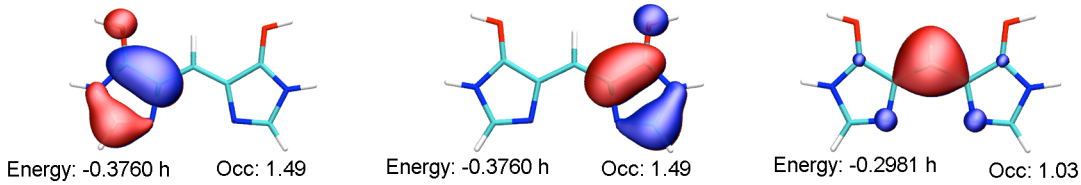


Table S2.21 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImOH_E$,$ImOH_E$).

```
ATOMIC COORDINATES
C       -0.00000  -0.54313   0.00001
C       -0.00000   0.07306   1.25532
N       -0.00000   1.44506   1.52015
C       -0.00000   1.53773   2.82656
N       -0.00000   0.28430   3.46275
C       -0.00000  -0.65073   2.48635
O       -0.00000  -1.97115   2.62633
C       -0.00000   0.07304  -1.25532
N       -0.00000   1.44506  -1.52011
C        0.00000   1.53773  -2.82655
N        0.00000   0.28434  -3.46272
C        0.00000  -0.65072  -2.48637
O        0.00000  -1.97114  -2.62640
H       -0.00000  -1.64221   0.00003
H       -0.00000   2.45940   3.40934
H        0.00001   0.12390   4.46879
H        0.00000   2.45943  -3.40929
H        0.00000   0.12395  -4.46877
H1      -0.00000  -2.22973   3.56274
H1       0.00000  -2.22965  -3.56283

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.503771040689 h
 !MCSCF STATE 2.1 Energy              -636.350342176125 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.2911772     1.0000     0.0000
    2    -638.1776914     0.0000     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

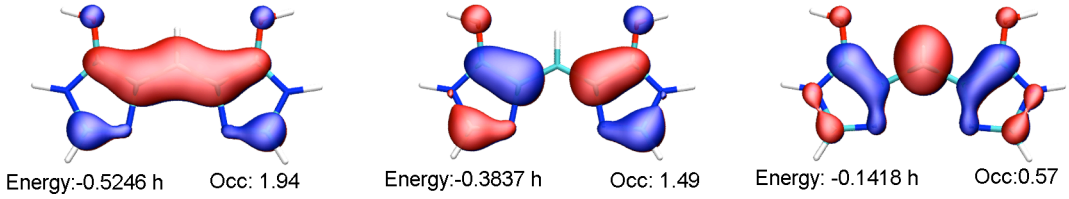
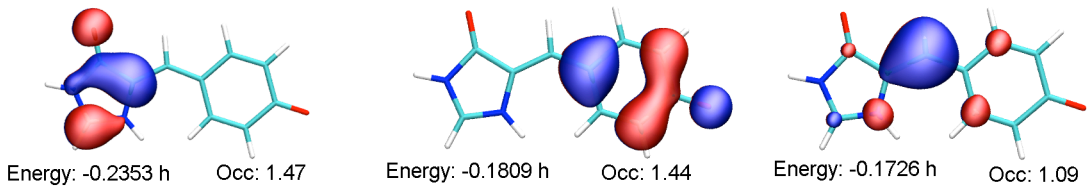


Table S2.22 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImNH,PhO-).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01995
C4I      0.00000    1.08876   -0.77029
H1C4    -0.00001    2.11828   -0.40919
N5I      0.00001    0.75042   -2.05785
C1I      0.00002   -0.65095   -2.19607
C2I      0.00001   -1.20047   -0.81962
O1C2     0.00002   -2.33347   -0.40137
C1B      0.00004   -1.44981   -3.31409
H1C1     0.00004   -2.51352   -3.03318
C1P      0.00006   -1.17197   -4.72537
C2P      0.00009    0.12234   -5.31329
H1C2     0.00010    1.03248   -4.70665
C3P      0.00011    0.28026   -6.69775
H1C3     0.00013    1.28666   -7.13147
C4P      0.00011   -0.85309   -7.54651
C5P      0.00008   -2.14723   -6.98031
H1C5     0.00007   -3.01280   -7.64870
C6P      0.00006   -2.29901   -5.59785
H1C6     0.00003   -3.30598   -5.16665
O1C4     0.00012   -0.77435   -8.88901
H1O4     0.00014    0.15811   -9.15879
H1N5     0.00001    1.43337   -2.80998

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.390812675191 h
 !MCSCF STATE 2.1 Energy              -642.229547563191 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1240181     0.9999    -0.0116
    2    -644.0183854     0.0116     0.9999
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

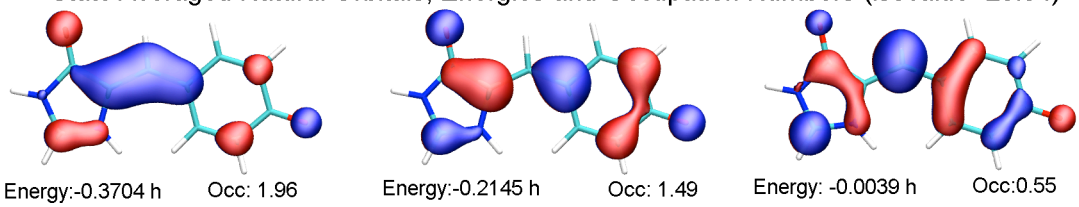
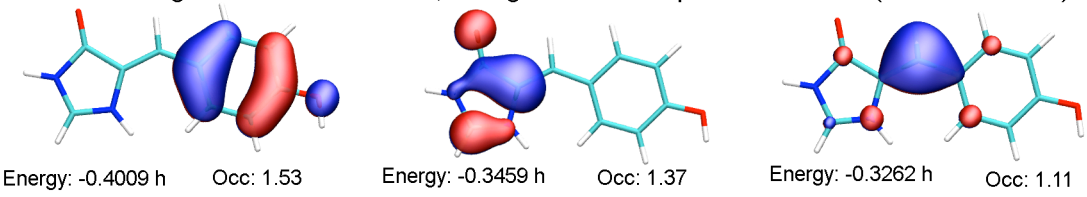


Table S2.23 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImNH,PhOH_Z$).

```
ATOMIC COORDINATES
N3I      0.00000    0.00000    0.00000
H1N3     0.00000    0.00000    1.01995
C4I      0.00000    1.08876   -0.77029
H1C4    -0.00001    2.11828   -0.40919
N5I      0.00001    0.75042   -2.05785
C1I      0.00002   -0.65095   -2.19607
C2I      0.00001   -1.20047   -0.81962
O1C2     0.00002   -2.33347   -0.40137
C1B      0.00004   -1.44981   -3.31409
H1C1     0.00004   -2.51352   -3.03318
C1P      0.00006   -1.17197   -4.72537
C2P      0.00009    0.12234   -5.31329
H1C2     0.00010    1.03248   -4.70665
C3P      0.00011    0.28026   -6.69775
H1C3     0.00013    1.28666   -7.13147
C4P      0.00011   -0.85309   -7.54651
C5P      0.00008   -2.14723   -6.98031
H1C5     0.00007   -3.01280   -7.64870
C6P      0.00006   -2.29901   -5.59785
H1C6     0.00003   -3.30598   -5.16665
O1C4     0.00012   -0.77435   -8.88901
H1O4     0.00014    0.15811   -9.15879
H1N5     0.00001    1.43337   -2.80998

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.390812675191 h
 !MCSCF STATE 2.1 Energy              -642.229547563191 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1240181     0.9999    -0.0116
    2    -644.0183854     0.0116     0.9999
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

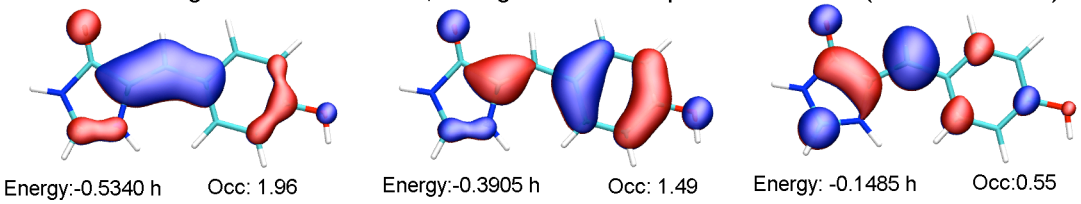
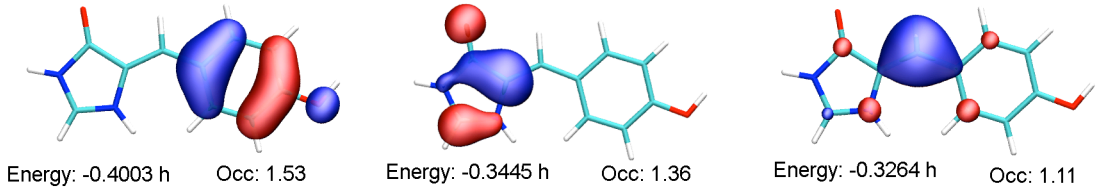


Table S2.24 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImNH,PhOH$_E$).

```
ATOMIC COORDINATES
N3I     -0.00000    0.42277    3.99763
H1N3     0.00001    0.34597    5.01466
C4I     -0.00002    1.56683    3.31178
H1C4    -0.00002    2.56616    3.74963
N5I     -0.00002    1.32688    2.00247
C1I     -0.00000   -0.06005    1.75888
C2I      0.00001   -0.71202    3.08967
O1C2     0.00002   -1.87363    3.42073
C1B      0.00000   -0.77147    0.58319
H1C1     0.00001   -1.85367    0.78113
C1P      0.00000   -0.38237   -0.80130
C2P      0.00002    0.95665   -1.28562
H1C2     0.00004    1.81529   -0.60821
C3P      0.00002    1.22394   -2.64995
H1C3     0.00004    2.25012   -3.02834
C4P      0.00000    0.16199   -3.58724
C5P     -0.00002   -1.17295   -3.12750
H1C5    -0.00003   -2.00251   -3.84337
C6P     -0.00002   -1.43500   -1.75864
H1C6    -0.00003   -2.47336   -1.40962
O1C4     0.00000    0.50866   -4.88655
H1O4    -0.00001   -0.29380   -5.43352
H1N5    -0.00003    2.06462    1.30380

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -642.392025042778 h
 !MCSCF STATE 2.1 Energy              -642.230201225716 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -644.1250978     0.9999    -0.0117
    2    -644.0190111     0.0117     0.9999
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

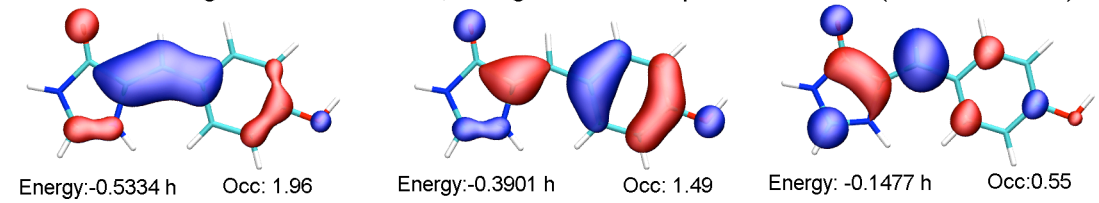
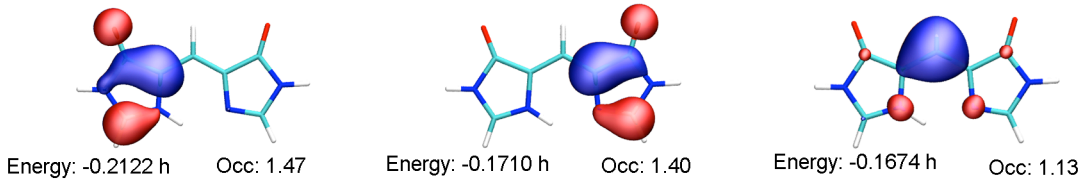


Table S2.25 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImNH,ImO-).

```
ATOMIC COORDINATES
C       -0.00000  -0.81763  -0.04302
C       -0.00000  -0.19275   1.22365
N       -0.00000   1.18755   1.36162
C       -0.00000   1.53982   2.64937
N       -0.00000   0.40304   3.38652
C       -0.00000  -0.77779   2.55296
O       -0.00000  -1.92950   2.97587
C       -0.00000  -0.13335  -1.24153
N       -0.00000   1.27575  -1.33991
C        0.00000   1.52222  -2.63437
N        0.00000   0.39365  -3.41854
C        0.00000  -0.75494  -2.58323
O        0.00000  -1.92059  -2.95572
H       -0.00000  -1.91364  -0.07037
H       -0.00000   2.55907   3.02792
H        0.00001   0.36153   4.40141
H        0.00000   2.52466  -3.06887
H        0.00000   0.35929  -4.43253
H1      -0.00000   1.77641   0.51043

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.134512776219 h
 !MCSCF STATE 2.1 Energy              -635.978402975174 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -637.9432825     1.0000     0.0008
    2    -637.8436273    -0.0008     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

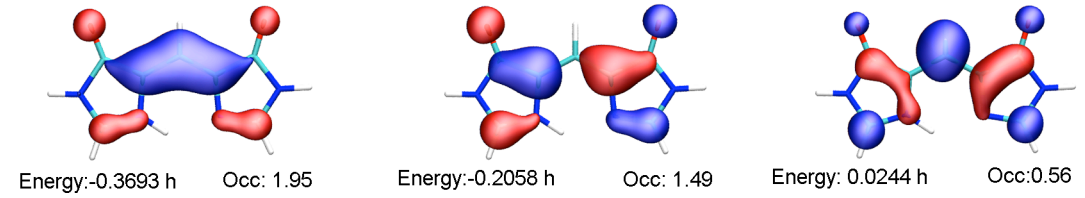
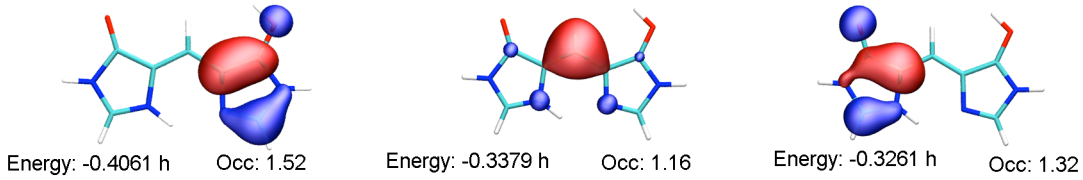


Table S2.26 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImNH,ImOH$_Z$).

```
ATOMIC COORDINATES
C       -0.00000  -0.80842  -0.01247
C       -0.00000  -0.10344   1.22356
N       -0.00000   1.29606   1.28125
C       -0.00000   1.60156   2.56729
N       -0.00000   0.47176   3.35805
C       -0.00000  -0.61527   2.53984
O       -0.00000  -1.83171   3.08848
C       -0.00000  -0.18400  -1.23508
N       -0.00000   1.20789  -1.41300
C        0.00000   1.49545  -2.70650
N        0.00000   0.37042  -3.43814
C        0.00000  -0.79769  -2.57821
O        0.00000  -1.94993  -2.94674
H       -0.00000  -1.90561  -0.03887
H       -0.00000   2.60598   2.98900
H        0.00001   0.43348   4.37622
H        0.00000   2.50774  -3.11319
H        0.00000   0.33122  -4.45657
H1      -0.00000  -2.50364   2.38792
H1      -0.00000   1.83204  -0.58596

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.531650466906 h
 !MCSCF STATE 2.1 Energy              -636.365478098275 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.3279346     0.9991    -0.0432
    2    -638.2117288     0.0432     0.9991
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

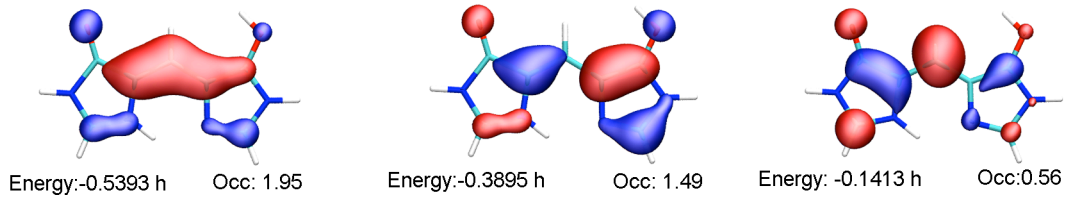
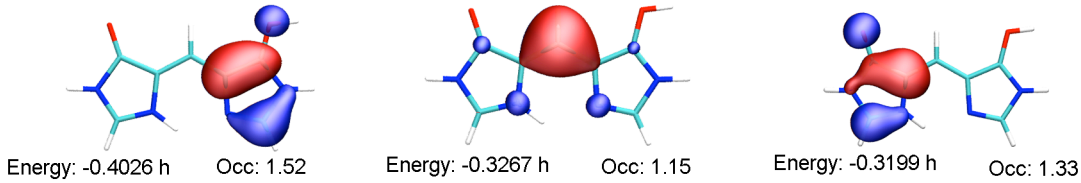


Table S2.27 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye ($ImNH,ImOH_E$).

```
ATOMIC COORDINATES
C       -0.00000  -0.80289  -0.00687
C       -0.00000  -0.08651   1.22379
N       -0.00000   1.30972   1.29519
C       -0.00000   1.60175   2.58372
N       -0.00000   0.45945   3.36561
C       -0.00000  -0.61893   2.52576
O       -0.00000  -1.90964   2.86507
C       -0.00000  -0.18505  -1.23150
N       -0.00000   1.20800  -1.41135
C        0.00000   1.49579  -2.70461
N        0.00000   0.37108  -3.43598
C        0.00000  -0.79861  -2.57561
O        0.00000  -1.94888  -2.94809
H       -0.00000  -1.89938   0.00289
H       -0.00000   2.59964   3.02078
H        0.00001   0.44179   4.38367
H        0.00000   2.50814  -3.11108
H        0.00000   0.33164  -4.45426
H1      -0.00000  -2.02121   3.82897
H1      -0.00000   1.83661  -0.58913

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.529384221051 h
 !MCSCF STATE 2.1 Energy              -636.361804779076 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.3251641     0.9993    -0.0385
    2    -638.2076781     0.0385     0.9993
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)

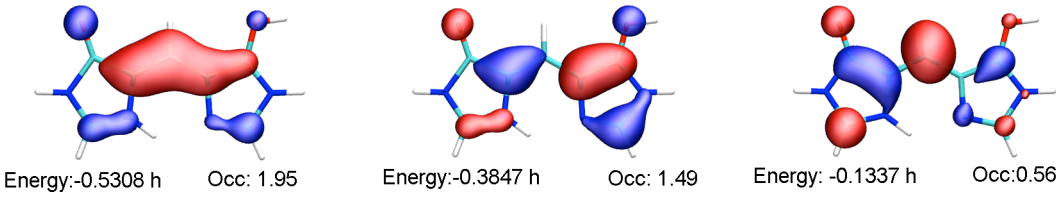
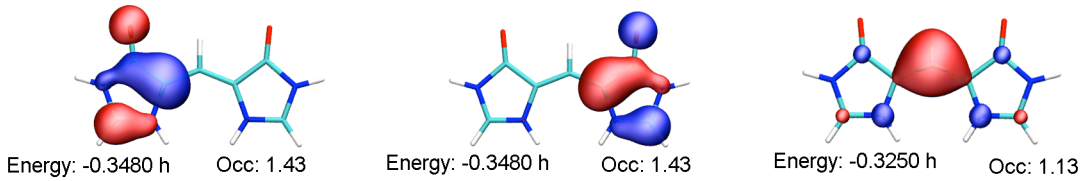


Table S2.28 Cartesian coordinates (Å), SA-CASSCF and MS-MRPT2 absolute energies (h), MS-MRPT2 mixing matricies, and state-averaged Boys localized and natural active space orbitals with energies (diagonal Fock matrix elements, h) and occupation numbers for the dye (ImNH,ImNH).

```
ATOMIC COORDINATES
C       -0.00000  -0.46174   0.00000
C       -0.00000   0.05015   1.29325
N       -0.00000   1.36780   1.77463
C       -0.00000   1.38064   3.11180
N       -0.00000   0.12499   3.56815
C       -0.00000  -0.82733   2.47179
O       -0.00000  -2.03399   2.59170
C       -0.00000   0.05021  -1.29325
N        0.00000   1.36780  -1.77468
C        0.00000   1.38059  -3.11185
N        0.00000   0.12492  -3.56816
C        0.00000  -0.82734  -2.47177
O        0.00000  -2.03401  -2.59159
H       -0.00000  -1.56362  -0.00008
H       -0.00000   2.28508   3.71949
H        0.00001  -0.14206   4.55196
H        0.00000   2.28502  -3.71957
H        0.00000  -0.14216  -4.55195
H1      -0.00000   2.22604   1.23406
H1       0.00000   2.22612  -1.23422

SA-CASSCF RESULTS
 !MCSCF STATE 1.1 Energy              -636.502800499318 h
 !MCSCF STATE 2.1 Energy              -636.344840655342 h

MS-MRPT2 Results
 STATE        ENERGY      MIXING COEFFICIENTS
    1    -638.3013924     1.0000     0.0000
    2    -638.2125540     0.0000     1.0000
```

State Averaged Localized Orbitals, Energies and Occupation Numbers (isovalue=±0.04)



State Averaged Natural Orbitals, Energies and Occupation Numbers (isovalue=±0.04)