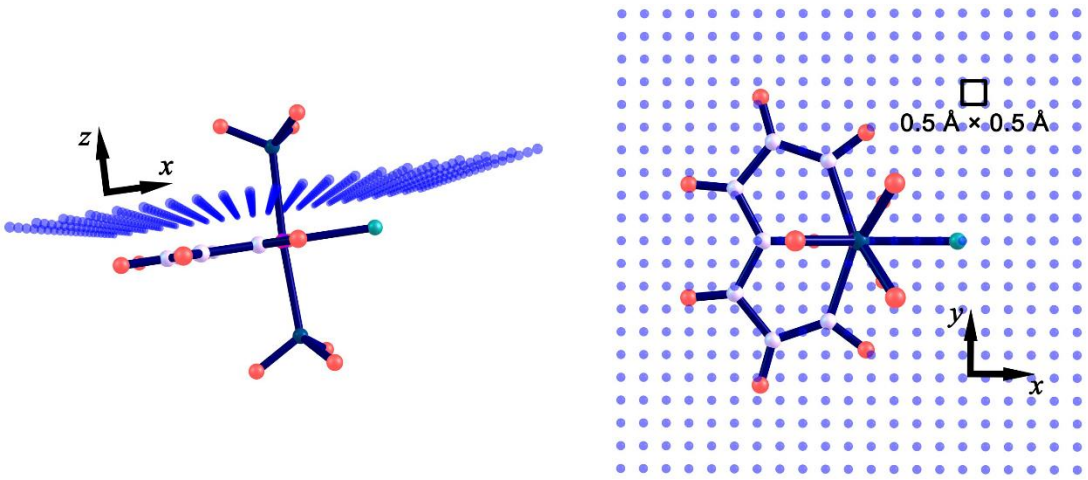
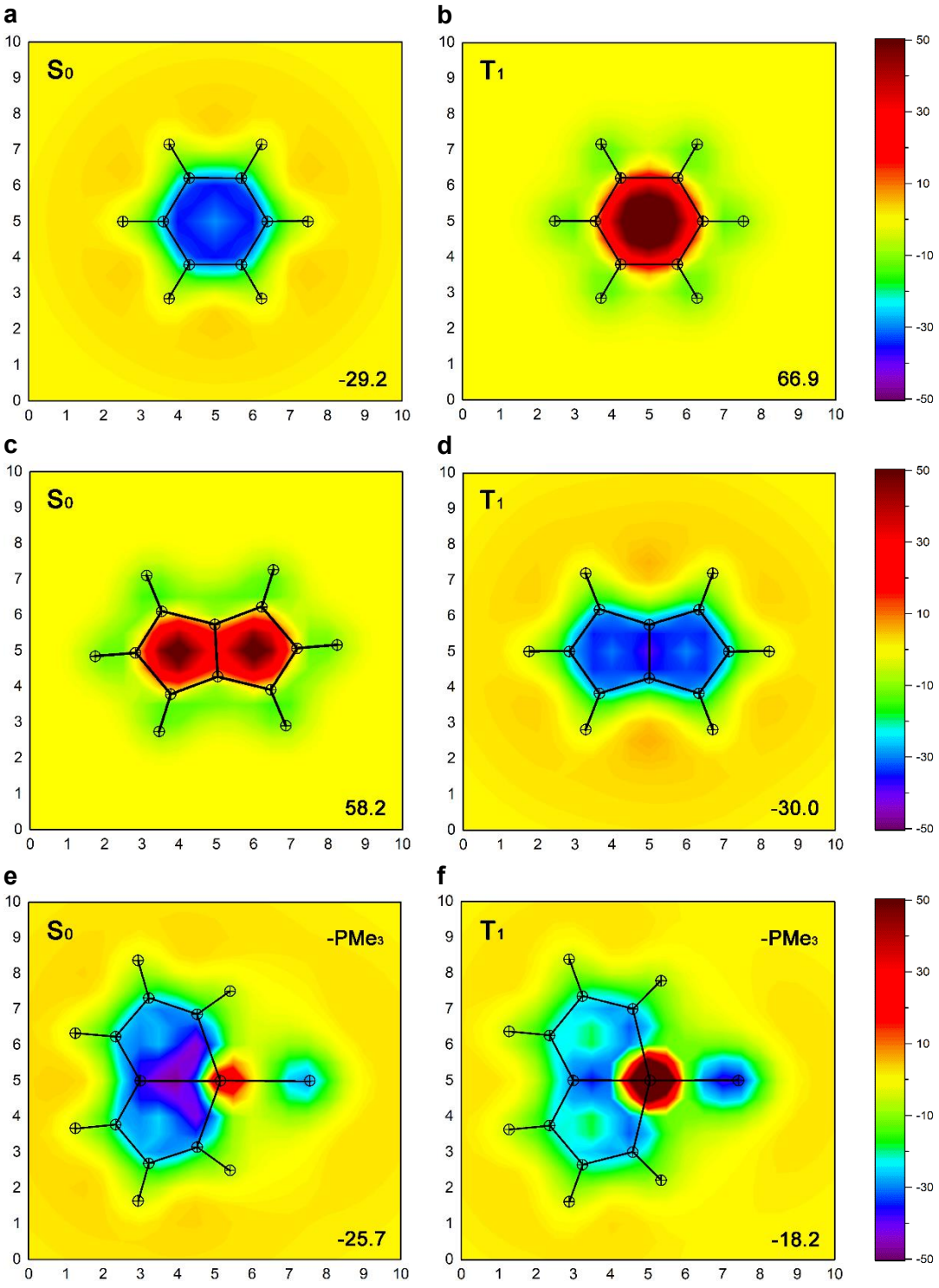
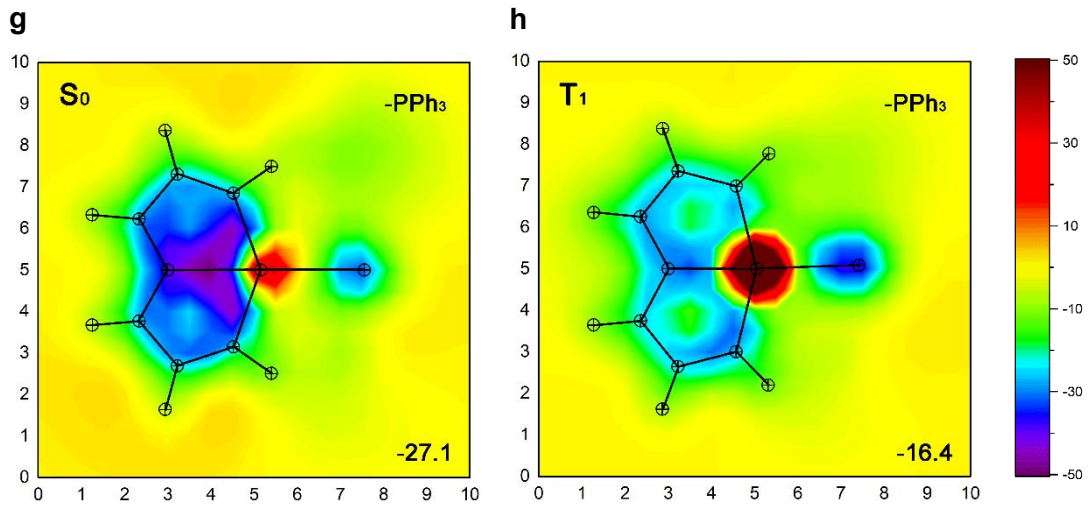
	1	2	3
S_0	0.933	0.956	0.935
T_1	0.721	0.950	0.801
ΔHOMA	0.212	0.006	0.133
Aromaticity	reversed	adaptive	neutralized

Supplementary Table 1. HOMA values of 1, 2, and 3 in the S_0 and the T_1 states. The HOMA values were calculated using equation $\text{HOMA}=1-\frac{\alpha}{n}\sum_{i=1}^{n}(R_i-R_{opt})^2$, where n is the number of C-C bonds in the metallacycle, R_{opt}(=1.388) is the optimal value for C-C bond, α(=257.7) is an empirical constant that determines the range of HOMA index, and R_i corresponds to individual C-C bond lengths[1]. Note that the HOMA method was originally developed for closed-shell systems, while in the T_1 state positive values do not necessarily indicate aromaticity (several instances have been reported for anti-aromatic systems)[2,3]. Given a specific electronic state, a larger HOMA value is supposed to arise from better equalization of C-C bond lengths. The variation of HOMA values (ΔHOMA) correlates well to the aromaticity change.

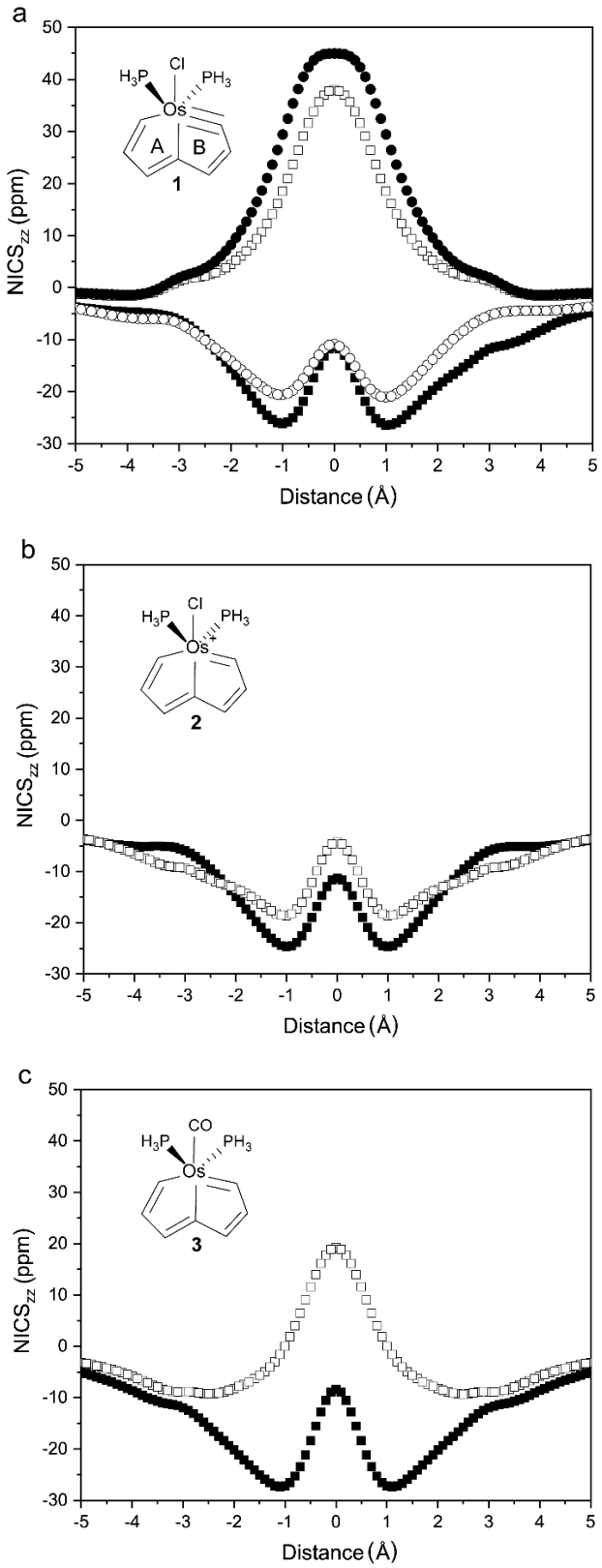


Supplementary Figure 1. Representations of NICS(1) grid setup. Each grid contains 441 grid points which are equally spaced with 0.5 Å distance. Ghost atoms were placed at grid points to probe the NICS values. The grid covers a 10 Å × 10 Å area that lies 1 Å above the molecular plane. When the grid coincides with the molecular plane, one gets a NICS(0) grid.


a
S0
-29.2
b
T1
66.9
c
S0
58.2
d
T1
-30.0
e
S0
-PMe3
-25.7
f
T1
-PMe3
-18.2
50
30
10
-10
-30
-50

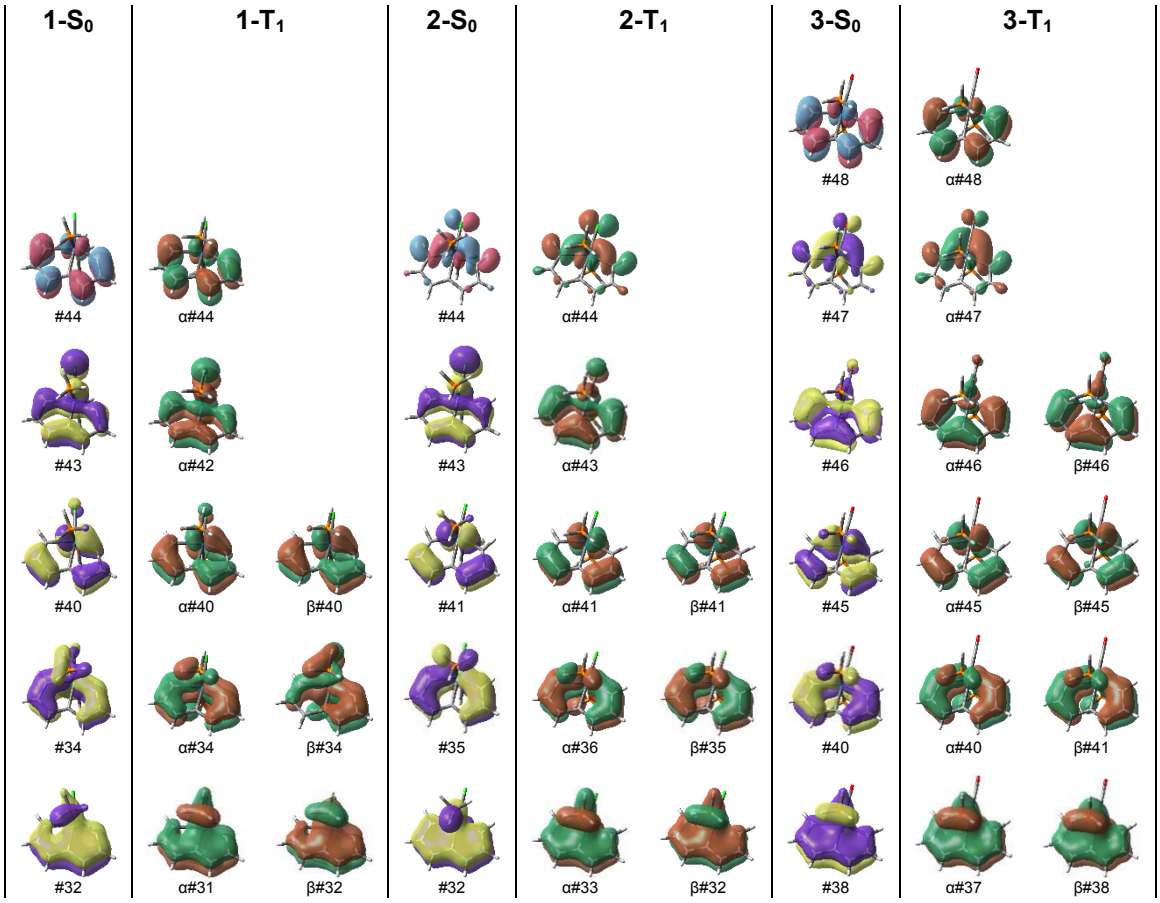


Supplementary Figure 2. NICS(1)$_{zz}$ grids for benzene, pentalene, and 16e osmapentalenes with bulkier phosphorus ligands. Ring-center NICS(1)$_{zz}$ values are commented on the bottom-right of each grid. All values are given in p.p.m. **a**-**b**, benzene; **c**-**d**, pentalene (C_8H_6, D_{2h} symmetric in both the S_0 and T_1 states); **e**-**h**, 16e osmapentalenes. To reduce computational cost, the 6-31G* instead of 6-311++G** basis set for C and H atoms was used for geometry optimizations of **e**-**h**. The 6-311++G** basis set was used for all the NMR calculations except **h** in which we used 6-311G** basis set because the 6-311++G** basis set is too large to perform the NMR calculations.

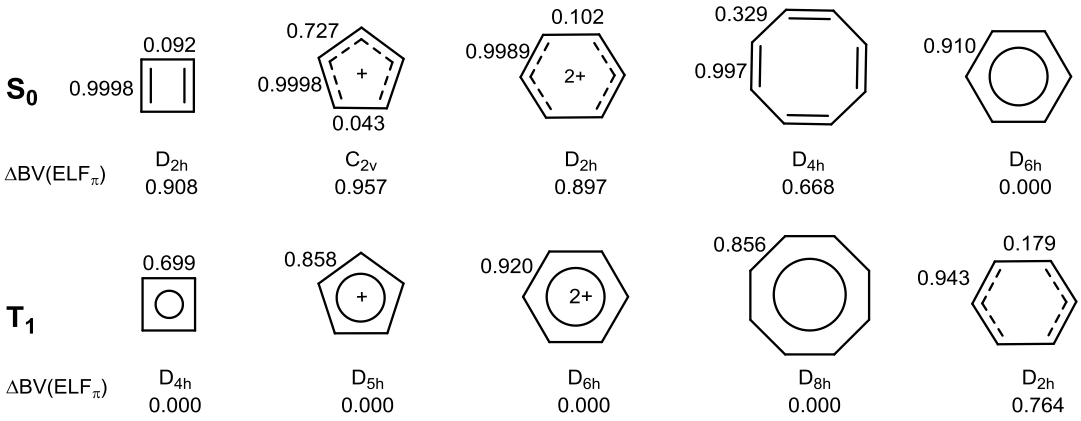


Supplementary Figure 3. NICS scans for complexes 1-3 in the S_0 and the T_1 states. a, complex **1** (S_0 A, solid squares; S_0 B, hollow circles; T_1 A, hollow squares; T_1 B, solid circles); **b**, complex

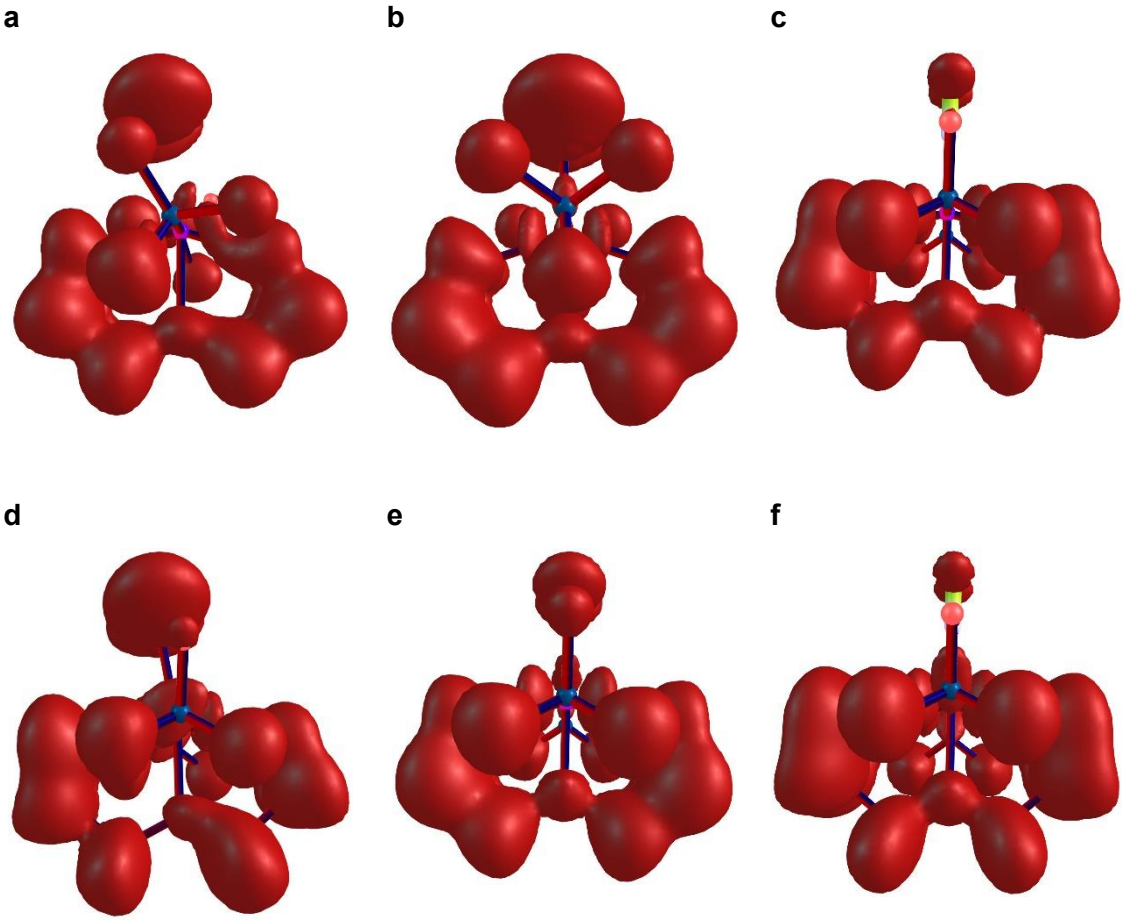
2 (S_0, solid squares; T_1, hollow squares); **c**, complex 3 (S_0, solid squares; T_1, hollow squares). Ghost atoms were placed equidistantly along the axis passing through the center of each ring, at distances to the plane ranging from 0.0 Å to 5.0 Å. Subscript "zz" represents the out-of-plane component of isotropic NICS. These $NICS_{zz}$ curves are consistent with the aromaticity in **1-S$_0$**, **2-S$_0$**, **3-S$_0$**, and **2-T$_1$**, anti-aromaticity in **1-T$_1$**, and non-aromaticity in **3-T$_1$**[4].



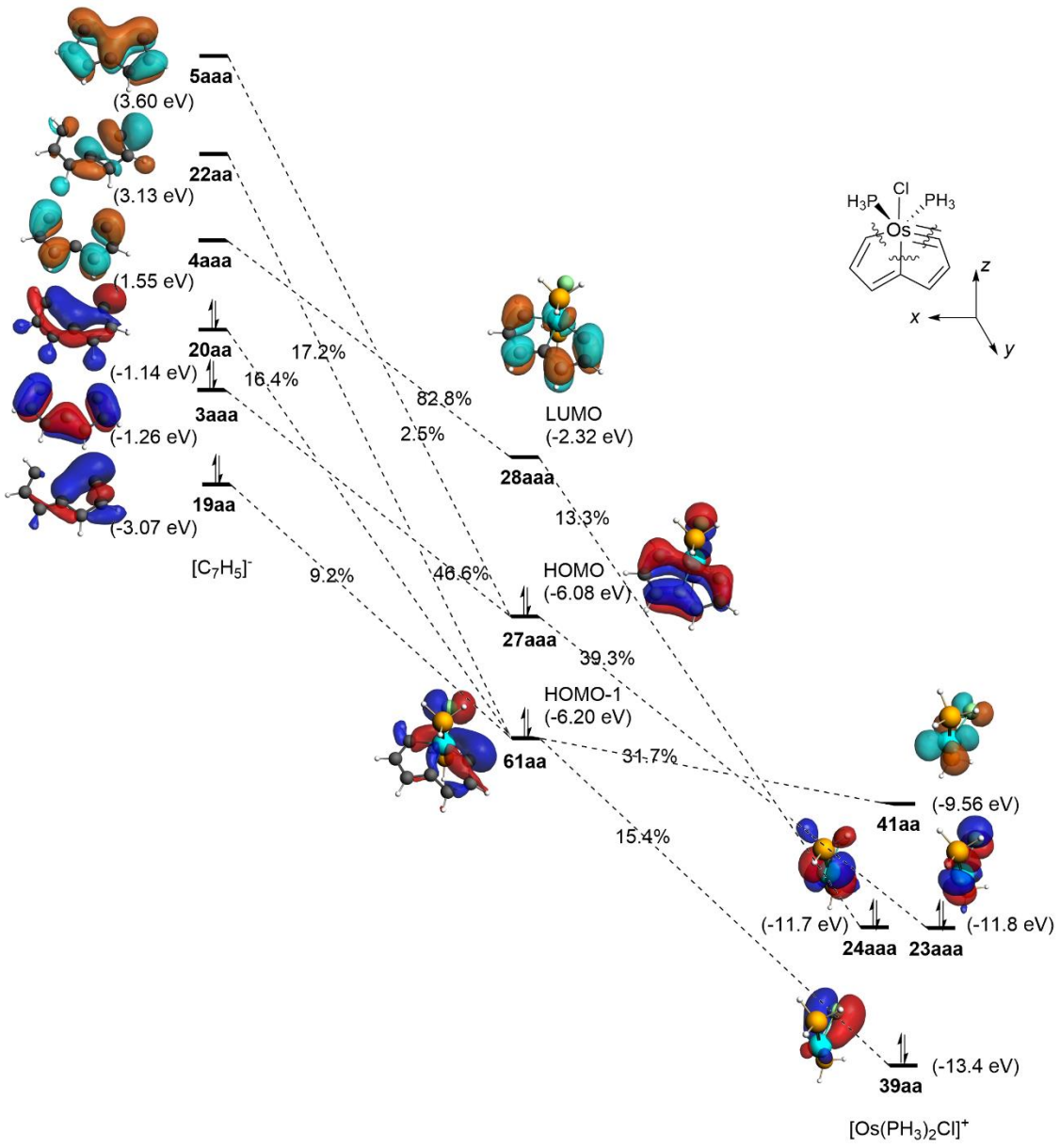
Supplementary Figure 4. Key MOs of complexes 1-3. According to previous studies, the Möbius aromaticity of complexes **1-3** in the S_0 state can be attributed to MOs #32, #34, #40, and #43 in **1-S$_0$**, #32, #35, #41, and #43 in **2-S$_0$** and #38, #40, #45, and #46 in **3-S$_0$**. The occupied out-of-plane π MOs were used in ELF_π analyses, where the occupation number for each closed-shell MO was set to 2 and for open-shell MOs the value was 1. The isovalue for surfaces is 0.03 a.u.



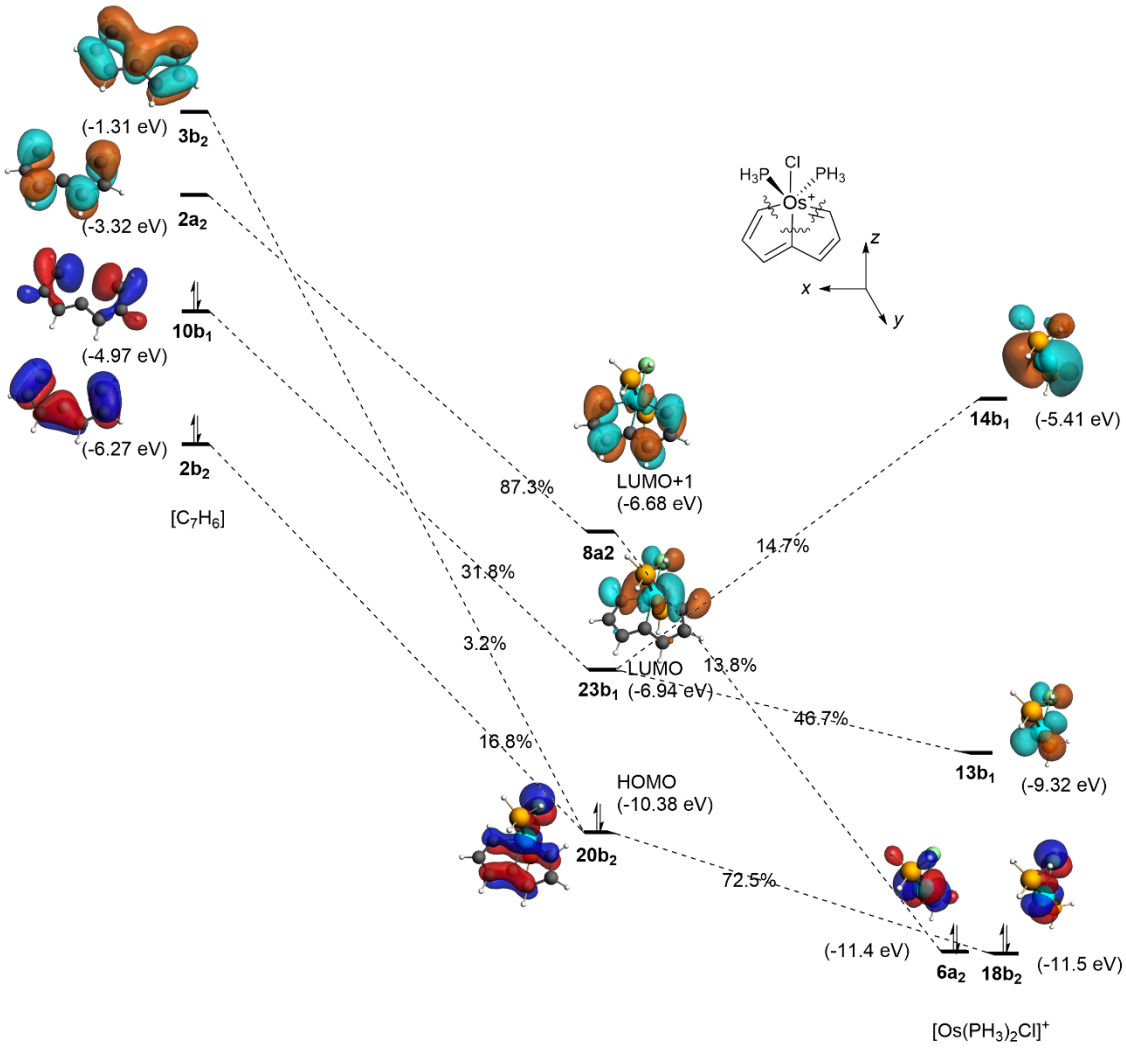
Supplementary Figure 5. ELF$_\pi$ analysis for 4*n*π species (C_4H_4, $C_5H_5^+$, $C_6H_6^{2+}$, and C_8H_8) and benzene. The ELF$_\pi$ bifurcation values are annotated along the corresponding bonds. Symmetry constraint was applied to geometry optimizations.



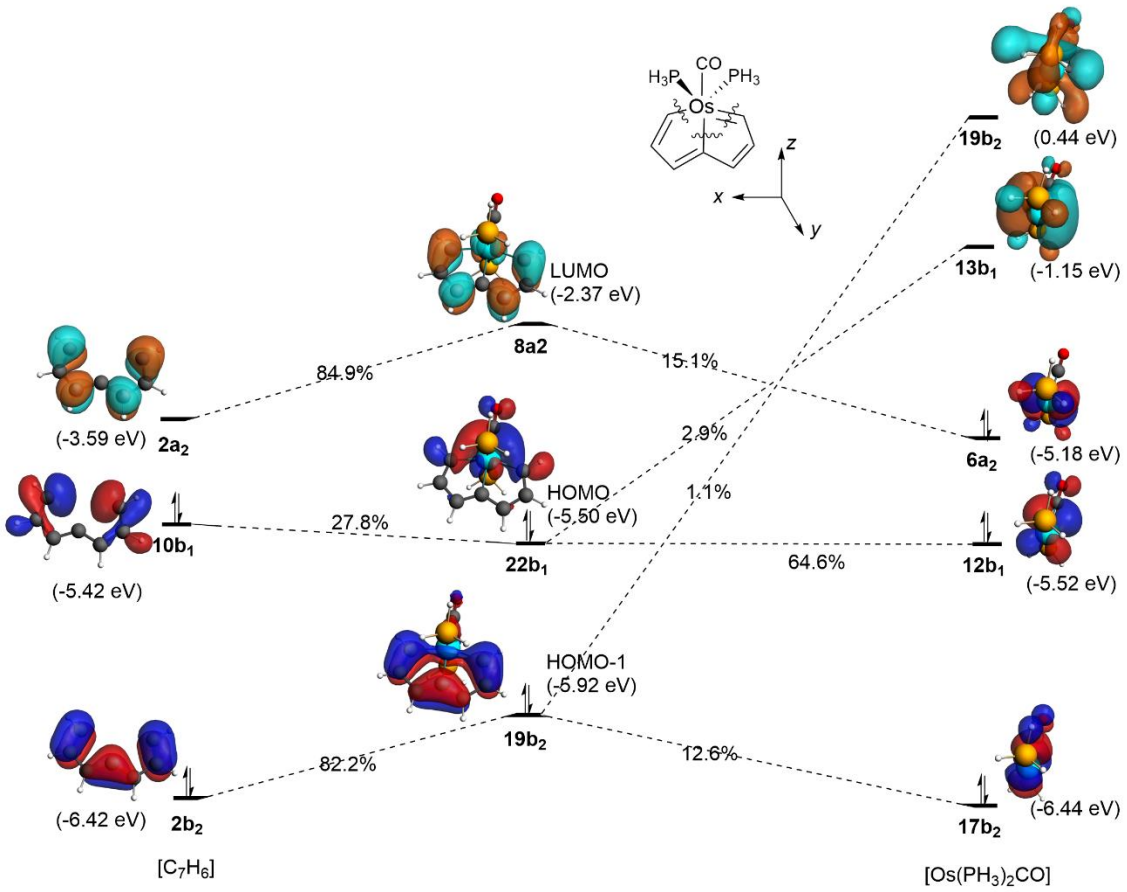
Supplementary Figure 6. ELF_{π} domains at the isovalue of 0.70. The ELF_{π} domains of C_7H_6 moiety in **1-S_0** (**a**), **2-S_0** (**b**), **3-S_0** (**c**), and **2-T_1** (**e**) are not bifurcated at this isovalue, indicating that the π electrons are highly delocalized in these systems. In contrast, π-electron delocalization in anti-aromatic **1-T_1** (**d**) and non-aromatic **3-T_1** (**f**) are particularly low, indicated by discontinuous ELF_{π} density surfaces.



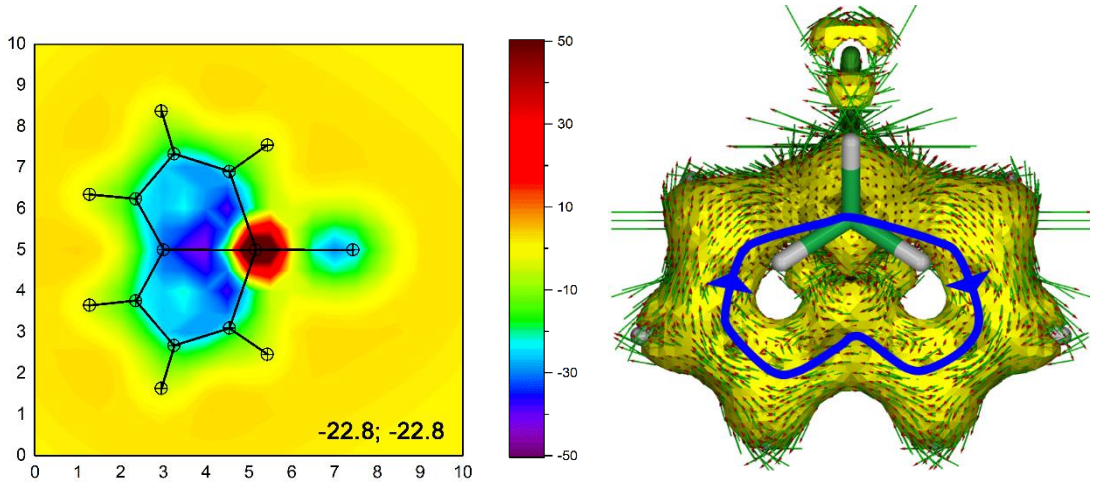
Supplementary Figure 7. Illustration of LUMO, HOMO, and HOMO-1 of 1-S_0 with major contributions from fragment molecular orbitals (FMOs). Level of theory: B3LYP/TZ2P. The isovalue for MO surfaces is 0.03 a.u. With C_s symmetry applied, all the out-of-plane π orbitals are aaa symmetric whereas the in-plane π orbitals are aa symmetric. Note that some FMOs which are also involved in the formation of these MOs are not presented due to the minor contributions.



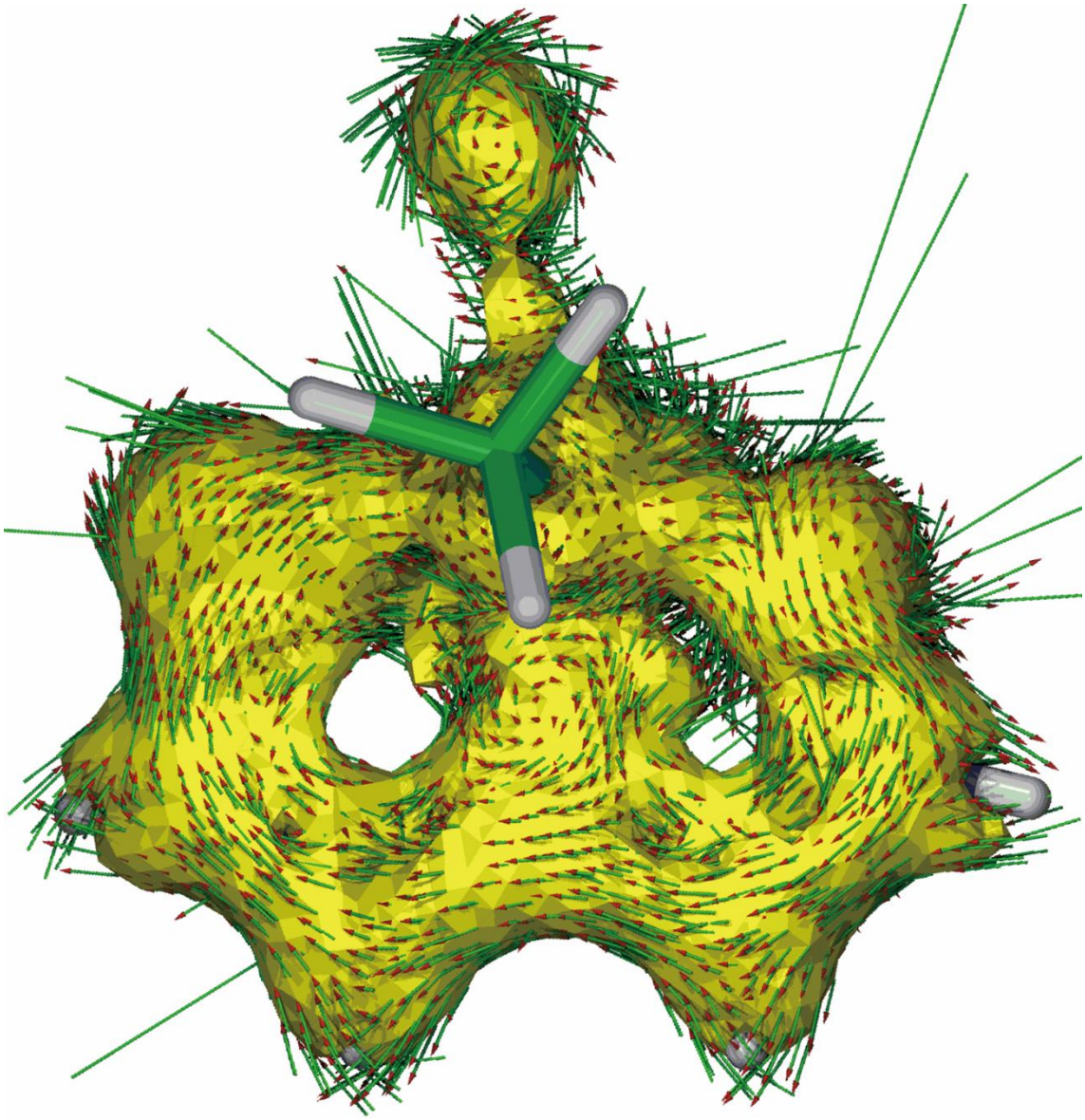
Supplementary Figure 8. Illustration of LUMO+1, LUMO, and HOMO of 2-S_0 with major contributions from FMOs. Level of theory: B3LYP/TZ2P. The isovalue for MO surfaces is 0.03 a.u. With C_{2v} symmetry applied, the out-of-plane π orbitals are a_2/b_2 symmetric whereas the in-plane π orbitals are a_1/b_1 symmetric. Note that some FMOs which are also involved in the formation of these MOs are not presented due to the minor contributions.



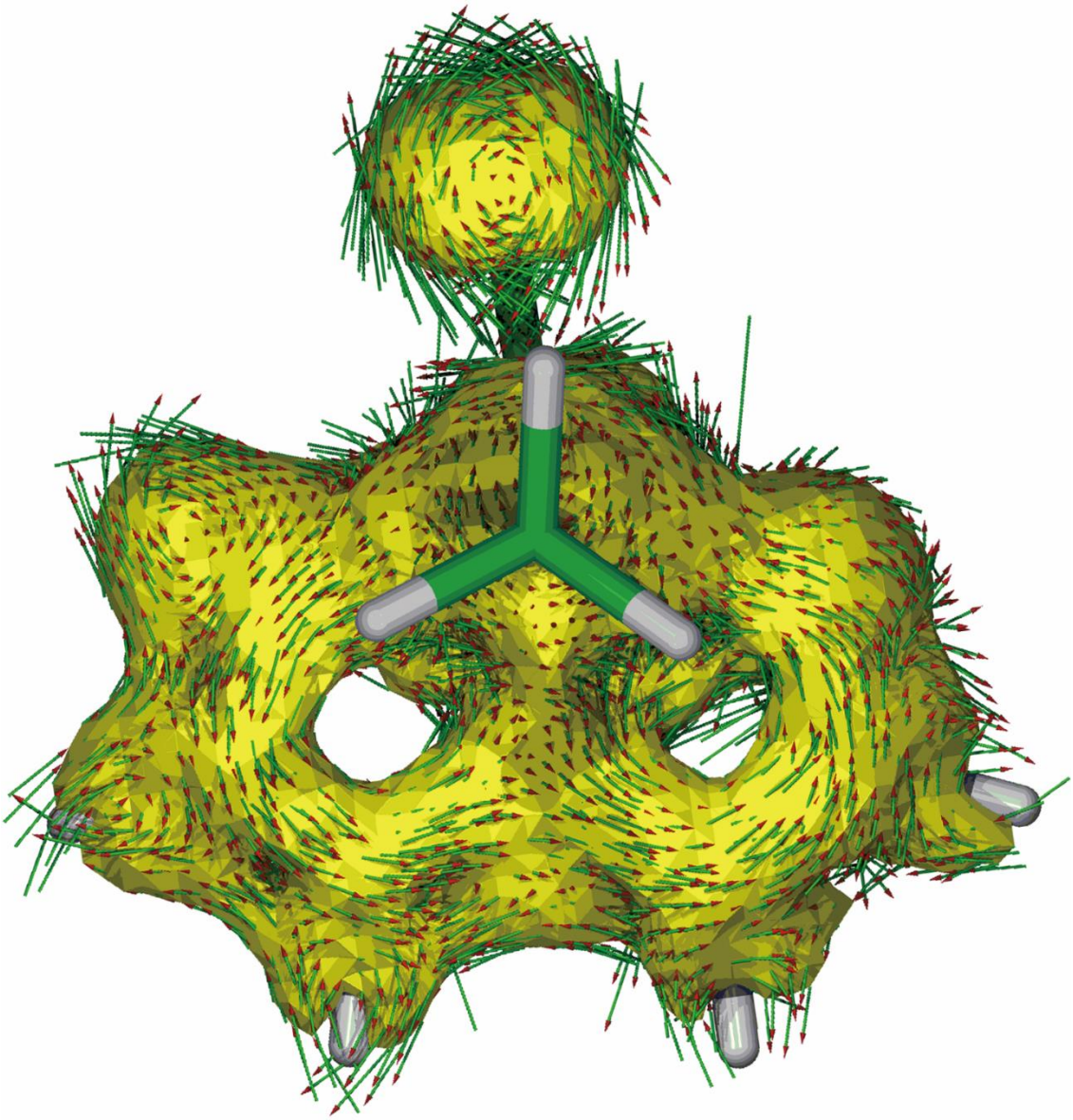
Supplementary Figure 9. Illustration of LUMO, HOMO, and HOMO-1 of 3-S_0 with major contributions from FMOs. Level of theory: B3LYP/TZ2P. The isovalue for MO surfaces is 0.03 a.u. With C_{2v} symmetry applied, the out-of-plane π orbitals are a_2/b_2 symmetric whereas the in-plane π orbitals are a_1/b_1 symmetric. Note that some FMOs which are also involved in the formation of these MOs are not presented due to the minor contributions. The HOMO-1 of **1-S_0**, LUMO of **2-S_0**, and HOMO of **3-S_0** are very similar, all generated from the interaction between an in-plane antibonding FMO of the OsL_3 (L, ligand) moiety and one σ FMO of the hydrocarbon moiety (Figs. S7-S9). Complexes **2** and **3** have the same arrangement of the listed MOs, whereas the HOMO-1 of **1-S_0** appears to be stabilized by the in-plane lone pair of the *sp* hybridized carbon. Since the osmium in **3** has two more valence electrons than **2**, the LUMO of **2-S_0** becomes the HOMO of **3-S_0**.



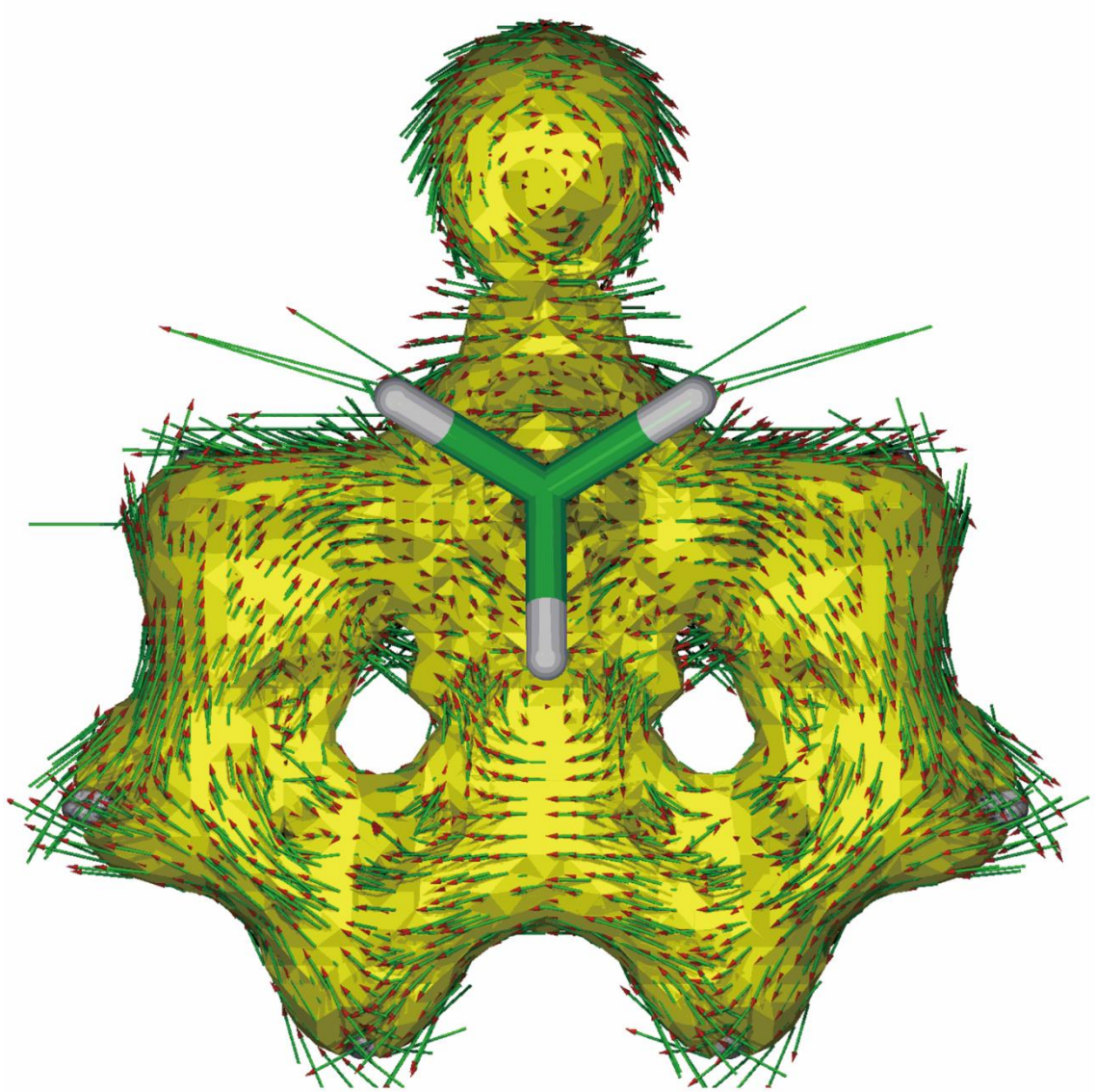
Supplementary Figure 10. NICS(1)$_{zz}$ grid and total ACID plot for radical dication 2′. Ring-center NICS(1)$_{zz}$ values are commented on the bottom-right of the grid. The isovalue for ACID surface is 0.035 a.u. Calculations were performed in the doublet ground state (D_0) for this species.



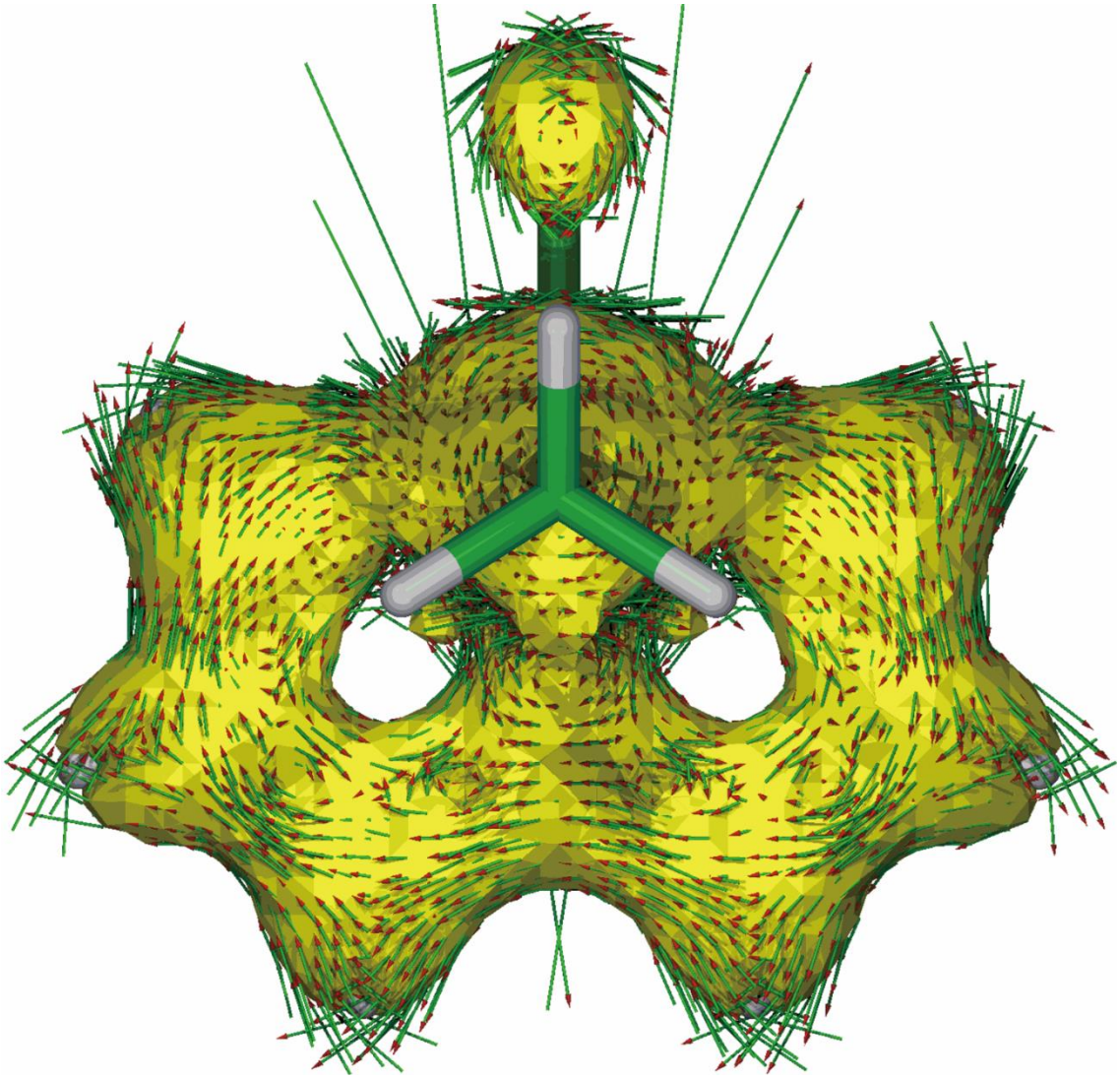
Supplementary Figure 11. ACID plot of 1-S_0. Isovalue: 0.035 a.u.



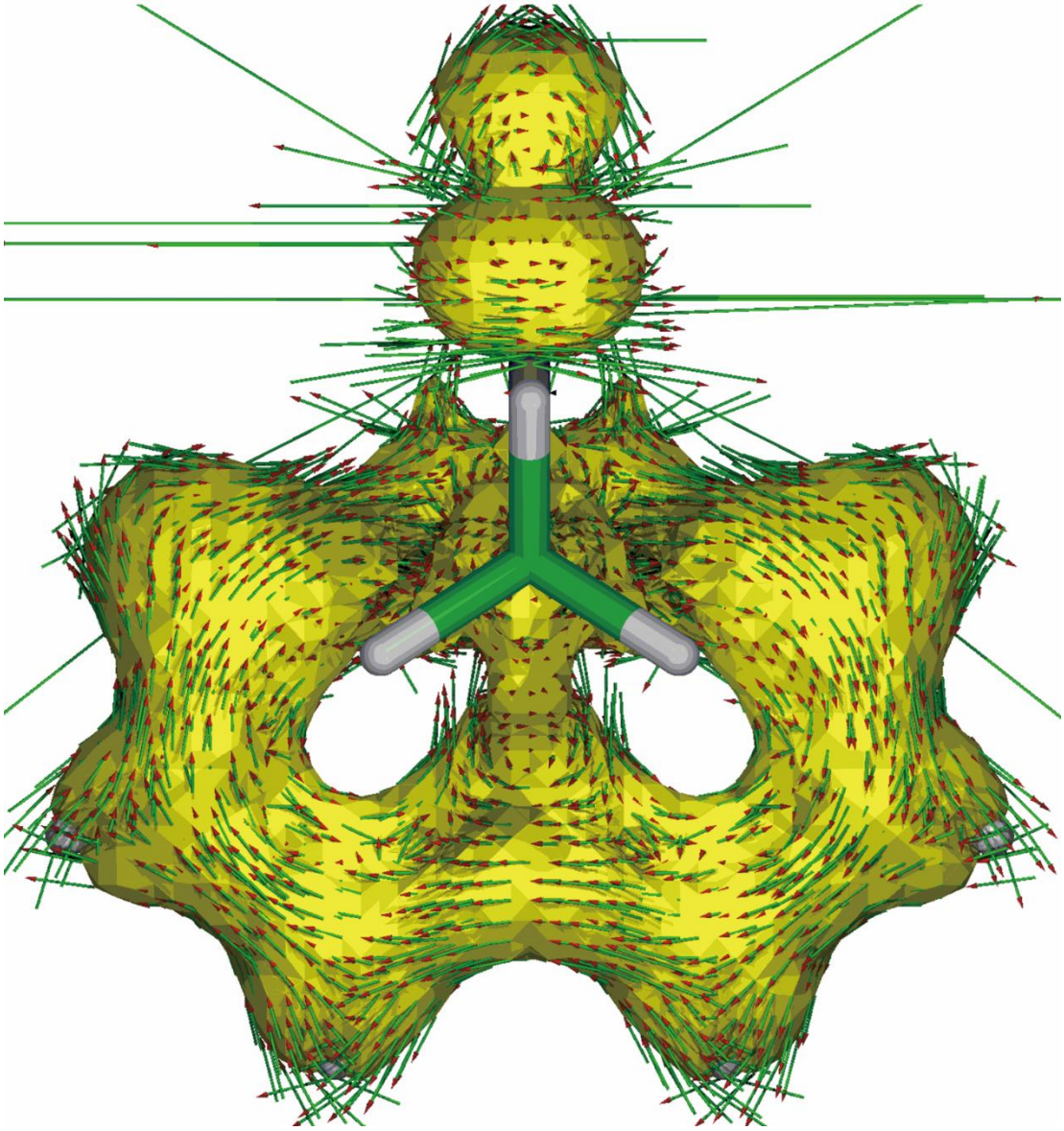
Supplementary Figure 12. ACID plot of 1-T_1. Isovalue: 0.035 a.u.



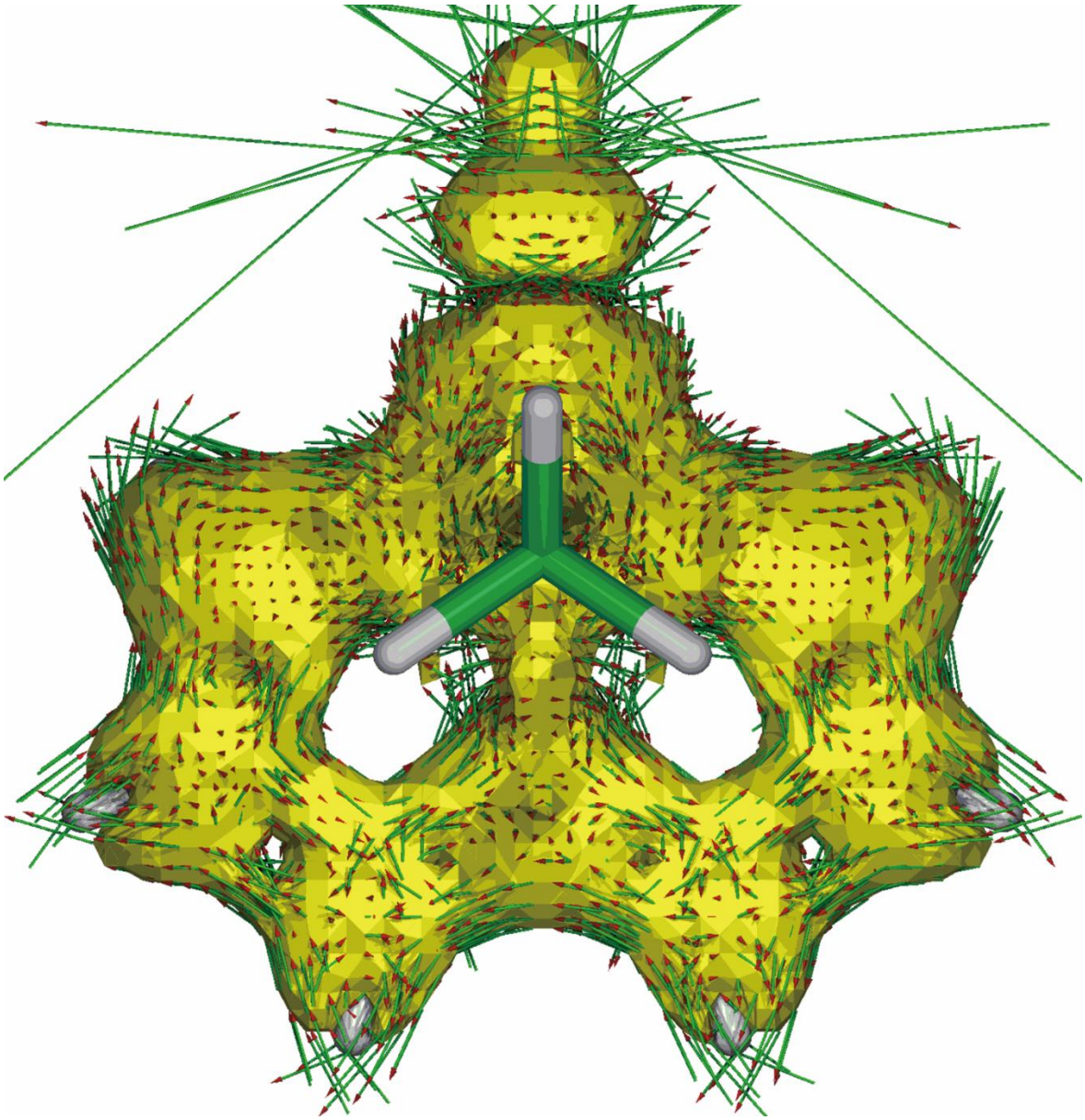
Supplementary Figure 13. ACID plot of 2-S_0. Isovalue: 0.035 a.u.



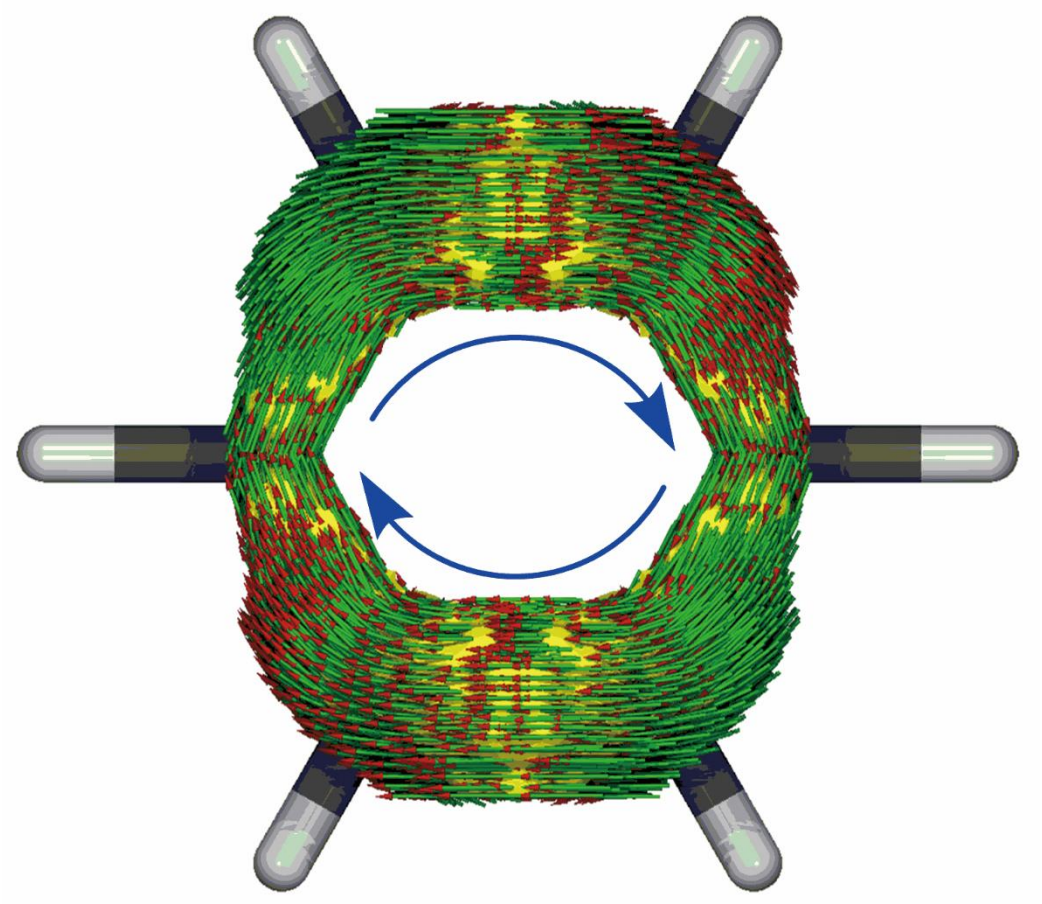
Supplementary Figure 14. ACID plot of 2-T_1. Isovalue: 0.035 a.u.



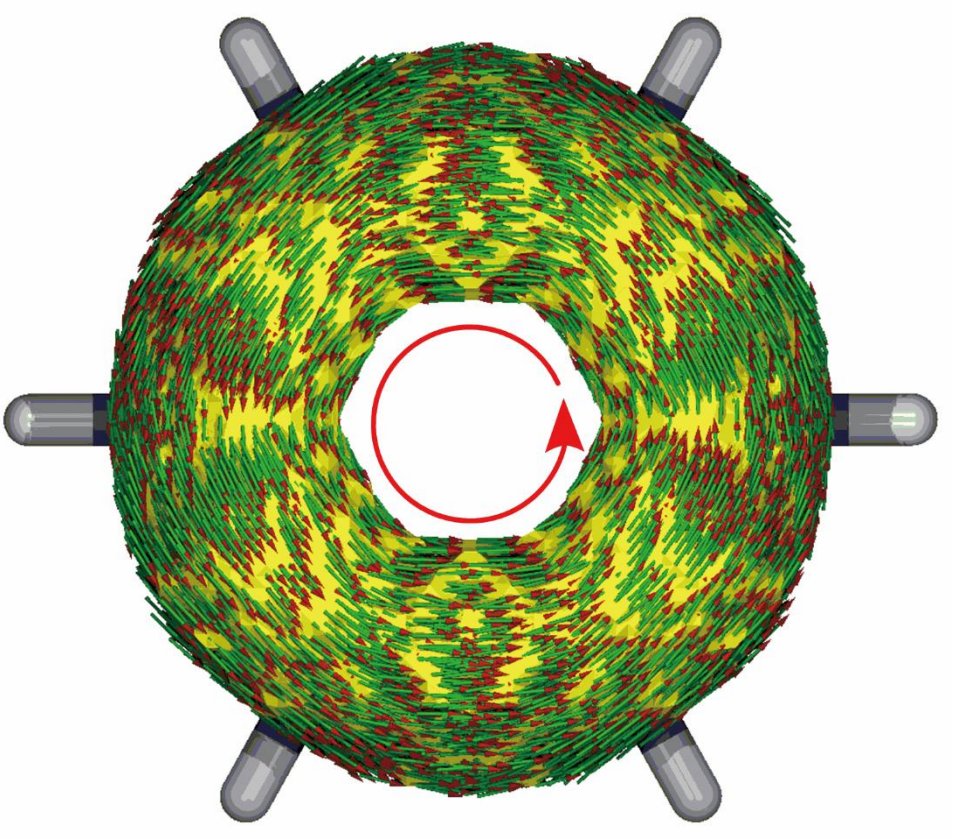
Supplementary Figure 15. ACID plot of 3-S_0. Isovalue: 0.035 a.u.



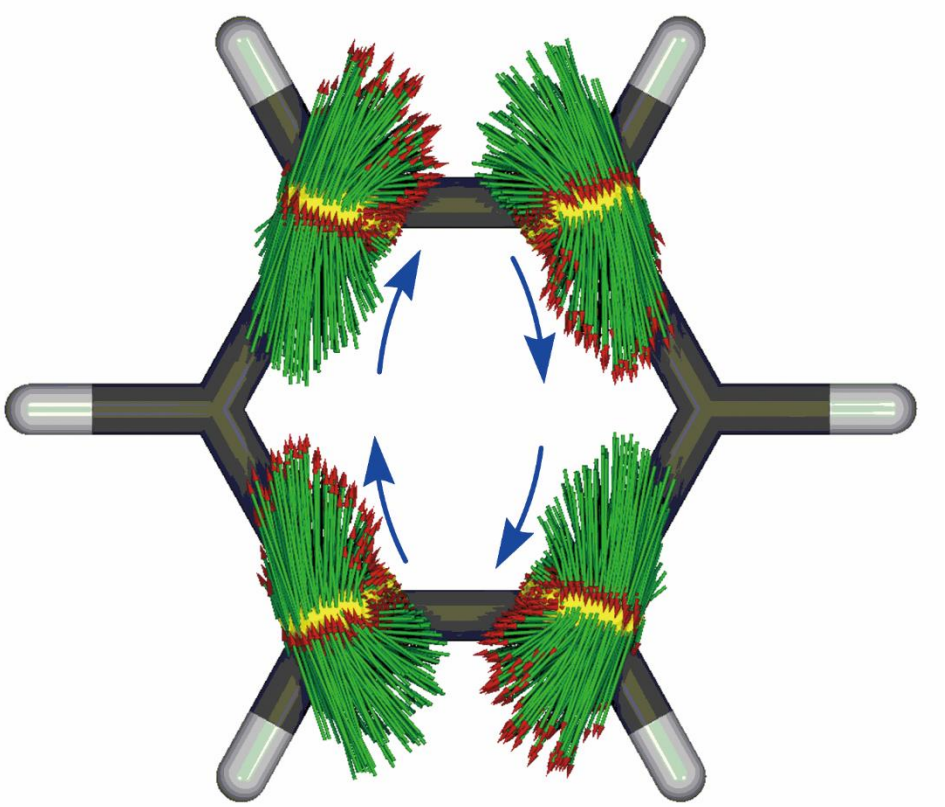
Supplementary Figure 16. ACID plot of 3-T_1. Isovalue: 0.035 a.u.



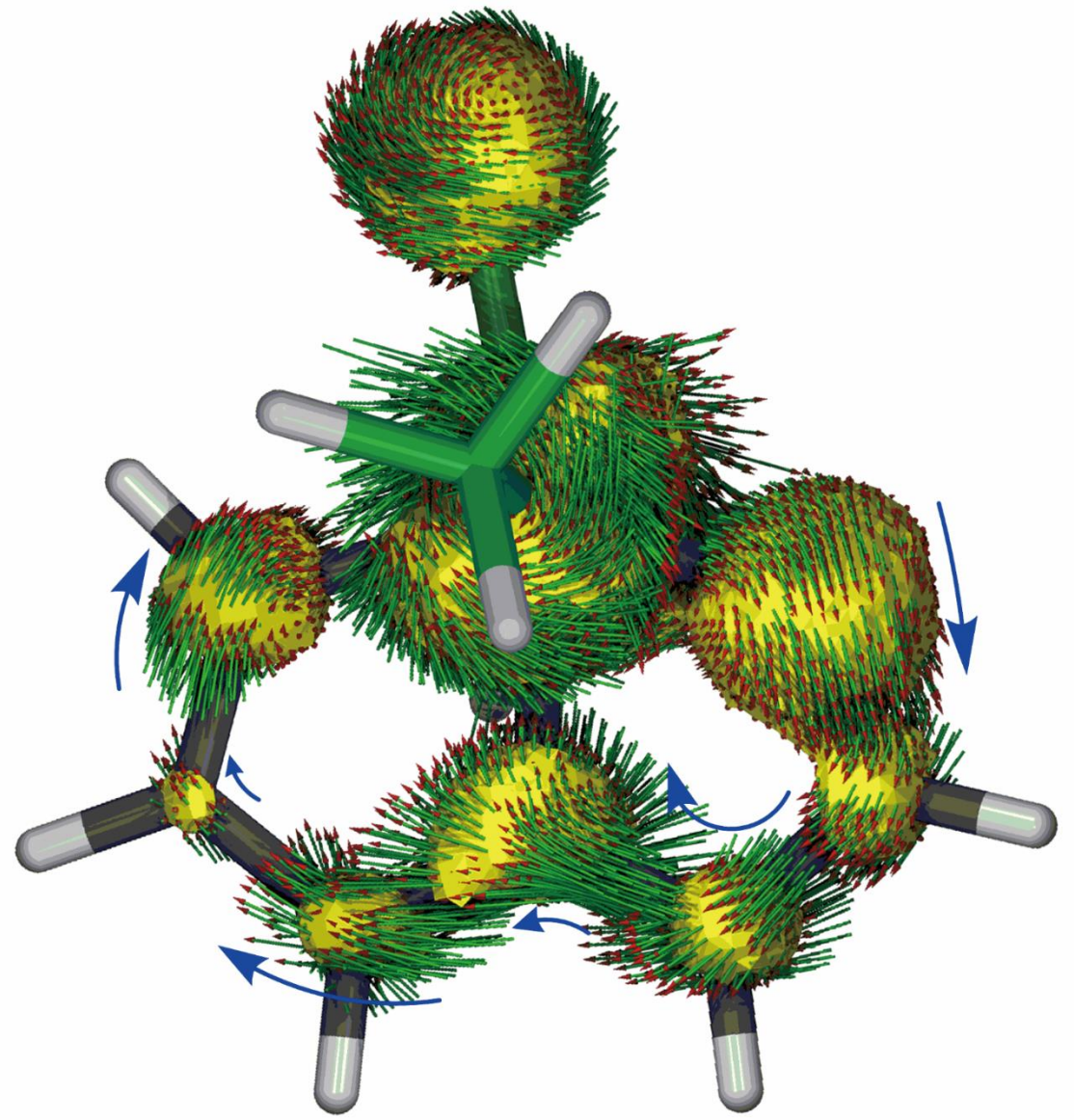
Supplementary Figure 17. ACID plot of the HOMO of benzene in the S_0 state. Isovalue: 0.024 a.u.



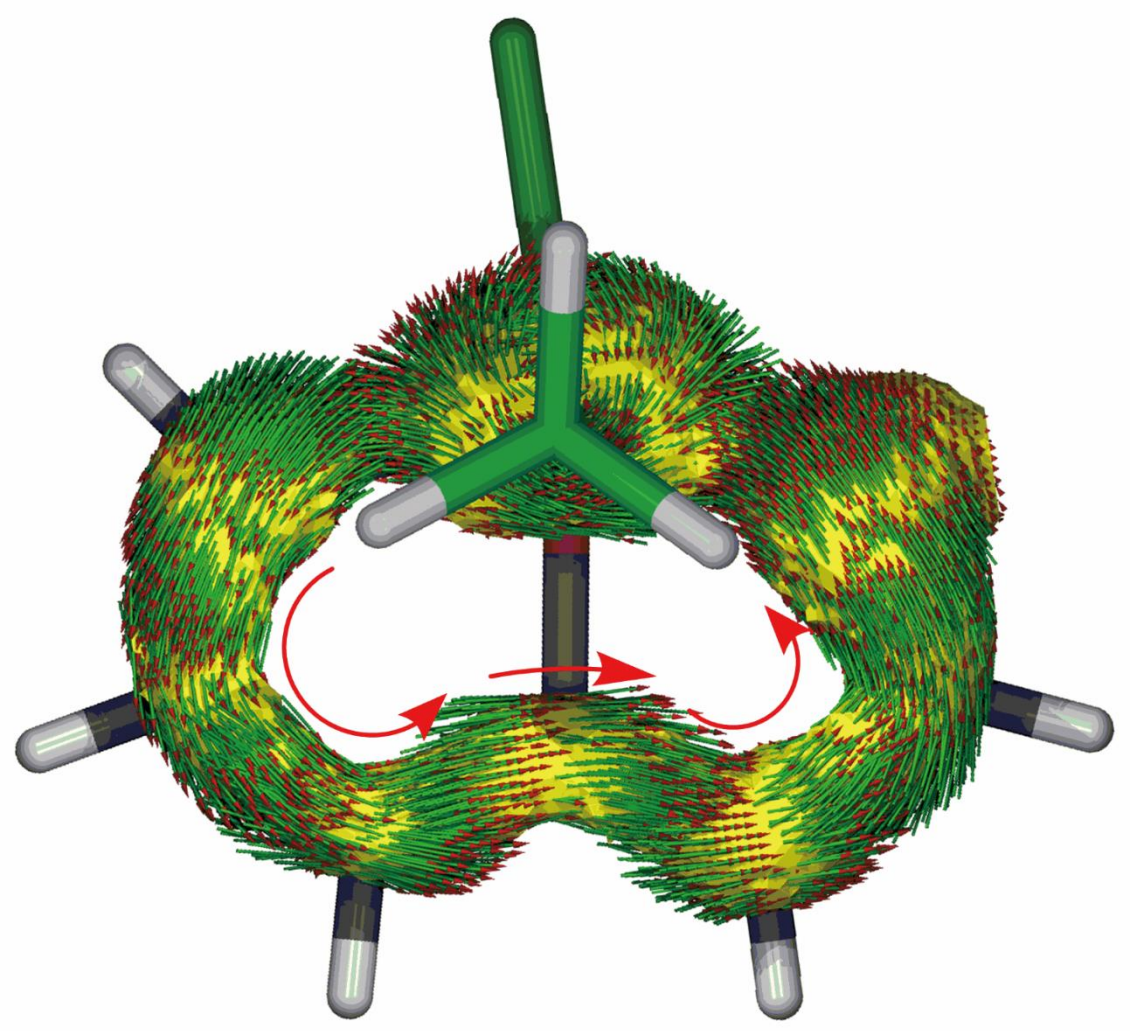
Supplementary Figure 18. ACID plot of the HSOMO of benzene in the T_1 state. Isovalue: 0.024 a.u.



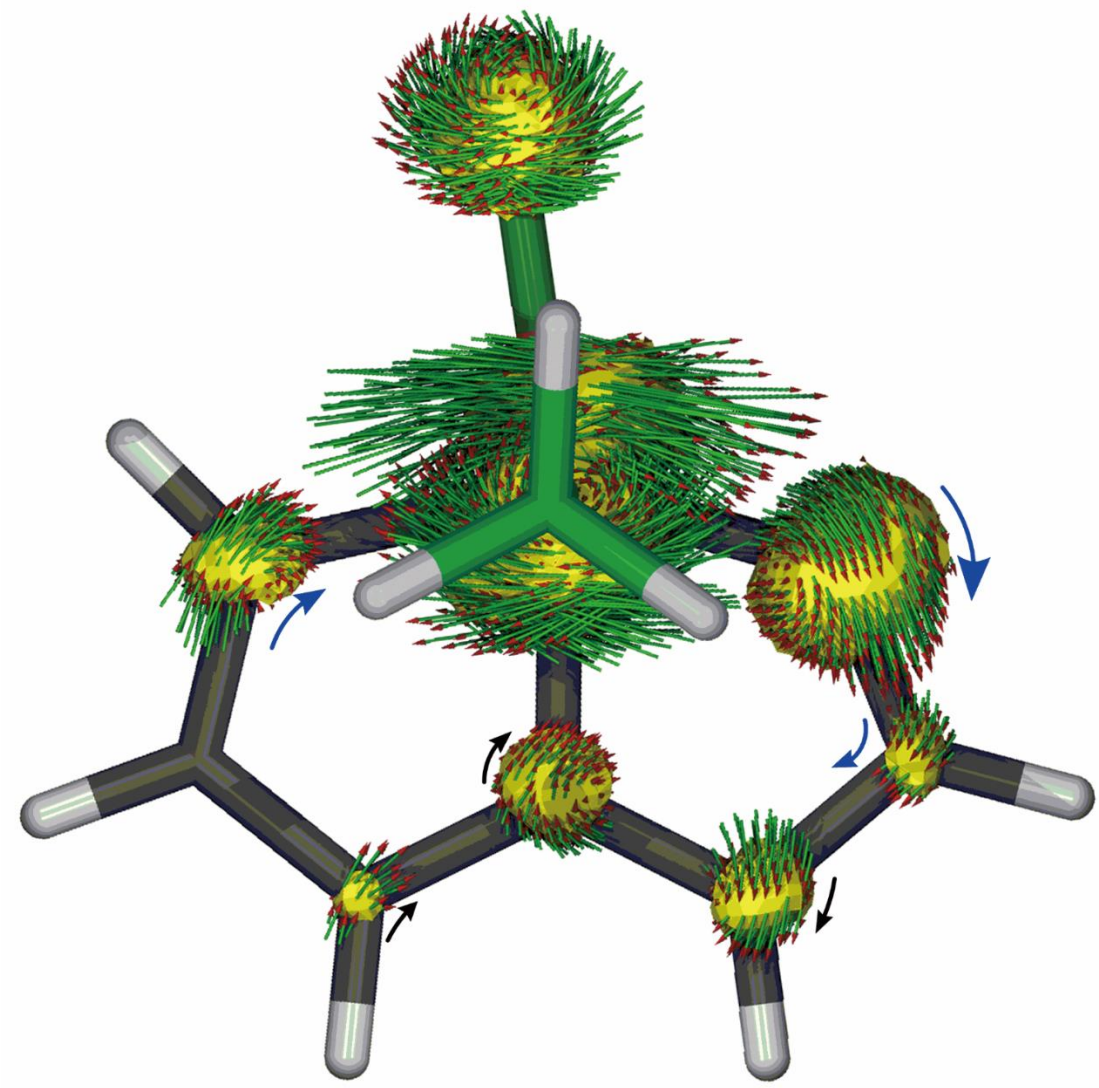
Supplementary Figure 19. ACID plot of the HSOMO-1 of benzene in the T_1 state. Isovalue: 0.024 a.u.



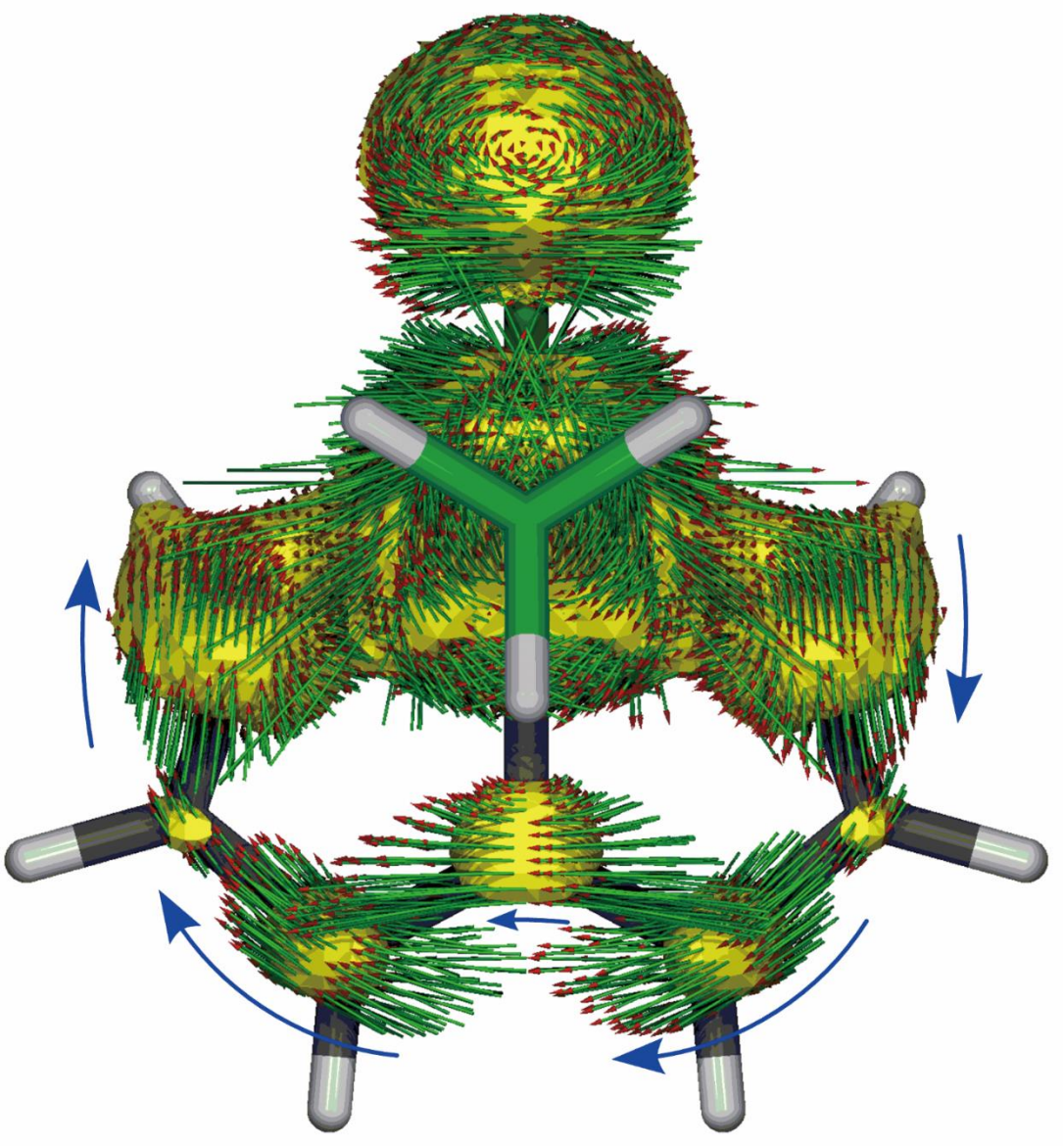
Supplementary Figure 20. ACID plot of the HOMO of 1-S_0. Isovalue: 0.024 a.u.



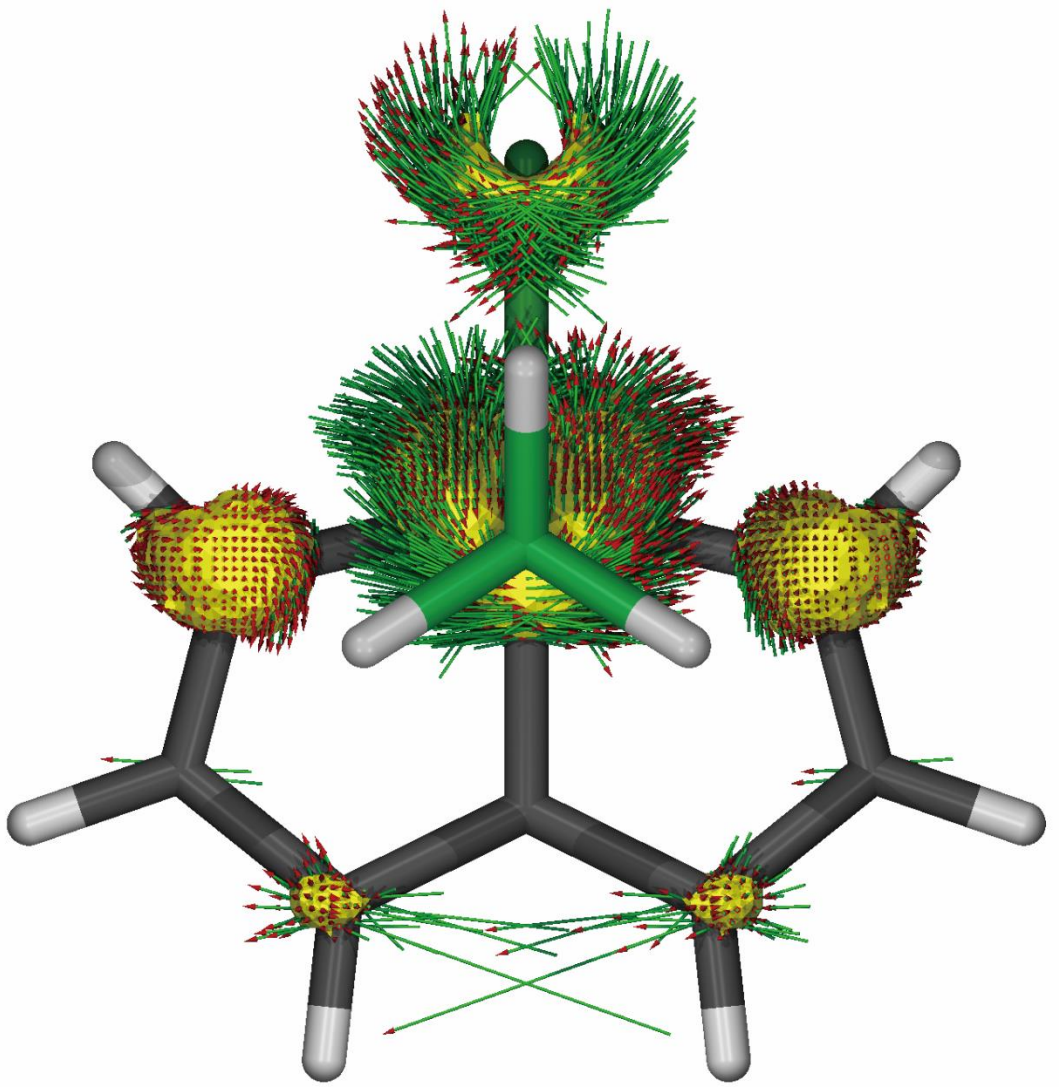
Supplementary Figure 21. ACID plot of the HSOMO of 1-T_1. Isovalue: 0.024 a.u.



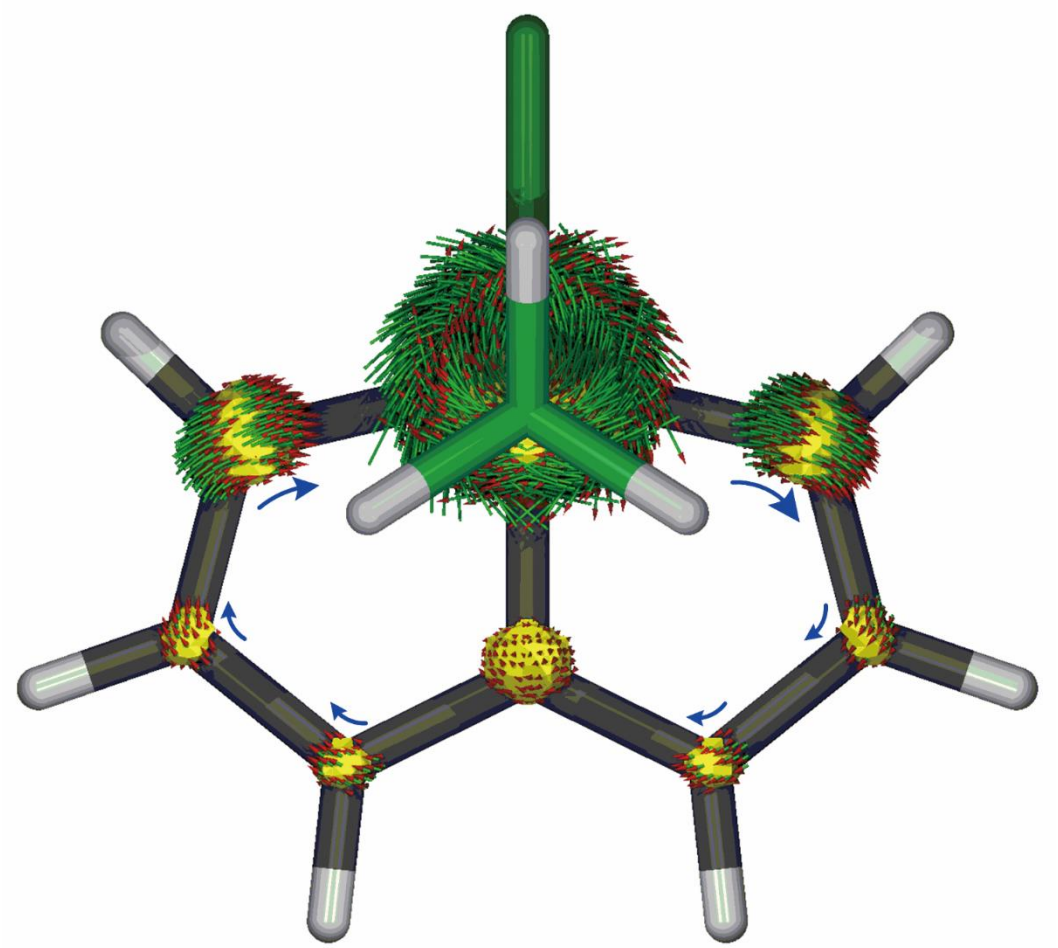
Supplementary Figure 22. ACID plot of the HSOMO-1 of 1-T_1. Isovalue: 0.024 a.u.



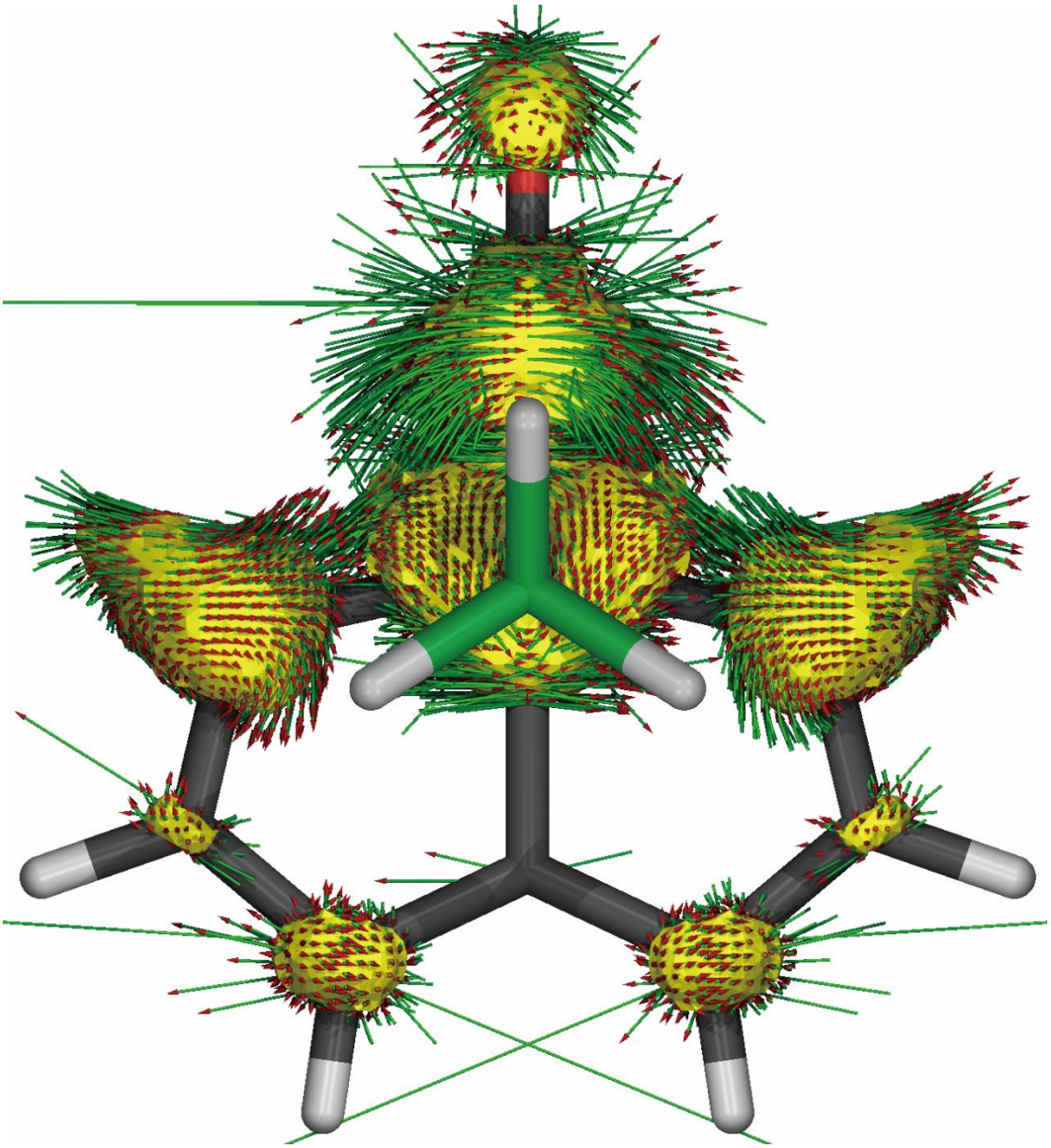
Supplementary Figure 23. ACID plot of the HOMO of 2-S_0. Isovalue: 0.024 a.u.



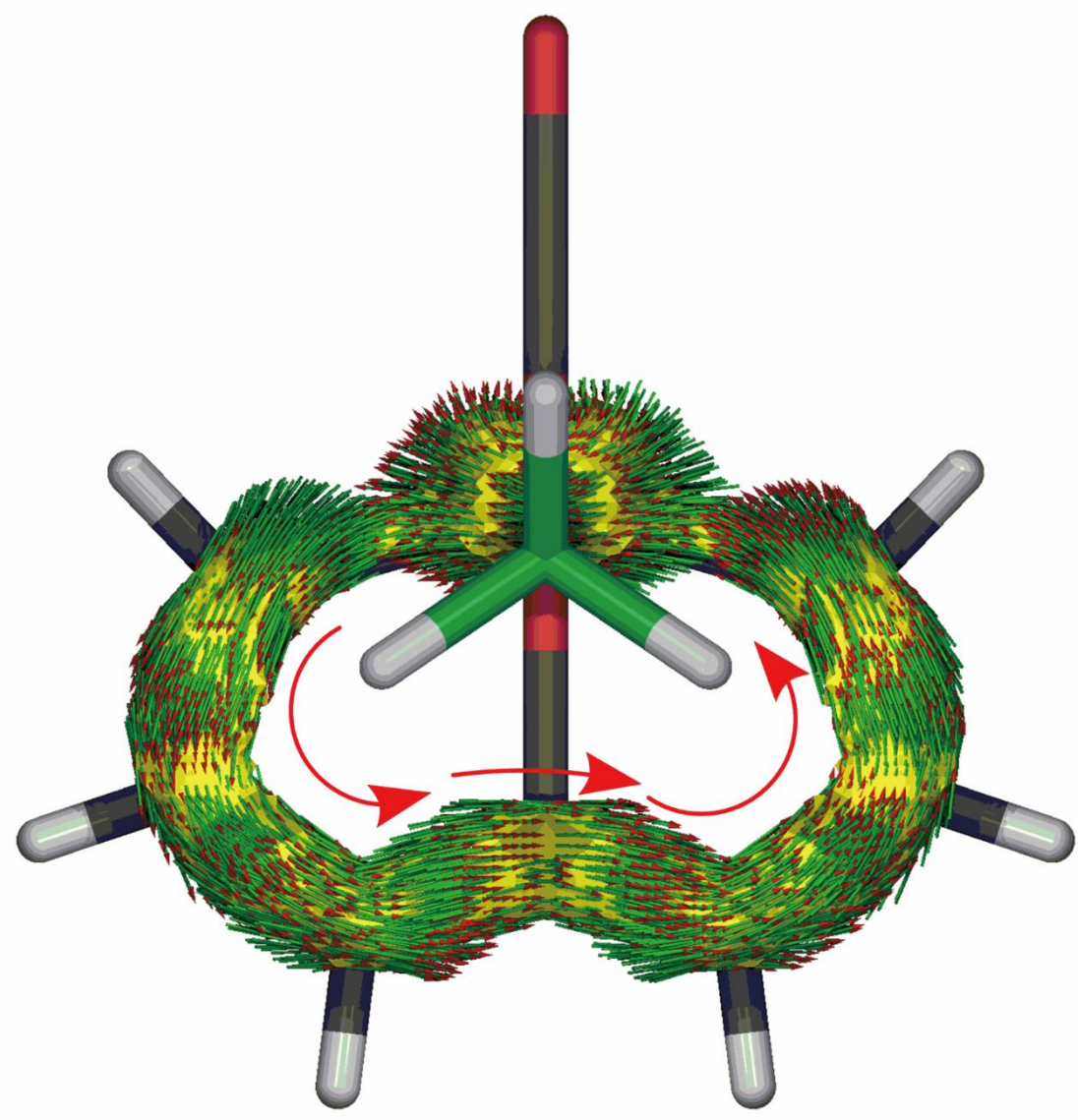
Supplementary Figure 24. ACID plot of the HSOMO of 2-T_1. Isovalue: 0.024 a.u.



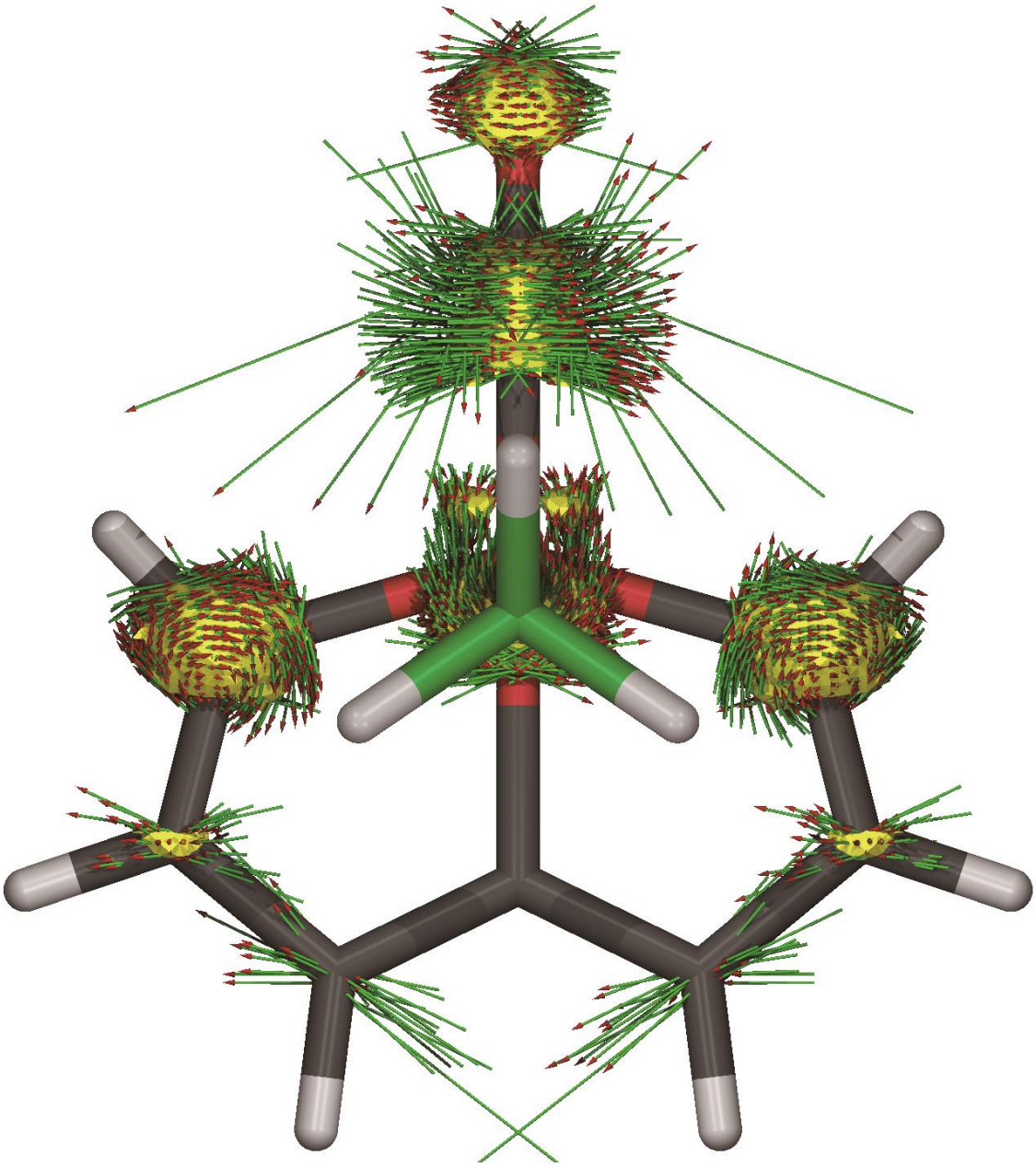
Supplementary Figure 25. ACID plot of the HSOMO-1 of 2-T_1. Isovalue: 0.024 a.u.



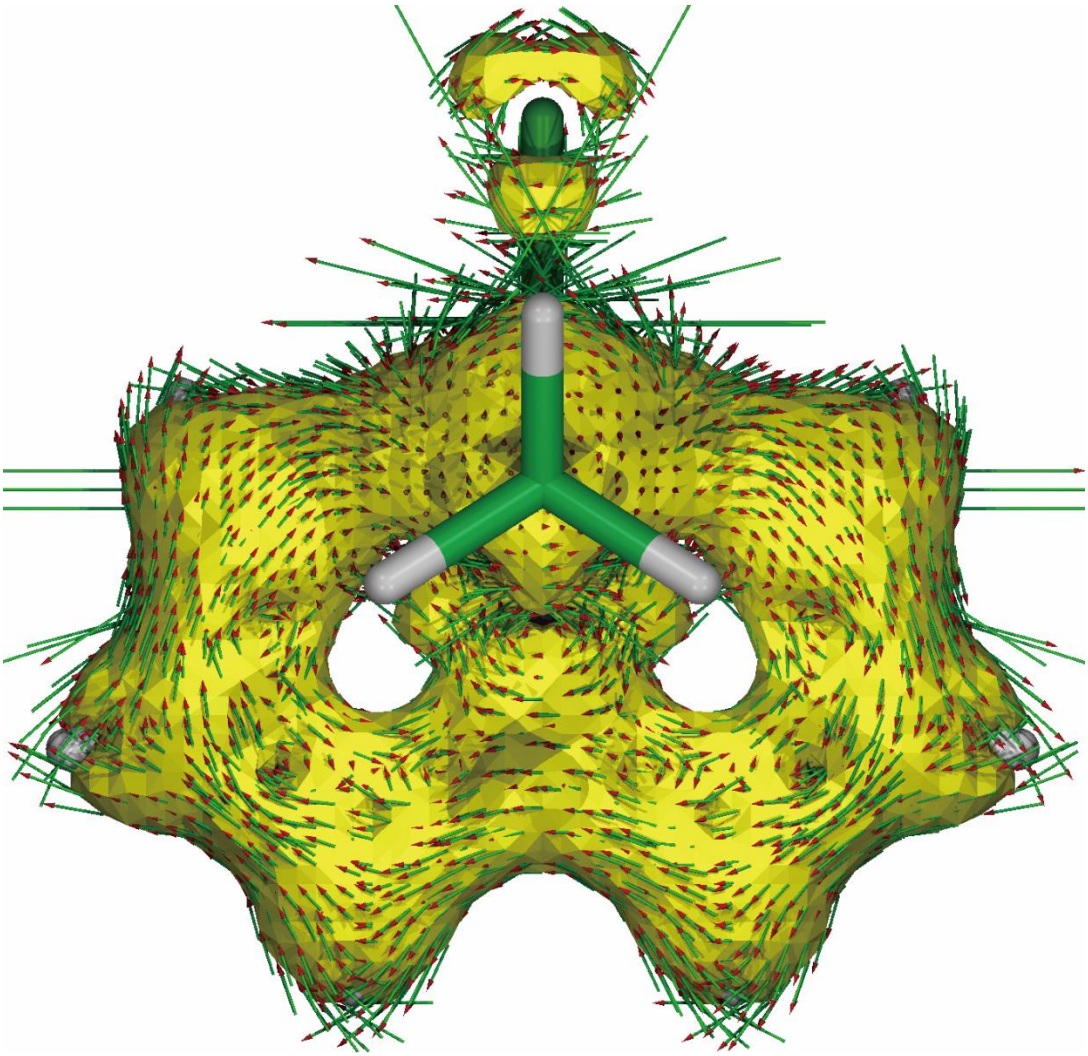
Supplementary Figure 26. ACID plot of the HOMO of 3-S_0. Isovalue: 0.024 a.u.



Supplementary Figure 27. ACID plot of the HSOMO of 3-T_1. Isovalue: 0.024 a.u.



Supplementary Figure 28. ACID plot of the HSOMO-1 of 3-T_1. Isovalue: 0.024 a.u.



Supplementary Figure 29. ACID plot of 2'. Isovalue: 0.035 a.u.

Complexes	Electronic Energy	Coordinates			
1-S_0	E = -392.461065623 a.u.	Os	-0.20502925	-0.00196346	-0.13306109
		P	-0.60114251	-2.32822923	0.22066365
		C	0.62406603	-0.34582343	-1.74430006
		C	1.97770235	-0.52531835	-2.03027428
		C	2.69632865	-0.37893670	-0.84136176
		H	3.78011521	-0.46023851	-0.77053196
		C	1.85704297	-0.11225177	0.26539834
		C	2.26611435	0.08282994	1.57441514
		H	3.31115548	0.05355413	1.87590483
		Cl	-2.67771695	0.20815998	-0.08530888
		H	-1.47504455	-2.97556739	-0.69384176
		H	0.50115677	-3.23087586	0.19058455
		H	-1.19669437	-2.71715638	1.45118587
		C	-0.04352015	0.31243686	1.89827747
		C	1.20719728	0.31864246	2.48062864
		H	-0.92672103	0.47825655	2.51710965
		P	-0.06882061	2.36389130	-0.35182476
		H	0.15825138	2.93871107	-1.63432612
		H	-1.22377283	3.08985033	0.04623141
		H	0.93613291	3.04165487	0.39598872
		H	2.40054487	-0.73479458	-3.00584833
		H	1.37814559	0.48270373	3.54115294
1-T_1	E = -392.389256653 a.u.	Os	-0.25629500	0.00005900	-0.11761800
		P	-0.14117100	-2.38452500	-0.02648500
		C	0.50656100	0.00027000	-1.85471800
		C	1.82797300	0.00015800	-2.18723600
		C	2.59044600	-0.00010600	-0.97292400
		H	3.67804200	-0.00017800	-0.94964700
		C	1.77487200	-0.00019000	0.18973000
		C	2.22741500	-0.00040400	1.55021900
		H	3.28333800	-0.00046200	1.80679000
		Cl	-2.68428700	0.00007900	-0.10223600
		H	-1.33667600	-3.12118200	-0.25099500
		H	0.73486300	-3.02255500	-0.94787700
		H	0.29660100	-2.96702800	1.19451600
		C	-0.08311800	-0.00020600	2.01627200
		C	1.19993000	-0.00039800	2.51639500
		H	-0.95031000	-0.00016500	2.67396100
		P	-0.14081600	2.38455700	-0.02602200
		H	0.73512600	3.02259600	-0.94749400
		H	-1.33623700	3.12146900	-0.25016900
		H	0.29726800	2.96680200	1.19499300
		H	2.24782700	0.00021700	-3.18599000
		H	1.42679100	-0.00054100	3.58007300

2-S_0	E = -392.854840453 a.u.	Os	0.00000000	0.00000000	0.25561500
		P	0.00000000	2.42447200	0.34041800
		C	1.87404600	-0.00252600	-0.37099400
		C	2.31435200	-0.00360200	-1.66928300
		C	1.22768300	-0.00202000	-2.55549900
		H	1.32434000	-0.00235100	-3.63707900
		C	0.00000000	0.00000000	-1.89645000
		C	-1.22768300	0.00202000	-2.55549900
		H	-1.32434000	0.00235100	-3.63707900
		Cl	0.00000000	0.00000000	2.63454900
		H	1.09884000	3.02367500	1.00906100
		H	-0.00722500	3.13490100	-0.88838400
		H	-1.09217800	3.02218500	1.02113100
		C	-1.87404600	0.00252600	-0.37099400
		C	-2.31435200	0.00360200	-1.66928300
		H	-2.53207300	0.00297200	0.50591100
		P	0.00000000	-2.42447200	0.34041800
		H	1.09217800	-3.02218500	1.02113100
		H	-1.09884000	-3.02367500	1.00906100
		H	0.00722500	-3.13490100	-0.88838400
		H	3.35799000	-0.00525600	-1.95994800
		H	-3.35799000	0.00525600	-1.95994800
		H	2.53207300	-0.00297200	0.50591100
2-T_1	E = -392.818756577 a.u.	Os	0.00000000	0.00000000	0.30281500
		P	-2.41767400	0.00000000	0.04282200
		C	0.00000000	2.00607800	-0.16584700
		C	0.00000000	2.35287600	-1.51133600
		C	0.00000000	1.25889600	-2.39215900
		H	0.00000000	1.37434300	-3.47347800
		C	0.00000000	0.00000000	-1.76907100
		C	0.00000000	-1.25889600	-2.39215900
		H	0.00000000	-1.37434300	-3.47347800
		Cl	0.00000000	0.00000000	2.63575100
		H	-3.17843900	0.00000000	1.24083400
		H	-2.96686100	1.10407200	-0.65836300
		H	-2.96686100	-1.10407200	-0.65836300
		C	0.00000000	-2.00607800	-0.16584700
		C	0.00000000	-2.35287600	-1.51133600
		H	0.00000000	-2.79860300	0.58526500
		P	2.41767400	0.00000000	0.04282200
		H	2.96686100	1.10407200	-0.65836300
		H	3.17843900	0.00000000	1.24083400
		H	2.96686100	-1.10407200	-0.65836300
		H	0.00000000	3.37847900	-1.86582200
		H	0.00000000	-3.37847900	-1.86582200
		H	0.00000000	2.79860300	0.58526500

3-S_0	E = -491.485842262 a.u.	Os	0.31596800	0.00022000	-0.00004800
		P	0.14099800	2.36406100	-0.00023400
		C	-0.14056300	-0.00045900	-2.01952300
		C	-1.50398300	-0.00124900	-2.34029600
		C	-2.41326800	-0.00158400	-1.26179200
		H	-3.49215500	-0.00225200	-1.41921200
		C	-1.81578500	-0.00106000	0.00010900
		C	-2.41303500	-0.00126200	1.26214800
		H	-3.49189800	-0.00185400	1.41972800
		H	1.30757700	3.18354600	-0.00010500
		H	-0.55652800	2.97052400	-1.08519700
		H	-0.55685200	2.97052200	1.08451300
		C	-0.14019400	0.00010200	2.01942100
		C	-1.50357000	-0.00060400	2.34048000
		H	0.53772400	0.00053400	2.87635300
		P	0.14448400	-2.36384200	0.00029600
		H	-0.55249900	-2.97168500	-1.08424600
		H	1.31251600	-3.18124400	0.00002600
		H	-0.55180300	-2.97142500	1.08543100
		H	-1.86298700	-0.00164800	-3.36827300
		H	-1.86233500	-0.00067800	3.36853600
		H	0.53720600	-0.00029700	-2.87657400
		C	2.23123100	0.00129400	-0.00006500
		O	3.39140700	0.00186100	-0.00014400
3-T_1	E = -491.427992484 a.u.	Os	0.00000000	0.00000000	0.37293800
		P	0.00000000	2.34102400	0.02930700
		C	2.02339700	0.00000200	-0.20354600
		C	2.35282600	-0.00002600	-1.52427600
		C	1.25403400	-0.00001800	-2.43471800
		H	1.38802300	0.00000500	-3.51403900
		C	0.00000000	0.00000000	-1.82061100
		C	-1.25403400	0.00001800	-2.43471800
		H	-1.38802300	-0.00000500	-3.51403900
		H	-0.00014100	3.26738400	1.11532400
		H	1.09460900	2.86397600	-0.71404700
		H	-1.09448300	2.86393700	-0.71425700
		C	-2.02339700	-0.00000200	-0.20354600
		C	-2.35282600	0.00002600	-1.52427600
		H	-2.77800400	-0.00005600	0.58549300
		P	0.00000000	-2.34102400	0.02930700
		H	1.09448300	-2.86393700	-0.71425700
		H	0.00014100	-3.26738400	1.11532400
		H	-1.09460900	-2.86397600	-0.71404700
		H	3.38430200	-0.00006000	-1.87106600
		H	-3.38430200	0.00006000	-1.87106600
		H	2.77800400	0.00005600	0.58549300

		C	0.00000000	0.00000000	2.33241900
		O	0.00000000	0.00000000	3.48528700
2-PMe$_3$-S$_0$	E = -628.732039159 a.u.	Os	-0.00006000	-0.13635900	-0.00019100
		P	-2.45607500	-0.28717500	-0.00107300
		C	-0.00235800	0.48848100	1.85666800
		C	-0.00311200	1.78778000	2.31562400
		C	-0.00144500	2.67775300	1.23179800
		H	-0.00164000	3.76169100	1.33203100
		C	0.00028700	2.01674000	0.00072800
		C	0.00213500	2.67887800	-1.22977500
		H	0.00271200	3.76290500	-1.32904500
		Cl	-0.00021500	-2.53894400	0.00020000
		C	0.00227900	0.49020800	-1.85634500
		C	0.00339900	1.78985000	-2.31436100
		H	0.00208400	-0.39592500	-2.50570700
		P	2.45602000	-0.28738800	0.00068300
		H	-0.00451200	2.07136700	3.36381800
		H	0.00481600	2.07427200	-3.36232600
		H	-0.00231300	-0.39800200	2.50548300
		C	3.12059200	-1.18702100	1.47467400
		H	2.88686100	-0.62866100	2.38557600
		H	2.65673900	-2.17517700	1.53062400
		H	4.20622200	-1.29806100	1.39235100
		C	3.11895000	-1.23629800	-1.44281100
		H	2.65532800	-2.22584200	-1.46444600
		H	2.88392700	-0.70982800	-2.37221900
		H	4.20479500	-1.34344800	-1.35795900
		C	3.38652300	1.31018900	-0.02804900
		H	3.11686900	1.87475800	-0.92426100
		H	3.12636200	1.90320300	0.85244100
		H	4.46441300	1.12070500	-0.03073800
		C	-3.11922900	-1.23804800	1.44100300
		H	-2.65540200	-2.22752000	1.46147300
		H	-2.88455900	-0.71276200	2.37117100
		H	-4.20502900	-1.34526500	1.35569300
		C	-3.12081500	-1.18450800	-1.47641200
		H	-2.88653600	-0.62506400	-2.38650300
		H	-2.65749200	-2.17285100	-1.53354600
		H	-4.20652100	-1.29512400	-1.39453900
		C	-3.38597000	1.31067500	0.03011900
		H	-3.11558300	1.87383300	0.92699000
		H	-3.12597200	1.90478300	-0.84967300
		H	-4.46394500	1.12168900	0.03308000
2-PMe$_3$-T$_1$	E = -628.694909820 a.u.	Os	0.00002600	-0.19621500	-0.00008600
		P	-2.48302200	-0.16319000	-0.00023200
		C	-0.00020200	0.26944400	1.99684700

		C	-0.00025600	1.61564200	2.35620200
		C	-0.00023600	2.49465100	1.25787600
		H	-0.00029300	3.57906200	1.37125400
		C	-0.00017300	1.86253800	0.00072300
		C	-0.00017400	2.49563200	-1.25592100
		H	-0.00028900	3.58013100	-1.36845600
		Cl	0.00019200	-2.56397100	-0.00010400
		C	0.00018400	0.27101500	-1.99671700
		C	0.00002900	1.61749300	-2.35496700
		H	0.00035700	-0.48910600	-2.78455300
		P	2.48304700	-0.16304700	-0.00014800
		H	-0.00031500	1.97073800	3.38454900
		H	0.00006500	1.97343500	-3.38302300
		H	-0.00021600	-0.49133500	2.78404200
		C	3.28848200	1.49917700	-0.00401500
		H	2.98190700	2.05331400	-0.89455700
		H	2.98269900	2.05721800	0.88435600
		H	4.37675800	1.38449200	-0.00423300
		C	3.20563900	-1.03435400	1.46247000
		H	2.93499700	-0.50149000	2.37767300
		H	2.80405400	-2.04987500	1.51077500
		H	4.29605500	-1.07666200	1.37629700
		C	3.20527600	-1.04084800	-1.45905600
		H	2.80381900	-2.05662300	-1.50264900
		H	2.93434400	-0.51223000	-2.37663600
		H	4.29571700	-1.08259800	-1.37294600
		C	-3.20521400	-1.03816200	-1.46086300
		H	-2.93393200	-0.50794900	-2.37741600
		H	-2.80402400	-2.05397300	-1.50621300
		H	-4.29568800	-1.07978700	-1.37508200
		C	-3.20546100	-1.03750600	1.46066900
		H	-2.80386600	-2.05311900	1.50682900
		H	-2.93472200	-0.50655500	2.37695700
		H	-4.29588500	-1.07962700	1.37452700
		C	-3.28870800	1.49891500	-0.00074000
		H	-2.98270200	2.05534200	0.88856500
		H	-2.98258000	2.05478200	-0.89035700
		H	-4.37696000	1.38399900	-0.00077900
2-PPh$_3$-S$_0$	E = -1779.08698654 a.u.	Os	-0.00002100	0.08355400	0.00064300
		P	-2.51060900	-0.08198900	0.02549500
		C	-0.00873400	0.71367600	1.85403200
		C	-0.07644900	2.01144900	2.31109000
		C	-0.06627500	2.89871400	1.22679200
		H	-0.09928100	3.98226100	1.32229000
		C	0.00009000	2.23552500	0.00032900
		C	0.06656300	2.89838400	-1.22629000

		H	0.09970900	3.98190000	-1.32207900
		Cl	-0.00007000	-2.31843600	0.00167000
		C	0.00875300	0.71320000	-1.85294100
		C	0.07665300	2.01081300	-2.31036600
		H	-0.07202500	-0.16676500	-2.50256100
		P	2.51054800	-0.08205200	-0.02557400
		H	-0.11176500	2.29487700	3.35867900
		H	0.11203200	2.29395500	-3.35803000
		C	3.09940300	-1.33715300	-1.25710300
		C	3.83745500	-2.45690600	-0.85018800
		C	2.79115300	-1.18176700	-2.61955800
		C	4.25567200	-3.40306300	-1.78859300
		H	4.08689300	-2.59972300	0.19486600
		C	3.21768400	-2.12488300	-3.55367000
		H	2.23283300	-0.31537400	-2.96027400
		C	3.94802700	-3.24138300	-3.13906500
		H	4.82437000	-4.26717600	-1.45774100
		H	2.97940100	-1.98692500	-4.60455800
		H	4.27583800	-3.97859600	-3.86612500
		C	3.43220100	1.47879300	-0.42177800
		C	3.30280200	2.57185700	0.45256000
		C	4.27398300	1.59346100	-1.53652200
		C	3.99543500	3.75630300	0.20953400
		H	2.67464800	2.48996500	1.33312000
		C	4.95849600	2.78735400	-1.78163000
		H	4.41137300	0.75647900	-2.21105100
		C	4.82056100	3.86938900	-0.91320800
		H	3.89504500	4.58980400	0.89911300
		H	5.60758800	2.86217100	-2.64930300
		H	5.35922400	4.79340200	-1.10260700
		C	3.23047800	-0.62785700	1.59825500
		C	4.46347700	-0.12061600	2.03854200
		C	2.57434200	-1.59874300	2.37442900
		C	5.01762600	-0.56206600	3.24149900
		H	4.99554600	0.61659500	1.44829500
		C	3.13623500	-2.03818200	3.57383200
		H	1.64276000	-2.03685300	2.03176100
		C	4.35495500	-1.51701700	4.01360500
		H	5.97095200	-0.15859400	3.57038200
		H	2.62094900	-2.79305400	4.16093800
		H	4.78861800	-1.85835600	4.94913000
		C	-3.22933900	-0.62889900	-1.59849000
		C	-4.46184500	-0.12168600	-2.04016500
		C	-2.57282100	-1.60057700	-2.37335000
		C	-5.01514900	-0.56396700	-3.24320800
		H	-4.99418100	0.61612200	-1.45091400

		C	-3.13386200	-2.04083100	-3.57285100
		H	-1.64162300	-2.03864800	-2.02957400
		C	-4.35210200	-1.51970700	-4.01401300
		H	-5.96811200	-0.16052500	-3.57317800
		H	-2.61830200	-2.79630500	-4.15894100
		H	-4.78510400	-1.86168400	-4.94961200
		C	-3.43248100	1.47912800	0.42010600
		C	-4.27485800	1.59451300	1.53432800
		C	-3.30251900	2.57166800	-0.45479800
		C	-4.95943300	2.78859200	1.77834500
		H	-4.41262800	0.75795700	2.20930900
		C	-3.99521100	3.75630200	-0.21285900
		H	-2.67386100	2.48922800	-1.33494200
		C	-4.82096100	3.87010100	0.90934900
		H	-5.60898900	2.86396700	2.64562200
		H	-3.89437400	4.58939100	-0.90287100
		H	-5.35967300	4.79425900	1.09790600
		C	-3.10036300	-1.33620600	1.25748500
		C	-3.83886100	-2.45582300	0.85101300
		C	-2.79241100	-1.18017300	2.61992900
		C	-4.25778700	-3.40123800	1.78985100
		H	-4.08809700	-2.59911300	-0.19402400
		C	-3.21965900	-2.12253400	3.55447200
		H	-2.23372900	-0.31386400	2.96027000
		C	-3.95042800	-3.23892300	3.14031200
		H	-4.82681000	-4.26527100	1.45934900
		H	-2.98158800	-1.98409200	4.60534500
		H	-4.27879000	-3.97555900	3.86770900
		H	0.07192200	-0.16607200	2.50394900
2-PPh$_3$-T$_1$	E = -1779.05200444 a.u.	Os	0.01020800	-0.10008300	0.00544300
		P	-2.52584500	-0.05899500	-0.00060100
		C	-0.02114700	0.59857100	1.94119500
		C	-0.07763400	1.97606200	2.13670000
		C	-0.06880500	2.71231200	0.94242800
		H	-0.10851500	3.80101300	0.91877100
		C	-0.00796800	1.93157000	-0.22801800
		C	0.02942300	2.42550400	-1.54686900
		H	0.04571200	3.49135900	-1.77319200
		Cl	0.04188900	-2.46913600	0.19233000
		C	0.00657300	0.13905100	-2.02802300
		C	0.03454200	1.43540800	-2.53959800
		H	-0.04584100	-0.70323300	-2.72199100
		P	2.54434400	-0.05753700	0.00737900
		H	-0.11415700	2.45066400	3.11463800
		H	0.04203100	1.67185200	-3.60122100
		C	3.28264800	-1.43064000	-1.00042400

		C	4.27836800	-2.25581000	-0.45659500
		C	2.86966500	-1.64245500	-2.32520700
		C	4.84519800	-3.27359900	-1.22641700
		H	4.61576600	-2.11364600	0.56325800
		C	3.44573600	-2.65396800	-3.09274600
		H	2.10237500	-1.01561600	-2.76507400
		C	4.43224100	-3.47577600	-2.54309700
		H	5.61214100	-3.90722800	-0.79068100
		H	3.11860800	-2.80290300	-4.11777600
		H	4.87434500	-4.26932100	-3.13839500
		C	3.32014500	1.50000400	-0.62695200
		C	3.22605400	2.67542200	0.13965600
		C	3.99088500	1.53984900	-1.85686500
		C	3.79120100	3.86411200	-0.31953200
		H	2.72990200	2.65795700	1.10442000
		C	4.54710400	2.73641200	-2.31657500
		H	4.09559000	0.64239700	-2.45571400
		C	4.44869000	3.89892200	-1.55236700
		H	3.72344500	4.76156000	0.28895600
		H	5.06662200	2.75175000	-3.27027300
		H	4.88930300	4.82547200	-1.90885100
		C	3.25873900	-0.30640200	1.70029900
		C	4.42384300	0.35943000	2.11102200
		C	2.66849400	-1.25019300	2.55787900
		C	4.97278300	0.10245000	3.36946900
		H	4.90822200	1.07526600	1.45650900
		C	3.22718000	-1.50796200	3.80966400
		H	1.78718400	-1.79880800	2.24157700
		C	4.37564300	-0.82733000	4.22138100
		H	5.87179000	0.62817800	3.67790400
		H	2.76543300	-2.24393100	4.46166700
		H	4.80618900	-1.02634500	5.19849100
		C	-3.24718400	-0.56471800	-1.63356300
		C	-4.32498900	0.10415500	-2.23176900
		C	-2.73662500	-1.72574400	-2.23935300
		C	-4.86765100	-0.37061100	-3.42929900
		H	-4.75155000	0.98786500	-1.77087700
		C	-3.28772700	-2.19724100	-3.42977000
		H	-1.92048600	-2.27006100	-1.77394500
		C	-4.35025600	-1.51735500	-4.03124400
		H	-5.70026300	0.15786000	-3.88466200
		H	-2.88877600	-3.09922000	-3.88498200
		H	-4.77629900	-1.88465800	-4.96037700
		C	-3.27788700	1.57645000	0.42798600
		C	-3.85013100	1.78136400	1.69178200
		C	-3.24502300	2.64113500	-0.48950200

		C	-4.38250900	3.02764700	2.02977200
		H	-3.89025200	0.97147400	2.41192900
		C	-3.78794100	3.88045800	-0.15060300
		H	-2.80846600	2.50101100	-1.47293100
		C	-4.35599300	4.07707100	1.11082000
		H	-4.82529800	3.17175900	3.01103000
		H	-3.77143000	4.69047700	-0.87436900
		H	-4.77981400	5.04226700	1.37259000
		C	-3.31873600	-1.24518100	1.18745700
		C	-4.65382800	-1.62628200	0.96958700
		C	-2.64386300	-1.72937400	2.31494600
		C	-5.29754900	-2.47319200	1.87067100
		H	-5.19199900	-1.26908300	0.09777200
		C	-3.29332400	-2.57723000	3.21460100
		H	-1.60975900	-1.46286900	2.48760700
		C	-4.61935400	-2.95035000	2.99477600
		H	-6.32869500	-2.76272400	1.69031700
		H	-2.75684200	-2.95095900	4.08189500
		H	-5.12163200	-3.61418800	3.69243300
		H	0.01583300	-0.04746500	2.82239800
2'	E = -392.424142276 a.u.	Os	0.00000000	0.00000000	0.34983900
		P	0.00000000	2.46386500	0.03119600
		C	1.90012200	-0.00004200	-0.26565800
		C	2.32763000	-0.00003300	-1.56142500
		C	1.23928500	-0.00003800	-2.45312300
		H	1.34691200	-0.00006200	-3.53453000
		C	0.00000000	0.00000000	-1.81038500
		C	-1.23928500	0.00003800	-2.45312300
		H	-1.34691200	0.00006200	-3.53453000
		Cl	0.00000000	0.00000000	2.61791800
		H	-0.00027200	3.21721100	1.23369900
		H	1.12013300	2.95625700	-0.68279400
		H	-1.12016200	2.95593200	-0.68300000
		C	-1.90012200	0.00004200	-0.26565800
		C	-2.32763000	0.00003300	-1.56142500
		H	-2.54504200	0.00024700	0.62160900
		P	0.00000000	-2.46386500	0.03119600
		H	1.12016200	-2.95593200	-0.68300000
		H	0.00027200	-3.21721100	1.23369900
		H	-1.12013300	-2.95625700	-0.68279400
		H	3.37124000	0.00003200	-1.85673200
		H	-3.37124000	-0.00003200	-1.85673200
		H	2.54504200	-0.00024700	0.62160900

Supplementary Table 2. Cartesian coordinates together with electronic energies for all the complexes calculated in this study. The electronic energies are given by atomic unit (a.u.).

Supplementary Reference

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