Supplementary information

Property	charge-compensated COT^{2-}	uncompensated COT^{2-}
$\rho_{C\text{-}C}$	0.301	0.301
$\nabla^2\rho_{C\text{-}C}$	-0.832	-0.839
$\rho_{C\text{-}H}$	0.282	0.272
$\nabla^2\rho_{C\text{-}H}$	-0.964	-1.01
N_C	6.191	6.049
q_C	-0.191	-0.049
λ_C	4.150	4.086
δ_{CC}	1.384	1.286
N_H	1.059	1.201
q_H	-0.059	-0.201
λ_H	0.0485	0.598
δ_{CH}	0.986	1.014

Table S1. Comparison of QTAIM date for the COT dianion in the presence and absence of a compensating dicationic point charge. All values are given in a.u.

Property	C_6H_6	COT^{2-}	ThCOT2: $1\,^1A_g$	PuCOT2: $1\,^5A_g$	CeCOT2: $1\,^1A_g$	CeCOT2: $2\,^1A_g$
$\rho_{M\text{-}C}$	-	-	0.0401	0.0415	0.0374	0.0362
$\nabla^2\rho_{M\text{-}C}$	-	-	0.107	0.124	0.110	0.114
$\rho_{C\text{-}C}$	0.323	0.301	0.303	0.304	0.305	0.305
$\nabla^2\rho_{C\text{-}C}$	-0.969	-0.832	-0.840	-0.846	-0.854	-0.853
$\rho_{C\text{-}H}$	0.292	0.272	0.293	0.293	0.293	0.292
$\nabla^2\rho_{C\text{-}H}$	-1.13	-0.964	-1.12	-1.13	-1.12	-1.12

Table S2. BCP properties of the M-C, C-C and C-H bonds in benzene, the COT dianion, and the metallocenes considered in this study. Metallocene properties obtained at B3LYP-optimised geometries. All values are given in a.u.

Property	ThCOT2: $1\,^1A_g$	PuCOT2: $1\,^5A_g$	CeCOT2: $1\,^1A_g$	CeCOT2: $2\,^1A_g$
N_M	87.459	91.992	56.029	56.049
q_M	+2.541	+2.008	+1.971	+1.951
λ_M	85.844	90.356	54.546	54.777
$Z_M - \lambda_M$	4.156	3.644	3.454	3.223
$N_M - \lambda_M$	1.615	1.636	1.483	1.272
N_C	6.131	6.099	6.095	6.094
q_C	-0.131	-0.099	-0.095	-0.094
λ_C	0.408	4.027	4.065	4.072
δ_{MC}	0.194	0.197	0.178	0.153

Table S3. Integrated QTAIM properties associated with the metallocene M-C bond, obtained at the B3LYP geometry. All values are given in a.u.

Property	C_6H_6	COT^{2-}	ThCOT2: 1 ^1A_g	PuCOT2: 1 ^5A_g	CeCOT2: 1 ^1A_g	CeCOT2: 2 ^1A_g
N_C	5.968	6.049	6.131	6.099	6.095	6.094
q_C	+0.032	-0.049	-0.131	-0.099	-0.095	-0.094
λ_C	4.050	4.086	0.408	4.027	4.065	4.072
δ_{CC}	1.287	1.286	1.309	1.318	1.303	1.304
N_H	1.032	1.201	1.027	1.026	1.028	1.028
q_H	-0.032	-0.201	-0.027	-0.026	-0.028	-0.028
λ_H	0.476	0.598	0.462	0.461	0.462	0.462
δ_{CH}	0.987	1.014	0.975	0.972	0.976	0.977

Table S4. Integrated QTAIM properties associated with the C-C and C-H bond in benzene, the COT dianion and metallocenes. Metallocene properties are calculated at the B3LYP geometry. All values are given in a.u.