

Should environmental effects be included when performing QTAIM calculations on actinide systems? A comparison of QTAIM metrics for $\text{Cs}_2\text{UO}_2\text{Cl}_4$, $\text{U}(\text{Se}_2\text{PPh}_2)_4$ and $\text{Np}(\text{Se}_2\text{PPh}_2)_4$ in gas phase, COSMO and PEECM

Joseph P. W. Wellington,¹ Andrew Kerridge² and Nikolas Kaltsoyannis*³

¹Department of Chemistry, University College London,
20 Gordon Street, London WC1H 0AJ, UK

²Department of Chemistry, Lancaster University, Bailrigg, Lancaster LA1 4YP, UK

³School of Chemistry, The University of Manchester,
Oxford Road, Manchester, M13 9PL, UK

* nikolas.kaltsoyannis@manchester.ac.uk

Supplementary Information

The total SCF energies for the each of the five systems are given in the four different environments followed by the atomic coordinates of each system.

1) $[\text{UO}_2\text{Cl}_4]^{2-}$

Environment	SCF Energy/ Hartree
Gas Phase	-29953.29633965
COSMO	-29953.57111197
Embedded Formal	-29955.46224841
Embedded Natural	-29954.29182637

Atomic coordinates/ Å:

Atom	x	y	z
U	27.3796363	15.2819957	11.3470608
O	28.5344670	15.2819957	12.6967934
O	26.2248046	15.2819957	9.9973282
Cl	28.7973459	17.2155502	10.1724131
Cl	25.9619257	17.2155502	12.5217085
Cl	28.7973459	13.3484403	10.1724131
Cl	25.9619257	13.3484403	12.5217085

2) $\text{Cs}_2\text{UO}_2\text{Cl}_4$

Environment	SCF Energy/ Hartree
Gas Phase	-45520.13915495
COSMO	-45520.24573394
Embedded Formal	-45520.40975755
Embedded Natural	-45520.39255059

Atomic coordinates/ Å:

Atom	x	y	z
U	27.3796363	15.2819957	11.3470608
O	28.5344670	15.2819957	12.6967934
O	26.2248046	15.2819957	9.9973282
Cl	28.7973459	17.2155502	10.1724131
Cl	25.9619257	17.2155502	12.5217085
Cl	28.7973459	13.3484403	10.1724131
Cl	25.9619257	13.3484403	12.5217085
Cs	25.7713058	11.4614958	9.5277293
Cs	28.9879659	19.1024936	13.1663923

3) (Cs₂UO₂Cl₄)₇

Environment	SCF Energy/ Hartree
Gas Phase	-318641.2386523
COSMO	-318641.6109086
Embedded Formal	
Embedded Natural	-318642.2956328

Atomic coordinates/ Å:

Atom	x	y	z
U	21.4856370	11.4614958	11.3470608
U	21.4856370	19.1024936	11.3470608
U	26.3344576	15.2819957	17.0205912
U	27.3796363	15.2819957	11.3470608
U	28.4248140	15.2819957	5.6735304
U	33.2736337	11.4614958	11.3470608
U	33.2736337	19.1024936	11.3470608
O	20.3308063	11.4614958	9.9973282
O	20.3308063	19.1024936	9.9973282
O	22.6404696	11.4614968	12.6967934
O	22.6404696	19.1024936	12.6967934
O	25.1796269	15.2819957	15.6708576
O	26.2248046	15.2819957	9.9973282
O	27.2699833	15.2819957	4.3237978
O	27.4892903	15.2819957	18.3703238
O	28.5344670	15.2819957	12.6967934
O	29.5796447	15.2819957	7.0232630
O	32.1188040	11.4614958	9.9973282
O	32.1188040	19.1024936	9.9973282
O	34.4284643	11.4614968	12.6967934
O	34.4284643	19.1024936	12.6967934
Cl	20.0679284	9.5279413	12.5217085
Cl	20.0679284	13.3950522	12.5217085
Cl	20.0679284	17.1689382	12.5217085
Cl	20.0679284	21.0360491	12.5217085
Cl	22.9033486	9.5279413	10.1724131
Cl	22.9033486	13.3950522	10.1724131
Cl	22.9033486	17.1689382	10.1724131
Cl	22.9033486	21.0360491	10.1724131
Cl	24.9167490	13.3484403	18.1952389
Cl	24.9167490	17.2155502	18.1952389
Cl	25.9619257	13.3484403	12.5217085
Cl	25.9619257	17.2155502	12.5217085
Cl	27.0071054	13.3484403	6.8481781
Cl	27.0071054	17.2155502	6.8481781
Cl	27.7521682	13.3484403	15.8459426
Cl	27.7521682	17.2155502	15.8459426
Cl	28.7973459	13.3484403	10.1724131
Cl	28.7973459	17.2155502	10.1724131
Cl	29.8425226	13.3484403	4.4988827
Cl	29.8425226	17.2155502	4.4988827
Cl	31.8559251	9.5279413	12.5217085

Cl	31.8559251	13.3950522	12.5217085
Cl	31.8559251	17.1689382	12.5217085
Cl	31.8559251	21.0360491	12.5217085
Cl	34.6913433	9.5279413	10.1724131
Cl	34.6913433	13.3950522	10.1724131
Cl	34.6913433	17.1689382	10.1724131
Cl	34.6913433	21.0360491	10.1724131
Cs	19.8773084	15.2819957	9.5277293
Cs	23.0939665	15.2819957	13.1663923
Cs	24.1391462	15.2819957	7.4928609
Cs	24.7261281	11.4614968	15.2012587
Cs	24.7261281	19.1024936	15.2012587
Cs	25.7713058	11.4614958	9.5277293
Cs	25.7713058	19.1024936	9.5277293
Cs	28.9879659	11.4614968	13.1663923
Cs	28.9879659	19.1024936	13.1663923
Cs	30.0331436	11.4614958	7.4928609
Cs	30.0331436	19.1024936	7.4928609
Cs	30.6201254	15.2819957	15.2012587
Cs	31.6653051	15.2819957	9.5277293
Cs	34.8819632	15.2819957	13.1663923

4) U(Se₂PPh₂)₄

Environment	SCF Energy/ Hartree
Gas Phase	-50556.01928956
COSMO	-50556.05305963
Embedded Formal	-50556.04612146
Embedded Natural	-50556.07177235

Atomic coordinates/ Å:

Atom	x	y	z
U	23.9318924	20.8891317	42.5281130
Se	24.9093165	21.2940551	45.2086235
Se	26.0332209	22.5377061	41.4010779
Se	21.1992737	20.8799946	43.5185798
Se	23.0697040	18.8575808	40.5855522
Se	26.3741558	19.1350252	42.1828669
Se	22.3662737	22.3158879	40.3650379
Se	23.4997467	23.8149479	43.1851703
Se	23.6367224	18.3183616	44.1431817
P	21.5710619	18.9092896	44.3580420
P	24.6886763	23.4459365	44.9575766
P	27.3023101	20.7975350	41.1603970
P	22.0825968	20.3904276	39.4221040
C	26.3030312	24.2775831	44.9321329
C	20.4450005	17.7356606	43.5411970
C	27.5678812	20.4562386	39.3985448
C	22.7725272	20.3935615	37.7437455
C	21.0451156	18.9196706	46.0950155
C	20.3268234	19.9744084	39.1591830
C	23.8366382	24.0415646	46.4455769
C	26.9313742	19.4093349	38.7445410
C	28.9483546	21.2142398	41.8066734
C	23.7978793	21.2475370	37.3818758
C	22.4667920	23.7840011	46.5793930
C	20.9078207	16.5810308	42.9437351
C	20.8739923	20.1085766	46.7829447
C	29.1173412	22.2817093	42.6817569
C	27.3440821	23.7301377	45.6916817
C	19.7707800	18.7933371	39.6322520
C	26.4984926	25.4547375	44.2630514
C	19.0924203	18.0294598	43.5016171
C	28.3520809	21.3454700	38.6691515
C	19.5653753	20.8411151	38.3789020
C	22.3160929	19.4328392	36.8239942
C	20.7976902	17.7190122	46.7452498
C	24.5368063	24.6213279	47.4916060
C	30.0651575	20.4826805	41.3850577
C	27.1019193	19.2506841	37.3837608
C	24.3502201	21.1701699	36.1002552
C	20.0233763	15.7270555	42.3142331
C	21.8258066	24.0729031	47.7705473
C	20.3778643	20.0958453	48.0796438
C	28.5600848	24.4019579	45.7746104

C	27.7058925	26.1148055	44.3572882
C	30.3848209	22.6381852	43.0982838
C	18.4670331	18.4750547	39.2779215
C	18.2148824	17.1705876	42.8721150
C	28.4995110	21.1789839	37.2951781
C	27.8954203	20.1252252	36.6675610
C	18.6902395	16.0169373	42.2708841
C	22.9023367	19.3603685	35.5649899
C	28.7262760	25.6026163	45.1149525
C	18.2723638	20.4993291	38.0358797
C	20.3110729	17.7111774	48.0476036
C	23.9109551	20.2221789	35.2087739
C	23.8889666	24.9229614	48.6808748
C	31.3309509	20.8587431	41.8125161
C	22.5411691	24.6330795	48.8165759
C	20.0844191	18.9059600	48.6959530
C	17.7215672	19.3192369	38.4693693
C	31.4884565	21.9448197	42.6534857
H	26.3918009	18.8197787	39.2213794
H	21.9892778	23.4196904	45.8707321
H	24.1206559	21.8713700	37.9925308
H	21.0911054	20.9165236	46.3777263
H	28.3767875	22.7557047	42.9851994
H	21.8147105	16.3783098	42.9663523
H	20.2652970	18.2243461	40.1769410
H	27.2225576	22.9231701	46.1364799
H	25.8140005	25.8112131	43.7447486
H	18.7746298	18.8070477	43.9011814
H	19.9270829	21.6471036	38.0886524
H	28.7779231	22.0515669	39.1007564
H	20.9591151	16.9149827	46.3061062
H	21.6285921	18.8511172	37.0577011
H	29.9592608	19.7491626	40.8234059
H	25.4429153	24.8093589	47.3935991
H	26.6776688	18.5475249	36.9446172
H	25.0202380	21.7656023	35.8495841
H	20.9197502	23.8927065	47.8666680
H	20.3396802	14.9475092	41.9184381
H	20.2422633	20.8969370	48.5338656
H	30.4927434	23.3511373	43.6863217
H	29.2589154	24.0435233	46.2740654
H	18.0881718	17.6847358	39.5889032
H	27.8291663	26.9149181	43.8992969
H	17.3051249	17.3654745	42.8532678
H	29.0090633	21.7822509	36.8051459
H	17.7691385	21.0741956	37.5062688
H	28.0189257	19.9969330	35.7553473
H	22.6071334	18.7208663	34.9562187
H	18.1014554	15.4410910	41.8373945
H	29.5296881	26.0638806	45.1828030
H	20.1388609	16.9051890	48.4773231
H	24.3570209	25.3156723	49.3819976
H	24.2963991	20.1634190	34.3644119
H	32.0763897	20.3778922	41.5320672
H	22.1112452	24.8162142	49.6213585

H	19.7317067	18.9049806	49.5553930
H	16.8554107	19.0871357	38.2243535
H	32.3402213	22.2072801	42.9192339

5) Np(Se₂PPh₂)₄

Environment	SCF Energy/ Hartree
Gas Phase	-51343.24289143
COSMO	-51343.27587509
Embedded Formal	-51343.26907218
Embedded Natural	-51343.24289143

Atomic coordinates/ Å:

Atom	x	y	z
Np	29.3676680	19.3716647	32.8921332
Se	28.3911298	18.9711188	30.2163130
Se	27.2777022	17.7269135	34.0094736
Se	32.1150249	19.4697578	31.9564009
Se	30.1666038	21.3495734	34.8867763
Se	29.8367421	16.4780224	32.2058461
Se	26.9259267	21.1144098	33.2311569
Se	30.9305667	17.9193111	35.0295047
Se	29.6396671	21.9468046	31.3149688
P	31.7237351	21.4229448	31.1013465
P	28.6372373	16.8265312	30.4346497
P	25.9948745	19.4541069	34.2502463
P	31.1893420	19.8187658	36.0182375
C	27.0474941	15.9650278	30.4708502
C	25.7283417	19.7691209	35.9952351
C	32.8056922	22.6411307	31.9245362
C	30.5087515	19.7830771	37.6940306
C	32.2578072	21.4418851	29.3754007
C	32.9415960	20.2496164	36.2837094
C	26.3596405	20.7929169	36.6626871
C	29.4919562	16.2531263	28.9436305
C	24.3432506	19.0433927	33.5818514
C	29.5276790	18.8948573	38.0560375
C	30.8320835	16.5103212	28.8041072
C	32.4327054	20.2675608	28.6872087
C	32.3305897	23.7516547	32.5203401
C	24.1683522	17.9827126	32.7371676
C	25.9776866	16.4873933	29.7392934
C	33.4879965	21.4269318	35.8198879
C	26.8683116	14.7976820	31.1137905
C	34.1510933	22.4058673	31.9433905
C	24.9544314	18.9217734	36.7456463
C	33.7051376	19.4122382	37.0397779
C	30.9049997	20.7520440	38.6197901
C	32.5037147	22.6411307	28.7305746
C	26.1991183	20.9574020	38.0258719
C	23.2407120	19.7890582	34.0155063
C	28.8194338	15.6430357	27.9141722
C	28.9638487	18.9706203	39.3381488
C	31.4888411	16.2152451	27.6256974
C	33.1714015	24.6418687	33.1463119
C	32.9248376	20.2496164	27.3862441
C	24.7881581	15.7845929	29.6601034

C	25.6757928	14.1028568	31.0421426
C	22.9013795	17.6726832	32.2997423
C	34.7885459	21.7489239	36.1498423
C	35.0096973	23.2761438	32.5787900
C	24.7871220	19.0752928	38.1031749
C	25.4304862	20.1230131	38.7366875
C	34.5070802	24.3577583	33.1651662
C	30.3337889	20.8058761	39.8679621
C	24.6509619	14.6082749	30.2917314
C	35.0000241	19.7362237	37.3735039
C	32.9712158	22.6112253	27.4164119
C	29.3906960	19.9425776	40.2129994
C	29.4721303	15.3389875	26.7319914
C	21.9865585	19.4172222	33.5667686
C	30.8009336	15.6420393	26.6000095
C	33.2013676	21.4428821	26.7885549
C	35.5285922	20.9015758	36.9511614
C	21.8000183	18.3575391	32.7409391
H	26.9034753	21.3820719	36.1913225
H	31.3039252	16.8871418	29.5111524
H	29.2389432	18.2468855	37.4564631
H	32.2155361	19.4640756	29.1001234
H	24.8983590	17.4762974	32.4618914
H	31.4155629	23.9201280	32.5090278
H	32.9834500	22.0001376	35.2881888
H	26.0648734	17.3058312	29.3056385
H	27.5652801	14.4487741	31.6190922
H	34.4920944	21.6502331	31.5229338
H	33.3454082	18.6077559	37.3357941
H	24.5260909	18.2159825	36.3195334
H	31.5615614	21.3681156	38.3878788
H	32.3581902	23.4515941	29.1642295
H	23.3482852	20.5147864	34.5867998
H	27.9193924	15.4316972	28.0122159
H	26.6329082	21.6582082	38.4557552
H	32.3942265	16.4066457	27.5351951
H	28.3051567	18.3665114	39.5964564
H	32.8407593	25.4144503	33.5441428
H	33.0624516	19.4461317	26.9375059
H	22.7907525	16.9808486	31.6888544
H	35.1669677	22.5374558	35.8312002
H	24.0797597	16.1295129	29.1661141
H	25.5713023	13.3003686	31.4984233
H	35.9245122	23.1096646	32.5976443
H	24.2540823	18.4851401	38.5858508
H	35.5092145	19.1530492	37.8882333
H	25.3283521	20.2476235	39.6530182
H	30.6125902	21.4528502	40.4750785
H	35.0837247	24.9409323	33.6025915
H	23.8543592	14.1337602	30.2181988
H	33.1282756	23.4107224	26.9676732
H	29.0125248	14.9322608	26.0343728
H	21.2468287	19.9086840	33.8420448
H	29.0112817	19.9944156	41.0614559
H	31.2373084	15.4546257	25.8005763

H	33.5560980	21.4498603	25.9287872
H	36.3938866	21.1358433	37.2000412
H	20.9447646	18.1023384	32.4807457