Supporting information for:

SUPPORTING INFORMATION

Evaluating charge equilibration methods to generate electrostatic fields in nanoporous materials

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Contents

1 Supplementary information for ionization energies and computed radii

Table S1: Lowest energy multiplicity for the CCSD(T)/aug-cc-pvqz calculations, considering multiplicities up to 11 or 12, for atoms with an even or odd number of electrons, respectively. The multiplicity is reported for ions from -5 to +5 charge.

	-5	-4	-3	-2	-1	0	1	2	3	4	5
H	3	2	1	2	1	2	0	0	0	0	0
He	4	3	2	1	2	1	2	0	0	0	0
Li	3	6	3	2	1	2	1	2	0	0	0
Be	2	5	4	3	2	1	2	1	2	0	0
B	1	2	3	4	3	2	1	2	1	2	0
C	2	1	2	3	4	3	2	1	2	1	2
N	3	2	1	2	3	4	3	2	1	2	1
O	4	3	2	1	2	3	4	3	2	1	2
F	1	6	1	2	1	2	3	4	3	2	1
Ne	4	3	4	1	2	1	2	3	4	3	2
Na	3	2	3	2	1	2	1	2	3	4	3
Mg	2	5	4	3	2	1	2	1	2	3	4
Al	1	4	3	4	3	2	1	2	1	2	3
Si	2	1	2	3	4	3	2	1	2	1	2
P	3	2	1	2	3	4	3	2	1	2	1
S	4	3	4	1	2	3	4	3	2	1	2
Cl	5	2	1	2	1	2	3	4	3	2	1
Ar	4	5	2	1	2	1	2	3	4	3	2

	-5	-4	-3	-2	-1	0	1	2	3	4	5
Sc	5	4	5	4	3	2	3	2	1	2	3
Ti	2	1	6	1	4	3	4	1	2	1	2
V	3	4	3	2	5	4	5	4	3	2	1
Cr	8	9	4	5	6	7	6	3	4	3	2
Mn	9	2	3	4	5	6	7	6	3	4	3
Fe	4	5	8	3	6	5	6	5	6	5	4
Co	7	4	7	6	3	4	3	4	5	6	3
Ni	6	7	6	5	4	3	2	3	4	5	6
Cu	3	2	3	2	1	2	1	2	3	4	5
Zn	4	5	2	1	2	1	2	1	2	3	4
Ga	1	2	3	4	3	2	1	2	1	2	3
Ge	2	1	2	3	4	3	2	1	2	1	2
As	3	2	1	2	3	4	3	2	1	2	1
Se	4	1	4	1	2	3	4	3	2	1	2
Br	1	4	1	2	1	2	3	4	3	2	1
Kr	2	3	2	3	2	1	2	3	4	3	2

Table S2: Lowest energy multiplicity for the CCSD(T)/def2qzvpp calculations, considering multiplicities up to 11 or 12, for atoms with an even or odd number of electrons, respectively. The multiplicity is reported for ions from -5 to +5 charge.

	-5	-4	-3	-2	-1	0	1	2	3	4	5
H	3	2	1	2	1	2	0	0	0	0	0
He	4	3	2	1	2	1	2	0	0	0	0
Li	3	4	5	2	1	2	1	2	0	0	0
Be	2	3	4	3	2	1	2	1	2	0	0
B	1	2	3	4	3	2	1	2	1	2	0
C	2	1	2	3	4	3	2	1	2	1	2
N	3	2	1	2	3	4	3	2	1	2	1
O	4	3	2	1	2	3	4	3	2	1	2
F	3	4	3	2	1	2	3	4	3	2	1
Ne	2	3	4	3	2	1	2	3	4	3	2
Na	3	4	3	2	1	2	1	2	3	4	3
Mg	2	3	4	3	2	1	2	1	2	3	4
Al	1	2	3	4	3	2	1	2	1	2	3
Si	2	1	2	3	4	3	2	1	2	1	2
P	3	2	1	2	3	4	3	2	1	2	1
S	4	3	2	1	2	3	4	3	2	1	2
Cl	3	4	3	2	1	2	3	4	3	2	1
Ar	2	3	4	3	2	1	2	3	4	3	2
K	3	2	3	2	1	2	1	2	3	4	3
Ca	2	3	4	3	2	1	2	1	2	3	4
Sc	3	4	5	2	3	2	3	2	1	2	3
Ti	2	5	6	5	4	3	4	1	2	1	2
V	7	4	7	4	5	4	5	4	3	2	1
Cr	4	3	10	7	6	7	6	5	4	3	2
Mn	5	4	3	10	9	6	7	6	5	4	3
Fe	4	3	4	3	6	5	6	5	6	5	4
Co	3	2	5	6	3	4	3	4	5	6	5
Ni	4	5	6	5	4	3	2	3	4	5	6
Cu	3	2	3	2	1	2	1	2	3	4	5
Zn	2	3	4	3	2	1	2	1	2	3	4
Ga	1	2	3	4	3	2	1	2	1	4	3
Ge	2	1	2	3	4	3	2	1	2	1	2
As	3	2	1	2	3	4	3	2	1	2	1
Se	4	3	2	1	2	3	4	3	2	1	2
Br	5	4	1	2	1	2	3	4	3	2	1
Kr	4	5	4	1	2	1	2	3	4	3	2

	-5	-4	-3	-2	-1	0	1	2	3	4	5
Rb	7	4	3	2	1	2	1	2	3	4	3
Sr	4	5	4	3	2	1	2	1	2	3	4
Y	3	6	5	4	3	2	1	2	1	2	3
Zr	2	5	6	3	4	3	4	3	2	1	2
Nb	7	4	9	4	5	6	5	2	3	2	1
Mo	4	5	10	7	6	7	6	5	4	3	2
Tc	5	2	9	8	5	6	7	6	5	4	3
Ru	4	5	6	5	4	5	4	5	6	3	4
Rh	1	2	7	6	3	4	3	4	5	6	5
Pd	4	5	2	1	2	1	2	3	4	5	6
Ag	3	2	3	2	1	2	1	2	3	4	5
Cd	2	5	4	3	2	1	2	1	4	3	4
In	1	2	3	4	3	2	1	2	1	2	3
Sn	2	1	2	3	4	3	2	1	2	1	4
Sb	1	2	1	2	3	4	3	2	1	2	1
Te	2	1	2	1	2	3	4	3	2	1	2
I	5	2	1	2	1	2	3	4	3	2	1
Xe	4	5	4	1	2	1	2	3	4	3	2
Cs	1	4	3	2	1	2	1	2	3	4	3
Ba	2	3	4	3	4	1	2	1	2	3	4
La	7	6	3	2	3	4	3	2	1	2	3
Hf	8	7	6	1	4	3	4	3	2	1	2
Ta	7	8	7	4	5	4	5	2	3	2	1
W	10	3	10	7	6	7	6	5	4	3	2
Re	9	10	9	8	7	6	7	4	3	4	3
Os	4	7	8	5	4	5	6	7	6	5	4
Ir	3	6	5	4	3	4	3	4	5	6	3
Pt	4	5	4	1	2	3	2	3	4	5	6
Au	1	4	3	2	1	2	1	2	3	4	5
Hg	2	3	4	3	2	1	2	1	2	3	4
Tl	1	2	3	4	3	2	1	2	1	2	3
Pb	2	1	2	3	4	3	2	1	2	1	2
Bi	1	2	1	2	3	4	3	2	1	2	1
Po	4	1	2	1	2	3	4	3	2	1	2
At	5	4	3	2	1	2	3	4	3	2	1
Rn	4	5	4	1	2	1	2	3	4	3	2

Table S3: Ionization energies IP_n (with n ranging from -5 to +4), computed with CCSD(T)/aug-cc-pvqz. Values are expressed in eV.

atom	**-5**	**-4**	**-3**	**-2**	**-1**	**0**	**+1**	**+2**	**+3**	**+4**
H	-24.241	-31.633	2.521	-5.875	0.74	13.604	-	-	-	-
He	-35.625	-28.22	-20.746	-7.917	-2.631	24.564	54.418	-	-	-
Li	-6.944	-4.794	-3.476	-2.193	0.616	5.37	74.933	122.42	-	-
Be	-9.134	-6.552	-4.509	-2.619	-0.342	9.302	18.175	153.262	217.64	-
B	-11.643	-9.017	-6.665	-3.637	0.25	8.263	25.11	37.872	258.729	340.071
C	-16.938	-11.974	-8.891	-5.606	1.246	11.235	24.324	47.821	64.412	391.425
N	-20.767	-15.8	-10.163	-6.044	-0.227	14.525	29.563	47.362	77.373	97.772
O	-22.76	-17.924	-14.441	-6.08	1.404	13.529	35.113	54.881	77.277	113.747
F	-16.027	-23.187	-14.394	-11.236	3.386	17.362	34.842	62.714	87.101	114.047
Ne	-31.968	-20.689	-16.072	-10.615	-5.287	21.547	40.875	63.26	97.231	126.183
Na	-7.827	-2.21	-2.991	-1.868	0.547	5.016	47.613	74.748	102.995	140.046
Mg	-7.076	-5.229	-3.487	-2.01	-0.22	7.583	14.868	80.013	110.929	143.805
Al	-8.665	-7.292	-5.047	-2.678	0.427	5.951	18.687	28.185	120.037	155.68
Si	-12.753	-8.917	-6.456	-3.941	1.397	8.123	16.253	33.295	44.828	166.573
P	-15.16	-11.752	-7.291	-4.097	0.686	10.492	19.708	30.049	51.176	64.639
S	-16.101	-11.141	-11.894	-3.888	2.035	10.255	23.347	34.742	47.139	72.23
Cl	-23.672	-8.642	-8.784	-7.537	3.605	12.893	23.651	39.619	53.059	67.437
Ar	-21.206	-14.051	-10.072	-6.654	-2.758	15.738	27.521	40.496	59.163	74.577
Sc	-6.497	-5.809	-2.443	-2.617	-0.526	6.504	15.892	21.459	73.304	91.342
Ti	-11.91	-1.558	-6.418	-0.031	0.24	6.765	14.971	25.835	43.191	99.151
V	-8.304	-5.859	-3.821	-3.203	0.608	6.982	14.193	29.377	46.765	65.255
Cr	-8.467	-5.95	-4.374	-2.795	0.612	8.387	17.33	28.702	49.236	69.57
Mn	-7.738	-6.948	-4.441	-2.568	1.738	5.287	15.463	36.9	48.62	72.905
Fe	-11.15	-3.695	-5.36	-1.716	-0.382	6.879	15.992	30.625	55.139	75.681
Co	-9.959	-5.557	-4.743	-4.138	0.993	7.268	17.386	33.325	51.577	83.926
Ni	-9.598	-6.41	-4.716	-2.793	-0.4	7.411	18.295	35.614	55.034	76.196
Cu	-9.292	-7.797	-3.989	-4.554	3.008	5.631	20.511	37.069	59.246	79.112
Zn	-10.306	-6.743	-5.388	-2.48	-0.515	9.186	17.622	40.073	59.973	83.986
Ga	-8.666	-6.845	-4.913	-2.629	0.35	5.937	20.001	29.878	63.13	86.42
Ge	-12.231	-8.506	-6.15	-3.798	1.383	7.913	15.711	33.296	44.586	90.561
As	-13.93	-10.875	-6.727	-3.8	0.732	9.996	18.517	27.946	48.764	61.362
Se	-16.577	-7.737	-10.586	-3.483	2.045	9.577	21.432	31.525	42.373	66.25
Br	-14.594	-15.714	-5.012	-6.602	3.495	11.801	21.276	35.213	46.7	58.851
Kr	-22.202	-6.655	-8.134	-5.567	-2.11	14.136	24.296	35.346	51.138	63.912

Table S4: Ionization energies IP_n (with n ranging from -5 to +4), computed with CCSD(T)/def2qzvpp. Values are expressed in eV.

atom	-5	-4	-3	-2	-1	0	+1	+2	+3	+4
H	-50.081	-40.486	-67.823	31.562	0.291	13.605	-	-	-	-
He	-77.929	-64.211	-50.406	-20.552	-11.098	24.562	54.418	-	-	-
Li	-12.221	-8.802	-6.166	-3.709	0.574	5.37	75.423	122.42	-	-
Be	-17.777	-14.015	-9.103	-5.183	-0.726	9.291	18.172	153.645	217.69	-
B	-18.89	-14.925	-10.889	-5.43	0.096	8.259	25.097	37.858	258.601	340.141
C	-28.441	-18.548	-13.396	-7.987	1.093	11.227	24.322	47.8	64.396	391.26
N	-34.457	-28.776	-15.512	-8.65	-0.6	14.512	29.558	47.36	77.347	97.754
O	-37.818	-31.497	-25.149	-8.496	1.076	13.496	35.107	54.877	77.279	113.708
F	-43.978	-34.575	-27.453	-20.412	3.078	17.316	34.827	62.707	87.1	114.05
Ne	-42.864	-35.775	-25.995	-18.85	-11.68	21.486	40.849	63.233	97.206	126.159
Na	-10.369	-7.244	-5.229	-3.17	0.527	5.105	47.033	72.165	100.339	139.777
Mg	-13.84	-10.868	-6.998	-3.939	-0.516	7.61	14.983	79.942	109.781	142.428
Al	-15.522	-12.142	-8.676	-4.034	0.312	5.947	18.755	28.345	119.548	153.088
Si	-23.	-14.401	-10.106	-5.603	1.304	8.124	16.27	33.373	44.984	166.348
P	-27.41	-22.392	-11.172	-5.681	0.48	10.493	19.726	30.081	51.261	64.797
S	-28.259	-23.257	-18.327	-5.157	1.88	10.246	23.36	34.768	47.181	72.328
Cl	-31.103	-24.196	-18.912	-13.698	3.473	12.877	23.658	39.637	53.092	67.484
Ar	-32.621	-26.975	-19.453	-13.834	-8.218	15.714	27.522	40.51	59.187	74.617
K	-9.569	-7.549	-4.626	-2.905	0.448	4.299	31.406	45.548	60.82	82.178
Ca	-10.675	-8.394	-5.351	-2.931	-0.104	6.062	11.786	50.647	66.891	84.145
Sc	-11.376	-8.874	-5.613	-2.764	-0.646	6.501	15.807	21.441	73.346	91.636
Ti	-11.665	-9.208	-5.785	-3.586	0.122	6.759	14.958	25.753	43.174	99.413
V	-13.081	-9.345	-6.536	-3.961	0.483	6.968	14.187	29.293	46.736	65.477
Cr	-13.181	-9.522	-6.802	-4.129	0.498	8.319	14.852	31.098	49.201	69.732
Mn	-13.105	-10.259	-5.803	-3.243	0.264	5.194	15.456	33.799	51.577	72.988
Fe	-13.508	-10.385	-7.034	-2.829	-0.649	6.783	15.981	30.49	55.058	75.716
Co	-14.37	-10.477	-7.035	-5.693	0.93	7.252	17.273	33.203	51.422	79.969
Ni	-14.893	-12.232	-7.303	-4.228	-0.702	7.393	18.187	35.469	54.886	76.085
Cu	-13.973	-11.565	-7.809	-4.853	2.887	5.689	20.393	36.93	57.491	80.591
Zn	-15.606	-12.34	-7.997	-4.622	-0.862	9.161	17.594	39.947	59.833	83.834
Ga	-17.203	-13.468	-9.626	-4.462	0.162	5.934	20.115	30.133	90.457	59.139
Ge	-23.567	-15.271	-10.671	-5.87	1.24	7.914	15.735	33.46	44.907	90.785
As	-26.168	-21.263	-10.632	-5.371	0.514	9.998	18.541	27.997	48.987	61.759
Se	-27.863	-22.937	-18.286	-4.784	1.865	9.566	21.452	31.566	42.436	66.517
Br	-28.393	-27.521	-13.927	-14.316	3.355	11.785	21.292	35.249	46.755	58.93
Kr	-34.862	-24.077	-21.087	-10.969	-8.846	14.114	24.306	35.382	51.187	63.981
Rb	-7.486	-5.934	-4.241	-2.481	0.459	4.124	27.262	38.994	51.438	69.297
Sr	-11.427	-8.272	-5.576	-2.973	-0.122	5.653	10.965	43.079	56.289	70.148
Y	-12.159	-8.531	-6.309	-3.017	-0.438	5.985	13.092	19.532	60.891	75.361
Zr	-12.573	-9.407	-5.608	-3.873	0.375	6.589	14.978	25.298	29.58	81.045
Nb	-12.905	-9.234	-6.405	-3.872	0.776	6.982	15.268	23.525	37.375	50.172
Mo	-12.775	-9.173	-6.515	-3.954	0.62	8.573	14.548	26.991	39.957	54.092
Tc	-12.602	-9.957	-6.391	-3.908	0.214	7.036	15.186	29.71	42.528	57.196
Ru	-13.485	-9.477	-7.033	-4.247	1.495	6.702	16.432	27.917	48.574	57.794
Rh	-14.979	-10.194	-6.495	-4.238	-0.034	7.427	18.015	30.499	44.094	64.488
Pd	-15.666	-11.101	-8.78	-4.258	0.376	8.314	19.27	32.857	47.288	62.561
Ag	-16.56	-6.857	-7.543	-4.428	1.229	7.529	21.46	34.64	50.236	66.388
Cd	-16.107	-12.309	-8.045	-4.651	-0.876	8.93	16.845	50.931	39.052	69.861
In	-13.128	-10.306	-7.411	-3.503	0.351	5.559	18.757	27.889	55.717	72.334
Sn	-18.105	-11.574	-8.16	-4.544	1.37	7.297	14.203	30.372	40.62	103.421
Sb	-20.403	-16.188	-8.655	-4.299	0.756	9.099	16.534	24.665	43.547	54.789
Te	-25.726	-17.196	-12.701	-3.791	1.947	8.682	18.939	27.55	36.725	58.226
I	-23.516	-19.229	-13.502	-8.759	3.217	10.528	18.706	30.522	40.167	50.306
Xe	-25.667	-19.196	-15.222	-9.623	-4.427	12.418	21.101	30.459	43.626	54.204
Cs	-7.314	-5.586	-3.861	-2.2	0.456	3.865	23.259	33.028	43.365	57.924
Ba	-9.885	-7.747	-4.83	-4.066	1.5	5.126	9.848	36.472	47.338	58.599
La	-11.04	-7.81	-5.805	-3.467	0.812	5.323	12.364	17.334	50.708	62.43
Hf	-16.375	-12.3	-6.43	-5.5	-0.37	7.129	13.975	23.801	30.656	81.571
Ta	-16.881	-12.026	-8.274	-5.123	0.145	7.381	15.86	26.079	37.561	41.954
W	-16.669	-12.65	-9.095	-5.093	0.947	8.413	15.348	25.536	37.545	50.601
Re	-17.445	-13.298	-8.588	-4.689	-0.485	7.634	18.122	28.521	37.715	53.136
Os	-15.856	-11.954	-8.119	-5.266	0.629	8.992	16.212	26.504	42.78	55.827
Ir	-17.353	-11.999	-8.771	-5.409	1.701	9.152	16.994	28.844	40.928	61.786
Pt	-16.702	-12.114	-9.35	-4.928	2.398	8.532	19.402	29.961	43.901	57.292
Au	-15.313	-10.653	-7.839	-4.944	3.852	7.421	20.625	32.521	46.123	60.729
Hg	-16.759	-13.231	-8.536	-4.918	-0.996	10.316	18.627	35.084	48.411	63.592
Tl	-12.681	-9.959	-7.169	-3.445	0.262	5.374	20.283	29.664	51.241	65.685
Pb	-17.805	-11.503	-8.099	-4.514	1.258	7	13.665	31.718	42.072	69.214
Bi	-19.831	-15.604	-8.583	-4.274	0.657	8.703	15.819	23.574	44.549	55.8
Po	-19.952	-16.22	-11.884	-3.524	1.837	8.242	18.015	26.155	34.801	58.617
At	-20.186	-16.306	-12.525	-8.093	3.046	9.951	17.674	28.819	37.836	47.285
Rn	-22.144	-16.231	-12.801	-7.871	-2.984	11.697	19.852	28.618	40.939	50.753

Table S5: Comparison of experimental[S1,S2] and CCSD(T)/aug-cc-pvqz ionization potentials. The signed deviation (eV) is reported as $\Delta IP_n = IP_n^{exp.} - IP_n^{CC}$.

	-1	0	1	2	3	4
H	0.01	-0.01	-	-	-	-
He	2.63	0.02	0.00	-	-	-
Li	0.00	0.02	0.71	0.03	-	-
Be	0.34	0.02	0.04	0.63	0.07	-
B	0.03	0.04	0.04	0.06	0.64	0.15
C	0.02	0.03	0.06	0.07	0.08	0.65
N	0.16	0.01	0.04	0.09	0.10	0.12
O	0.06	0.09	0.00	0.05	0.13	0.15
F	0.02	0.06	0.13	-0.01	0.04	0.19
Ne	5.29	0.02	0.09	0.19	-0.12	0.03
Na	0.00	0.12	-0.33	-3.11	-4.09	-1.66
Mg	0.22	0.06	0.17	0.13	-1.69	-2.54
Al	0.01	0.04	0.14	0.26	-0.05	-1.97
Si	-0.01	0.03	0.09	0.20	0.31	0.20
P	0.06	-0.03	0.02	0.13	0.19	0.38
S	0.04	0.10	-0.02	0.09	0.16	0.45
Cl	0.01	0.07	0.16	-0.01	0.40	0.36
Ar	2.76	0.02	0.11	0.24	0.65	0.44
Sc	0.71	0.04	-3.09	3.30	0.17	0.32
Ti	-0.16	0.06	-1.39	1.66	0.07	0.07
V	-0.08	-0.24	0.46	-0.07	-0.06	-0.03
Cr	0.06	-1.62	-0.83	2.26	-0.14	-0.27
Mn	-1.74	2.15	0.18	-3.23	2.58	-0.51
Fe	0.53	0.99	0.19	0.03	-0.34	-0.68
Co	-0.33	0.59	-0.33	0.17	-0.28	-4.43
Ni	1.56	0.22	-0.13	-0.44	-0.13	-0.70
Cu	-1.77	2.10	-0.22	-0.24	-4.05	0.79
Zn	0.52	0.21	0.34	-0.35	-0.57	-1.39
Ga	0.06	0.06	0.51	0.83	0.87	-
Ge	-0.15	-0.01	0.22	0.92	1.12	2.94
As	0.08	-0.19	0.12	0.40	1.37	1.27
Se	-0.02	0.18	-0.24	-0.70	0.57	2.05
Br	-0.13	0.01	0.52	0.79	0.60	0.85
Kr	2.11	-0.14	0.06	1.60	1.36	0.79

Table S6: Comparison of experimental[S1,S2] and CCSD(T)/def2qzvpp ionization energies. The signed deviation (eV) is reported as $\Delta IP_n = IP_n^{exp.} - IP_n^{CC}$.

	-1	0	1	2	3	4
H	0.46	-0.01	-	-	-	-
He	11.10	0.02	0.00	-	-	-
Li	0.04	0.02	0.22	0.03	-	-
Be	0.73	0.03	0.04	0.25	0.02	-
B	0.18	0.04	0.06	0.07	0.77	0.08
C	0.17	0.03	0.06	0.09	0.10	0.82
N	0.53	0.02	0.04	0.09	0.13	0.13
O	0.39	0.12	0.01	0.06	0.13	0.19
F	0.32	0.11	0.14	0.00	0.04	0.19
Ne	11.68	0.08	0.11	0.22	-0.10	0.05
Na	0.02	0.03	0.25	-0.52	-1.43	-1.39
Mg	0.52	0.04	0.05	0.20	-0.54	-1.17
Al	0.12	0.04	0.07	0.10	0.44	0.62
Si	0.09	0.03	0.08	0.12	0.16	0.42
P	0.27	-0.04	0.00	0.10	0.11	0.23
S	0.20	0.11	-0.03	0.06	0.12	0.35
Cl	0.14	0.09	0.15	-0.03	0.37	0.32
Ar	8.22	0.04	0.11	0.23	0.62	0.40
K	0.05	0.04	0.22	0.17	0.09	0.48
Ca	0.13	0.05	0.09	0.26	0.21	0.26
Sc	0.83	0.04	-3.01	3.32	0.12	0.02
Ti	-0.04	0.06	-1.38	1.74	0.09	-0.19
V	0.04	-0.23	0.46	0.02	-0.03	-0.25
Cr	0.18	-1.55	1.65	-0.14	-0.10	-0.43
Mn	-0.26	2.24	0.18	-0.13	-0.38	-0.59
Fe	0.80	1.09	0.20	0.16	-0.26	-0.72
Co	-0.27	0.61	-0.21	0.30	-0.12	-0.47
Ni	1.86	0.24	-0.02	-0.30	0.01	-0.59
Cu	-1.65	2.04	-0.10	-0.10	-2.29	-0.69
Zn	0.86	0.23	0.37	-0.23	-0.43	-1.23
Ga	0.25	0.06	0.40	0.58	-26.46	-
Ge	-0.01	-0.01	0.20	0.76	0.80	2.72
As	0.30	-0.19	0.09	0.35	1.14	0.87
Se	0.16	0.19	-0.26	-0.75	0.51	1.78
Br	0.01	0.03	0.51	0.75	0.54	0.77
Kr	8.85	-0.12	0.05	1.57	1.31	0.72

	-1	0	1	2	3	4
Rb	0.03	0.05	0.02	1.01	1.16	1.70
Sr	0.17	0.04	0.06	0.52	0.71	1.45
Y	0.75	0.40	-0.85	0.99	0.91	1.64
Zr	0.05	0.25	-1.85	-2.31	4.76	0.45
Nb	0.12	-0.10	-0.95	1.52	0.92	0.38
Mo	0.13	-1.47	1.60	0.17	6.44	7.11
Tc	0.34	0.24	0.07	-0.17	-	-
Ru	-0.45	0.67	0.33	0.55	-	-
Rh	1.18	0.03	0.06	0.56	-	-
Pd	0.19	0.03	0.16	0.07	-	-
Ag	0.08	0.05	0.03	0.19	-	-
Cd	0.88	0.06	0.06	-13.45	-	-
In	0.05	0.23	0.11	0.14	-1.72	-
Sn	-0.26	0.05	0.43	0.13	0.11	-31.14
Sb	0.29	-0.46	0.00	0.63	0.65	1.21
Te	0.02	0.33	-0.34	0.41	0.68	0.52
I	-0.16	-0.08	0.42	2.48	-	-
Xe	4.43	-0.29	0.11	1.64	-	-
Cs	0.02	0.03	1.84	-	-	-
Ba	-1.36	0.09	0.16	-	-	-
La	-0.34	0.25	-1.30	1.84	-	-
Hf	0.37	-0.13	0.93	-0.50	2.64	-
Ta	0.18	0.51	-	-	-	-
W	-0.13	-0.43	-	-	-	-
Re	0.50	0.25	-	-	-	-
Os	0.45	-0.29	-	-	-	-
Ir	-0.14	-0.05	-	-	-	-
Pt	-0.27	0.47	-0.84	-	-	-
Au	-1.54	1.80	-0.13	-	-	-
Hg	1.00	0.12	0.13	-0.88	-	-
Tl	0.12	0.73	0.15	0.17	-	-
Pb	-0.89	0.42	1.37	0.22	0.25	-0.41
Bi	0.29	-1.41	0.87	1.99	0.75	0.20
Po	0.06	0.18	-	-	-	-

Table S7: Comparison of CCSD(T)/def2qzvpp and CCSD(T)/aug-cc-pvqz ionization potentials. The signed deviation (eV) is reported as $\Delta IP_n = IP_n^{CC/def2} - IP_n^{CC/aug}$.

	-5	-4	-3	-2	-1	0	1	2	3	4
H	-25.84	-8.85	-70.34	37.44	-0.45	0.00	-	-	-	-
He	-42.30	-35.99	-29.66	-12.64	-8.47	0.00	0.00	-	-	-
Li	-5.28	-4.01	-2.69	-1.52	-0.04	0.00	0.49	0.00	-	-
Be	-8.64	-7.46	-4.59	-2.56	-0.38	-0.01	0.00	0.38	0.05	-
B	-7.25	-5.91	-4.22	-1.79	-0.15	0.00	-0.01	-0.01	-0.13	0.07
C	-11.50	-6.57	-4.51	-2.38	-0.15	-0.01	0.00	-0.02	-0.02	-0.16
N	-13.69	-12.98	-5.35	-2.61	-0.37	-0.01	-0.01	0.00	-0.03	-0.02
O	-15.06	-13.57	-10.71	-2.42	-0.33	-0.03	-0.01	0.00	0.00	-0.04
F	-27.95	-11.39	-13.06	-9.18	-0.31	-0.05	-0.02	-0.01	0.00	0.00
Ne	-10.90	-15.09	-9.92	-8.24	-6.39	-0.06	-0.03	-0.03	-0.03	-0.02
Na	-2.54	-5.03	-2.24	-1.30	-0.02	0.09	-0.58	-2.58	-2.66	-0.27
Mg	-6.76	-5.64	-3.51	-1.93	-0.30	0.03	0.12	-0.07	-1.15	-1.38
Al	-6.86	-4.85	-3.63	-1.36	-0.12	0.00	0.07	0.16	-0.49	-2.59
Si	-10.25	-5.48	-3.65	-1.66	-0.09	0.00	0.02	0.08	0.16	-0.23
P	-12.25	-10.64	-3.88	-1.58	-0.21	0.00	0.02	0.03	0.09	0.16
S	-12.16	-12.12	-6.43	-1.27	-0.16	-0.01	0.01	0.03	0.04	0.10
Cl	-7.43	-15.55	-10.13	-6.16	-0.13	-0.02	0.01	0.02	0.03	0.05
Ar	-11.42	-12.92	-9.38	-7.18	-5.46	-0.02	0.00	0.01	0.02	0.04
Sc	-4.88	-3.07	-3.17	-0.15	-0.12	0.00	-0.09	-0.02	0.04	0.29
Ti	0.25	-7.65	0.63	-3.56	-0.12	-0.01	-0.01	-0.08	-0.02	0.26
V	-4.78	-3.49	-2.72	-0.76	-0.13	-0.01	-0.01	-0.08	-0.03	0.22
Cr	-4.71	-3.57	-2.43	-1.33	-0.11	-0.07	-2.48	2.40	-0.04	0.16
Mn	-5.37	-3.31	-1.36	-0.68	-1.47	-0.09	-0.01	-3.10	2.96	0.08
Fe	-2.36	-6.69	-1.67	-1.11	-0.27	-0.10	-0.01	-0.14	-0.08	0.03
Co	-4.41	-4.92	-2.29	-1.56	-0.06	-0.02	-0.11	-0.12	-0.15	-3.96
Ni	-5.30	-5.82	-2.59	-1.44	-0.30	-0.02	-0.11	-0.15	-0.15	-0.11
Cu	-4.68	-3.77	-3.82	-0.30	-0.12	0.06	-0.12	-0.14	-1.76	1.48
Zn	-5.30	-5.60	-2.61	-2.14	-0.35	-0.03	-0.03	-0.13	-0.14	-0.15
Ga	-8.54	-6.62	-4.71	-1.83	-0.19	0.00	0.11	0.25	27.33	-27.28
Ge	-11.34	-6.77	-4.52	-2.07	-0.14	0.00	0.02	0.16	0.32	0.22
As	-12.24	-10.39	-3.91	-1.57	-0.22	0.00	0.02	0.05	0.22	0.40
Se	-11.29	-15.20	-7.70	-1.30	-0.18	-0.01	0.02	0.04	0.06	0.27
Br	-13.80	-11.81	-8.92	-7.71	-0.14	-0.02	0.02	0.04	0.06	0.08
Kr	-12.66	-17.42	-12.95	-5.40	-6.74	-0.02	0.01	0.04	0.05	0.07

Table S8: Comparison of electronegativity and idempotential (related to the neutral state) as reported or computed. Atoms from H to Sb. Values are expressed in eV.

	GMP		**experimental**[S1,S2]		**MEPO fit**		**CCSD(T)/def2qzvpp**	
Atom	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0
H	4.53	13.89	7.18	12.84	4.53	13.89	6.95	13.31
He	9.66	29.84	12.29	24.59			6.73	35.66
Li	3.01	4.77	3.01	4.77			2.97	4.80
Be	4.88	8.89	4.66	9.32			4.28	10.02
B	5.11	9.50	4.29	8.02			4.18	8.16
C	5.34	10.13	6.26	10.00	5.43	11.71	6.16	10.13
N	6.90	11.76	7.23	14.60	6.69	13.24	6.96	15.11
O	8.74	13.36	7.54	12.16	8.71	17.14	7.29	12.42
F	10.87	14.95	10.41	14.02	6.42	22.26	10.20	14.24
Ne	11.04	21.10	10.78	21.56			4.90	33.17
Na	2.84	4.59	2.84	4.59			2.82	4.58
Mg	3.95	7.39	3.82	7.65			3.55	8.13
Al	4.06	7.18	3.21	5.55			3.13	5.64
Si	4.17	6.97	4.77	6.76			4.71	6.82
P	5.46	8.00	5.60	9.71			5.49	10.01
S	6.93	8.97	6.22	8.28			6.06	8.37
Cl	8.56	9.89	8.29	9.35	5.82	14.55	8.18	9.40
Ar	9.47	12.71	7.88	15.76			3.75	23.93
K	2.42	3.84	2.42	3.84			2.37	3.85
Ca	3.23	5.76	3.07	6.09			2.98	6.17
Sc	3.40	6.16	3.36	6.35			2.93	7.15
Ti	3.47	6.76	3.45	6.74			3.44	6.64
V	3.65	6.82	3.63	6.22	4.09	8.43	3.73	6.49
Cr	3.42	7.73	3.72	6.09			4.41	7.82
Mn	3.33	8.21	3.72	7.44			2.73	4.93
Fe	3.76	8.28	4.01	7.72			3.07	7.43
Co	4.11	8.35	4.26	7.20			4.09	6.32
Ni	4.47	8.41	4.40	6.48			3.35	8.10
Cu	4.20	8.44	4.48	6.49	5.43	6.94	4.29	2.80
Zn	5.11	8.57	4.70	9.39	3.70	8.93	4.15	10.02
Ga	3.64	6.32	3.20	5.59			3.05	5.77
Ge	4.05	6.88	4.57	6.67			4.58	6.67
As	5.19	7.62	5.31	9.00			5.26	9.48
Se	6.43	8.26	5.89	7.73			5.72	7.70
Br	7.79	8.85	7.59	8.45	5.69	17.52	7.57	8.43
Kr	8.51	11.43	7.00	14.00			2.63	22.96
Rb	2.33	3.69	2.33	3.69			2.29	3.67
Sr	3.02	4.88	2.87	5.64			2.77	5.78
Y	3.83	5.62	3.34	6.07			2.77	6.42
Zr	3.40	7.10	3.63	6.41			3.48	6.21
Nb	3.55	6.76	3.89	5.99			3.88	6.21
Mo	3.47	7.51	3.92	6.35			4.60	7.95
Tc	3.29	7.98	3.92	6.73			3.63	6.82
Ru	3.58	8.03	4.21	6.32			4.10	5.21
Rh	3.98	8.01	4.30	6.32			3.70	7.46
Pd	4.32	8.00	4.45	7.78			4.35	7.94
Ag	4.44	6.27	4.44	6.27			4.38	6.30
Cd	5.03	7.91	4.50	8.99			4.03	9.81
In	3.51	5.79	3.10	5.38			2.96	5.21
Sn	3.99	6.25	4.23	6.23			4.33	5.93
Sb	4.90	6.68	4.84	7.59			4.93	8.34

Table S9: Comparison of electronegativity and idempotential (related to the neutral state) as reported or computed. Atoms from Te to Lr. Values are expressed in eV.

	GMP		**experimental**[S1,S2]		**MEPO fit**		**CCSD(T)/def2qzvpp**	
Atom	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0	χ_A^0	J_{AA}^0
Te	5.82	7.05	5.49	7.04			5.31	6.74
I	6.82	7.52	6.76	7.39	5.43	11.44	6.87	7.31
Xe	7.60	9.95	6.07	12.13			4.00	16.85
Cs	2.18	3.42	2.18	3.42			2.16	3.41
Ba	2.81	4.79	2.68	5.07			3.31	3.63
La	2.84	5.48	3.02	5.11			3.07	4.51
Ce	2.77	5.38	3.09	4.77				
Pr	2.86	5.13						
Nd	2.87	5.24						
Pm	2.88	5.35						
Sm	2.91	5.44						
Eu	2.88	5.58						
Gd	3.17	5.95						
Tb	3.02	5.67						
Dy	3.06	5.74						
Ho	3.13	5.78						
Er	3.19	5.83						
Tm	3.25	5.87						
Yb	3.29	5.93						
Lu	2.96	4.93						
Hf	3.70	6.80	3.50	7.00			3.38	7.50
Ta	5.10	5.70	4.11	7.57			3.76	7.24
W	4.63	6.62	4.40	7.17			4.68	7.47
Re	3.96	7.84	3.95	7.87			3.57	8.12
Os	5.14	7.26	4.89	7.62			4.81	8.36
Ir	5.00	8.00	5.33	7.54			5.43	7.45
Pt	4.79	8.86	5.56	6.87			5.47	6.13
Au	4.89	5.17	5.77	6.92			5.64	3.57
Hg	6.27	8.32	5.22	10.44			4.66	11.31
Tl	3.20	5.80	3.24	5.73			2.82	5.11
Pb	3.90	7.06	3.89	7.05			4.13	5.74
Bi	4.69	7.48	4.12	6.35			4.68	8.05
Po	4.21	8.42	5.16	6.52			5.04	6.41
At	4.75	9.50					6.50	6.91
Rn	5.37	10.74					4.36	14.68
Fr	2.00	4.00						
Ra	2.84	4.87						
Ac	2.84	5.67						
Th	3.18	5.81						
Pa	2.99	5.81						
U	3.34	5.71						
Np	3.55	5.43						
Pu	3.24	5.64						
Am	2.99	6.01						
Cm	2.83	6.38						
Bk	3.19	6.07						
Cf	3.20	6.20						
Es	3.33	6.18						
Fm	3.40	6.20						
Md	3.47	6.22						
No	3.48	6.35						
Lr	3.50	6.40						

Table S10: Radii of the isolated ions (Angstrom), computed at the UHF/def2qzvpp level of theory and used as input for the FC-Qeq and I-Qeq methods.

atom	-5	-4	-3	-2	-1	0	+1	+2	+3	+4	+5
H	1.396	1.422	1.464	1.430	1.100	0.752	0.000	0.000	0.000	0.000	0.000
He	0.909	0.914	0.923	0.938	0.791	0.487	0.397	0.000	0.000	0.000	0.000
Li	4.191	3.998	3.899	3.403	2.725	2.049	0.299	0.314	0.000	0.000	0.000
Be	2.665	2.537	2.374	2.120	1.777	1.402	1.208	0.258	0.226	0.000	0.000
B	2.227	2.078	1.890	1.628	1.321	1.084	0.953	0.867	0.187	0.177	0.000
C	1.690	1.560	1.413	1.230	1.017	0.873	0.781	0.727	0.679	0.227	0.146
N	1.356	1.253	1.111	0.985	0.848	0.729	0.661	0.614	0.590	0.559	0.108
O	1.166	1.072	0.960	0.826	0.723	0.637	0.573	0.535	0.508	0.497	0.475
F	1.002	0.932	0.849	0.750	0.627	0.562	0.512	0.474	0.450	0.433	0.429
Ne	0.944	0.884	0.812	0.728	0.627	0.501	0.461	0.430	0.405	0.389	0.378
Na	4.942	4.718	4.448	3.986	3.031	2.228	0.398	0.440	0.401	0.380	0.367
Mg	3.380	3.219	3.013	2.689	2.252	1.724	1.506	0.353	0.369	0.345	0.330
Al	2.755	2.597	2.398	2.135	1.817	1.519	1.289	1.187	1.496	0.322	0.304
Si	2.167	2.041	1.887	1.702	1.487	1.312	1.175	1.055	0.994	0.227	0.284
P	1.795	1.688	1.561	1.427	1.283	1.147	1.054	0.976	0.902	0.860	0.239
S	1.618	1.517	1.396	1.248	1.133	1.031	0.949	0.892	0.841	0.791	0.761
Cl	1.452	1.372	1.274	1.157	1.010	0.932	0.869	0.817	0.778	0.742	0.707
Ar	1.336	1.271	1.192	1.100	0.988	0.848	0.798	0.757	0.720	0.692	0.665
K	5.508	5.590	5.008	4.275	3.447	2.772	0.583	0.794	0.752	0.722	0.698
Ca	4.347	4.127	3.854	3.411	2.828	2.231	1.972	0.762	0.691	0.663	0.642
Sc	3.615	3.379	3.102	2.684	1.951	1.688	1.349	1.606	0.396	0.612	0.594
Ti	3.205	2.963	2.678	2.283	1.638	1.384	1.088	0.753	0.655	0.643	0.556
V	2.753	2.462	2.298	1.840	1.412	1.185	0.934	0.669	0.609	0.562	0.643
Cr	2.410	2.195	1.982	1.613	1.245	0.927	0.830	0.623	0.567	0.530	0.496
Mn	2.277	2.080	1.861	1.551	1.245	0.851	0.751	0.582	0.536	0.498	0.470
Fe	2.088	1.898	1.664	1.425	1.081	0.783	0.696	0.553	0.507	0.475	0.446
Co	1.847	1.661	1.496	1.252	0.929	0.725	0.577	0.524	0.485	0.453	0.428
Ni	1.621	1.488	1.382	1.157	0.912	0.677	0.546	0.497	0.463	0.435	0.412
Cu	1.635	1.463	1.270	1.052	0.810	0.682	0.517	0.475	0.444	0.420	0.399
Zn	1.516	1.368	1.206	1.020	0.824	0.641	0.540	0.454	0.424	0.399	0.380
Ga	1.360	1.235	1.100	0.951	0.798	0.660	0.553	0.479	0.408	0.433	0.365
Ge	1.194	1.092	0.987	0.874	0.757	0.656	0.570	0.494	0.434	0.373	0.355
As	1.122	1.029	0.930	0.830	0.732	0.643	0.574	0.511	0.451	0.399	0.345
Se	1.044	0.964	0.875	0.785	0.705	0.633	0.570	0.517	0.467	0.416	0.370
Br	0.992	0.921	0.831	0.757	0.679	0.618	0.565	0.517	0.474	0.431	0.387
Kr	0.936	0.885	0.821	0.732	0.669	0.601	0.556	0.515	0.476	0.439	0.402
Rb	6.846	6.196	5.547	4.780	3.772	2.930	0.484	0.938	0.899	0.868	0.834
Sr	4.270	4.140	3.793	3.445	2.985	2.418	2.148	1.095	0.844	0.817	0.796
Y	3.640	3.406	3.356	2.945	2.210	1.914	1.914	1.787	0.608	0.773	0.754
Zr	3.129	3.047	2.786	2.455	1.831	1.625	1.364	1.353	1.556	0.898	0.717
Nb	2.904	2.622	2.412	2.033	1.622	1.303	1.201	0.991	0.909	0.863	0.805
Mo	2.610	2.397	2.185	1.822	1.469	1.172	1.088	0.908	0.854	0.813	0.781
Tc	2.487	2.188	2.035	1.752	1.326	1.172	0.999	0.850	0.807	0.771	0.742
Ru	2.207	2.007	1.781	1.513	1.215	1.080	0.861	0.806	0.762	0.736	0.706
Rh	2.043	1.857	1.716	1.472	1.055	0.939	0.808	0.760	0.723	0.691	0.668
Pd	1.722	1.607	1.375	1.164	0.986	0.823	0.766	0.724	0.691	0.663	0.639
Ag	1.935	1.846	1.552	1.246	0.988	0.835	0.727	0.691	0.661	0.637	0.615
Cd	1.658	1.504	1.364	1.189	1.006	0.834	0.741	0.660	0.684	0.612	0.593
In	1.771	1.601	1.413	1.205	1.004	0.852	0.744	0.675	0.608	0.588	0.572
Sn	1.542	1.413	1.264	1.107	0.953	0.843	0.756	0.679	0.622	0.564	0.592
Sb	1.388	1.279	1.156	1.037	0.925	0.829	0.758	0.694	0.632	0.583	0.531
Te	1.275	1.195	1.094	0.982	0.893	0.816	0.751	0.696	0.644	0.592	0.548
I	1.221	1.142	1.052	0.961	0.862	0.797	0.742	0.692	0.647	0.603	0.557
Xe	1.168	1.105	1.032	0.939	0.861	0.778	0.730	0.687	0.646	0.607	0.568
Cs	6.467	6.100	5.835	5.309	4.210	3.207	0.976	1.134	1.093	1.064	1.039
Ba	4.831	4.610	4.357	3.900	2.769	2.705	2.417	0.352	1.042	1.017	1.000
La	3.828	3.802	3.411	3.357	2.507	1.965	1.851	2.038	0.174	0.961	0.940
Hf	1.737	1.653	1.494	1.280	1.134	1.009	0.891	0.830	0.783	0.690	0.674
Ta	1.649	1.517	1.413	1.239	1.079	0.974	0.869	0.805	0.777	0.742	0.660
W	1.548	1.449	1.314	1.176	1.032	0.904	0.816	0.768	0.729	0.695	0.664
Re	1.515	1.409	1.279	1.154	1.028	0.907	0.820	0.813	0.720	0.689	0.662
Os	1.534	1.368	1.277	1.094	0.947	0.874	0.825	0.761	0.704	0.679	0.655
Ir	1.464	1.309	1.183	1.048	0.911	0.845	0.765	0.721	0.692	0.667	0.647
Pt	1.325	1.261	1.139	0.957	0.877	0.816	0.736	0.705	0.677	0.655	0.634
Au	1.399	1.277	1.141	0.997	0.847	0.790	0.714	0.686	0.662	0.642	0.623
Hg	1.287	1.190	1.088	0.978	0.867	0.763	0.712	0.668	0.647	0.628	0.611
Tl	1.464	1.330	1.185	1.029	0.884	0.780	0.711	0.671	0.634	0.617	0.602
Pb	1.285	1.186	1.076	0.964	0.856	0.779	0.720	0.669	0.635	0.603	0.589
Bi	1.186	1.100	1.008	0.921	0.840	0.771	0.720	0.676	0.634	0.605	0.576
Po	1.159	1.069	0.988	0.896	0.824	0.764	0.716	0.677	0.640	0.605	0.579
At	1.117	1.044	0.966	0.882	0.805	0.754	0.712	0.675	0.642	0.610	0.579
Rn	1.087	1.030	0.962	0.875	0.810	0.742	0.705	0.671	0.640	0.612	0.584

2 EQeq charge centers

Table S11: For the elements included in the def2qzvpp basis set, the charge centers used as input for the EQeq program, are reported. The lowest common oxidation states[S3] is generally adopted. It is set to zero for non-metal atoms or lowered in case of unavailable experimental data.

atom	EQeq/ox input charge center	atom	EQeq/ox input charge center
H	0	Tc	2
He	0	Ru	2
Li	1	Rh	2
Be	2	Pd	2
B	0	Ag	1
C	0	Cd	2
N	0	In	3
O	0	Sn	2
F	0	Sb	3
Ne	0	Te	0
Na	1	I	0
Mg	2	Xe	0
Al	3	Cs	1
Si	4	Ba	1
P	0	La	2
S	0	Ce	3
Cl	0	Pr	3
Ar	0	Nd	1
K	1	Pm	1
Ca	2	Sm	1
Sc	3	Eu	1
Ti	4	Gd	1
V	4	Tb	1
Cr	3	Dy	1
Mn	2	Ho	1
Fe	2	Er	1
Co	2	Tm	2
Ni	2	Yb	2
Cu	2	Lu	1
Zn	2	Hf	3
Ga	3	Ta	0
Ge	2	W	0
As	0	Re	0
Se	0	Os	0
Br	0	Ir	0
Kr	0	Pt	1
Rb	1	Au	1
Sr	2	Hg	1
Y	3	Tl	1
Zr	4	Pb	2
Nb	4	Bi	3
Mo	4	Po	0

3 Structures containing ClO_4^- ions

Table S12: List of structures containing positive DDEC charges on Cl: this feature is attributed to the presence of perchlorate floating anions inside the pores of the frameworks.

MOF's code	DDEC Cl charge
FAZRAA	+1.57
IPIDAN	+1.53
ITIWOY	+1.67
MIFQEY	+1.54
OKITIM01	+1.53
OKITIM02	+1.52
OKITIM	+1.80
OKIVAG01	+1.61
OKIVAG	+1.62
OKIVIO	+1.80
OKIWEL01	+1.50
OKIWEL02	+1.33
OKIWEL	+1.47
OKIWUB01	+1.90
OKIWUB	+1.90
OKIXIQ01	+1.81
OKIXIQ02	+1.83
OKIXIQ	+1.85
OKIYAJ	+2.06
OKIYIR03	+1.37
OKIYIR	+1.29
SAHYAD01	+1.72
SAHYAD	+1.93
SAHYIL	+2.00
TAMJEY	+1.57
WUNSEE01	+1.75
WUNSEE02	+1.51
WUNSEE	+1.79
WUNSII01	+1.61
WUNSII02	+1.36
WUNSII	+1.37
WUNTAB	+1.56
WUNVIL	+1.91

4 Charge comparison for Cl and F

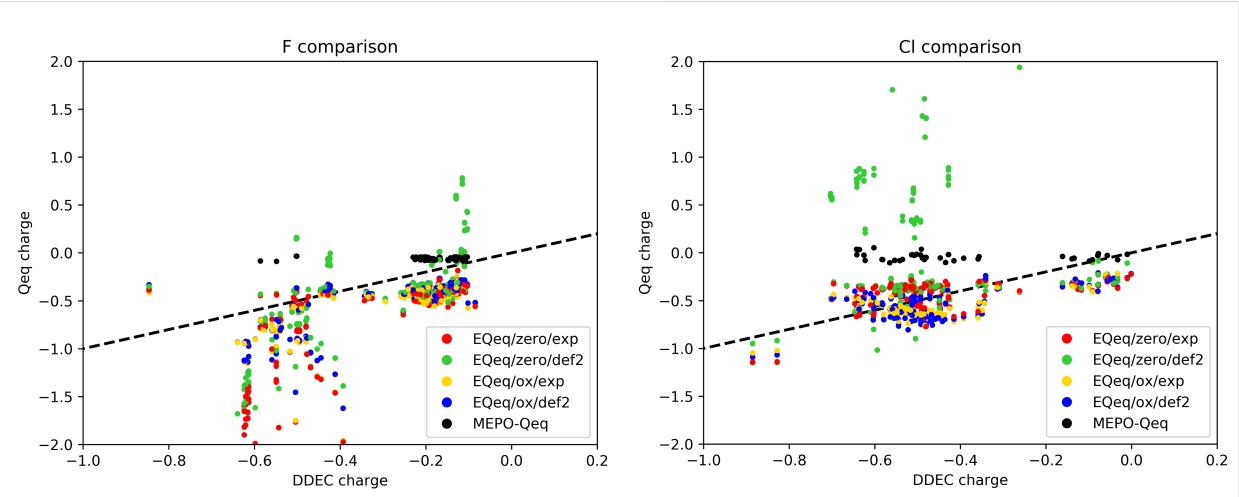

Figure S1: Comparison of point charges for the F and Cl atom types that are contained in the 2338 structures used as dataset in this work. Charges from calculations that are not converged, or from frameworks that contain elements that were not parametrized for MEPO-Qeq, are not shown.

5 Speed benchmark for the Qeq codes

Table S13: Speed benchmark for 13 common MOFs, on a 2.6 GHz Intel Ivy Bridge CPU and 3GB of RAM: the running time is reported in seconds. For Mg-MOF-74 the unit cell has been replicated on the xyz directions to test the performance for different number of atoms and volume. The EQeq method has been tested in its original form, applying a default expansion of the unit cell by 5x5x5 and the more precise expansion used in this work, i.e., 13x13x13.

Framework (UC)	**Atoms/UC**	**Vol./UC(Å^3)**	**FC-Qeq**	**I-Qeq**	**PQeq**	**EQeq (5)**	**EQeq (13)**
HKUST-1 (DOTSOV02)	156	4527.8	1	1	2	4	58
IRMOF-1 (cubic)	424	17237.4	6	7	10	27	431
Mg-MOF-74 (111)	54	1359.4	<1	<1	1	<1	7
Mg-MOF-74 (211)	108	2718.8	<1	1	2	2	27
Mg-MOF-74 (222)	432	10875.5	4	4	10	28	442
Mg-MOF-74 (322)	648	16313.3	11	13	16	60	970
Mg-MOF-74 (333)	1458	36705	60	64	91	341	4935
Mg-MOF-74 (422)	864	21751.1	23	25	25	111	1757
Mg-MOF-74 (444)	3456	87004.4	718	763	697	2496	27551
Mg-MOF-74 (555)	6750	169930.7	crash	crash	3939	11887	106881
MIL-100 (cubic)	11152	387511.3	crash	crash	crash	38113	>250000
MIL-100 (primitive)	2788	96877.7	322	diverge	424	1389	18964
UIO-66 (cubic)	432	8870.2	6	7	15	26	449

6 Partial charges of CO_2

In this work we focused on the partial charges of the frameworks. However, it is interesting to compare the different charges that can be derived for the adsorbate molecule. In the TraPPE force fields, charges have been fitted together with the dispersion parameters, to better reproduce experimental data such as the vapour-liquid equilibrium (VLE).[S4] The parameters are therefore meant to be used for molecule-molecule interactions, but this does not imply that also the molecule-framework interactions are properly described. Contrary to dispersion parameter, where we can use two different potentials for the molecule-molecule and molecule-framework interaction, in the case of charges, a compromise should be found to describe both Coulombic interactions. Indeed, for each atom (or dummy bead) only one point charge can be assigned. Table S14 reports the charges that one can obtain for CO_2 from different schemes, placing the molecule at the center of a 30x30x30 Angstrom box. The CO bonds are the same as for TraPPE, i.e., the experimental value of 1.16 Angstrom. For Qeq and EQeq the standard parameters proposed for the method are used, i.e., GMP and experimental, respectively. As for the electron density derived charges, the calculation is based on the PBE electronic structure calculated with CP2K[S5] using the following settings: 700 Ry cutoff, 50 Ry relative cutoff with 4 grids, TZVP-MOLOPT-GTH gaussian basis set and GTH-PBE pseudopotentials. Mulliken, Hirshfeld and REPEAT charges are computed using the routines implemented in CP2K. DDEC charges are computed using the latest release of the program Chargemol.[S6]

Table S14: Comparison of the partial charge on CO_2 as obtained from different methods.

Method	C charge	O charge
TraPPE	0.70	-0.35
PQeq	0.89	-0.45
EQeq	0.41	-0.21
Mulliken	0.32	-0.16
Hirshfeld	0.61	-0.30
REPEAT	0.64	-0.32
DDEC	0.73	-0.37

It is interesting to notice how the DFT derived charges that are supposed to fit the electrostatic potential (i.e., REPEAT and DDEC) get closer to the TraPPE's partial charges. In particular the closest agreement is obtained with DDEC: therefore, we can

say that the TraPPE's partial charges are consistent with the method we used to compute the partial charges of the framework. On the other side, PQeq and EQeq show larger deviations.

7 Statistical coefficients used for the comparisons

7.1 Mean signed deviation

Given two sets, $\{X\}$ and $\{Y\}$, composed by N values each:

$$\mathrm{MSD} = \sum_{i}^{N} \frac{X_i - Y_i}{n} \tag{1}$$

were Y is taken as the reference set. Notice that positive and negative deviations may cancel each other out. Indeed, the mean signed deviation is only informative if compared to the mean absolute deviation: if the two values are the same (opposite) one can identify a systematic overestimation (underestimation) with respect to the reference set.

7.2 Mean absolute deviation

$$\mathrm{MAD} = \sum_{i}^{N} \frac{|X_i - Y_i|}{n} \tag{2}$$

7.3 Pearson coefficient

The Pearson coefficient, ρ_{XY}, which quantifies the linear correlation between two sets of data, is defined as:

$$\rho_{XY} = \frac{\sum_i^N (X_i - \overline{X})(Y_i - \overline{Y})}{\sqrt{\sum_i^N (X_i - \overline{X})^2}\sqrt{\sum_i^N (Y_i - \overline{Y})^2}} \tag{3}$$

where $\overline{X}$ and $\overline{Y}$, are the mean values over the two sets. The coefficient can span the range from -1 to 1: if ρ_{XY} is null the two sets are linearly uncorrelated.

7.4 Spearman coefficient

To compute the Spearman correlation coefficient r_s, the two sets of data $\{X\}$ and $\{Y\}$, are first converted to ranks rgX and rgY. Then the Pearson coefficient is computed from

these ranks:

$$\rho_{XY} = \frac{\sum_i^N (rgX_i - \overline{rgX})(rgY_i - \overline{rgY})}{\sqrt{\sum_i^N (rgX_i - \overline{rgX})^2}\sqrt{\sum_i^N (rgY_i - \overline{rgY})^2}} \tag{4}$$

Two sets of data that are correlated in a non-linear fashion, e.g., exponentially, can give a near zero Pearson coefficient but they will lead to a Spearman coefficient close to 1 or -1. If instead the correlation is linear, the two coefficient must be similar.

References

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(S2) Moore, C. E. *Ionization potentials and ionization limits derived from the analyses of optical spectra*; 1970.

(S3) Greenwood, N. N.; Earnshaw, A. *Chemistry of the Elements*; Butterworth-Heinemann, 1997.

(S4) Potoff, J. J.; Siepmann, J. I. Vapor liquid equilibria of mixtures containing alkanes, carbon dioxide, and nitrogen. *AIChE J.* **2001**, *47*, 1676–1682.

(S5) CP2K code and reference material is available at the project web page http://www.cp2k.org.

(S6) Chargemol, release 09_26_2017, available at https://sourceforge.net/projects/ddec/.