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Supporting Information

Mixed Two-Dimensional Organic-Inorganic Halide Perovskites for Highly Efficient and Stable Photovoltaic Application

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Table S1. Calculated lattice constants (Å) of $BA_2MA_2B_3X_{10}$ (B = Pb^{2+} or Sn^{2+}; X = Br^- or I^-) with the mixed Cs atom.

Rb atom	SnI-based			PbI-based			SnBr-based			PbBr-based		
	a	b	c	a	b	c	a	b	c	a	b	c
0%	8.83	8.83	26.57	8.92	8.92	26.69	8.34	8.34	25.55	8.42	8.42	25.65
25%	8.79	8.79	26.5	8.91	8.91	26.63	8.33	8.33	25.28	8.4	8.4	25.61
50%	8.8	8.8	26.34	8.82	8.82	26.76	8.31	8.31	25.17	8.3	8.3	25.59
75%	8.77	8.77	26.3	8.76	8.76	26.64	8.24	8.24	25.18	8.26	8.26	25.58
100%	8.69	8.69	26.37	8.76	8.76	26.63	8.15	8.15	25.3	8.23	8.23	25.61

Table S2. Calculated lattice constants (Å) of $BA_2MA_2B_3X_{10}$ (B = Pb^{2+} or Sn^{2+}; X = Br^- or I^-) with the mixed Rb atom.

Cs atom	SnI-based			PbI-based			SnBr-based			PbBr-based		
	a	b	c	a	b	c	a	b	c	a	b	c
0%	8.83	8.83	26.57	8.92	8.92	26.69	8.34	8.34	25.55	8.42	8.42	25.65
25%	8.84	8.84	26.49	8.85	8.85	26.75	8.33	8.33	25.44	8.4	8.4	25.65
50%	8.83	8.83	26.44	8.85	8.85	26.77	8.33	8.33	25.27	8.33	8.33	25.7
75%	8.82	8.82	26.4	8.83	8.83	26.97	8.32	8.32	25.24	8.33	8.33	25.7
100%	8.8	8.8	26.48	8.8	8.8	26.85	8.28	8.28	25.15	8.28	8.28	25.75

Table S3. Calculated lattice constants (Å) of $BA_2MA_2Sn_3I_{10}$ with the mixed Ge atom.

Ge atom	SnI-based			SnBr-based		
	a	b	c	a	b	c
0%	8.83	8.83	26.57	8.34	8.34	25.55
16.67%	8.83	8.83	26.42	8.33	8.33	25.37
33.33%	8.81	8.81	26.35	8.33	8.33	25.22
50%	8.75	8.75	26.25	8.29	8.29	25.18
66.67%	8.72	8.72	26.17	8.25	8.25	25.25
83.33%	8.71	8.71	26.04	8.24	8.24	25.23

Table S4. Calculated lattice constants (Å) of $BA_2MA_2Sn_3I_{10}$ with the mixed Pb atom.

Pb atom	SnI-based			SnBr-based		
	a	b	c	a	b	c
0%	8.83	8.83	26.57	8.34	8.34	25.55
16.67%	8.86	8.86	26.54	8.31	8.31	25.51
33.33%	8.88	8.88	26.57	8.38	8.38	25.53
50%	8.89	8.89	26.59	8.38	8.38	25.53
66.67%	8.89	8.89	26.65	8.41	8.41	25.53
83.33%	8.91	8.91	26.77	8.4	8.4	25.63

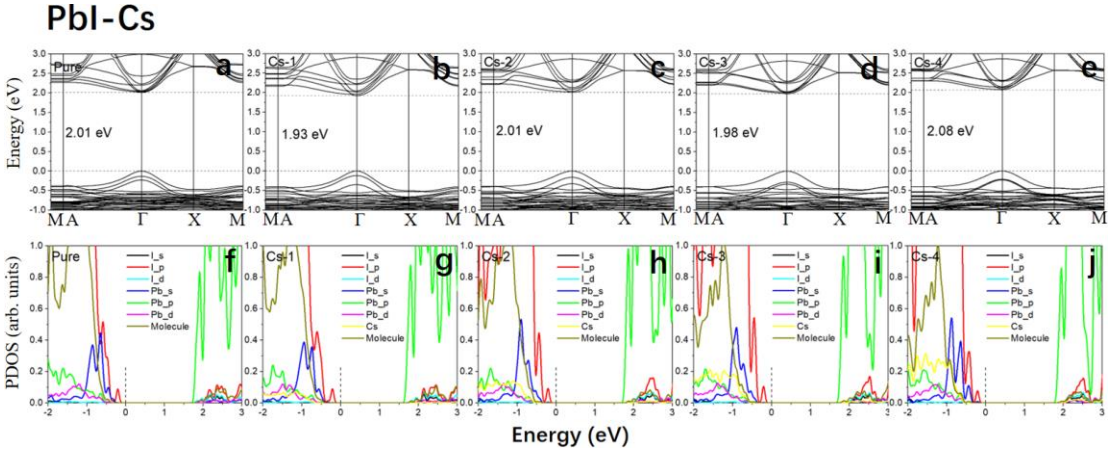

Figure S1. Calculated band structures of PbI-based 2D HOIP by mixing Cs atom with different concentrations: (a) pure (0%), (b) Cs-1 (25%), (c) Cs-2 (50%), and (d) Cs-3 (75%), (e) Cs-4 (100%). Calculated partial densities of states (PDOSs) of PbI-based 2D HOIP by mixing Cs atom with different concentrations: (f) pure, (g) Cs-1, (h) Cs-2, (i) Cs-3, and (j) Cs-4.

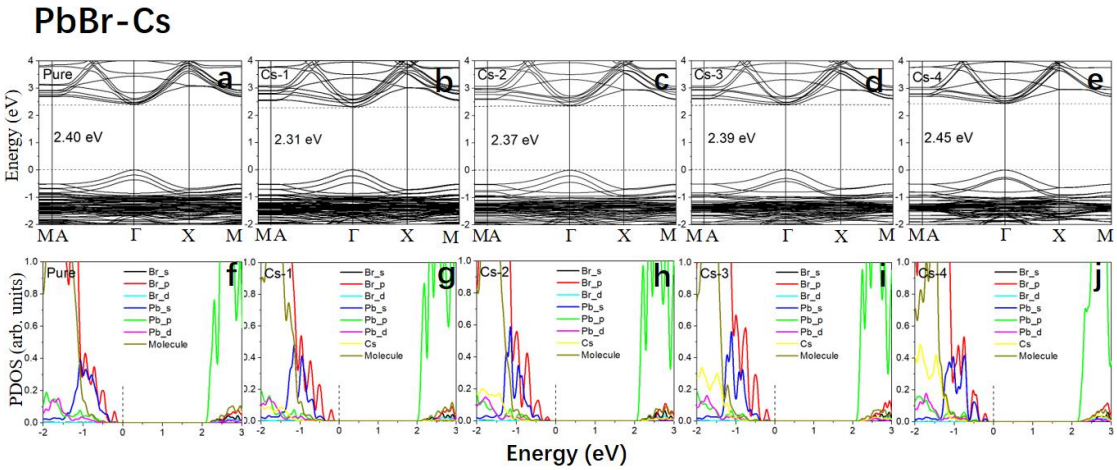

Figure S2. Calculated band structures of PbBr-based 2D HOIP by mixing Cs atom with different concentrations: (a) pure, (b) Cs-1, (c) Cs-2, (d) Cs-3, and (e) Cs-4. Calculated partial densities of states (PDOSs) of PbBr-based 2D HOIP by mixing Cs atom with different concentrations: (f) pure, (g) Cs-1, (h) Cs-2, (i) Cs-3, and (j) Cs-4.

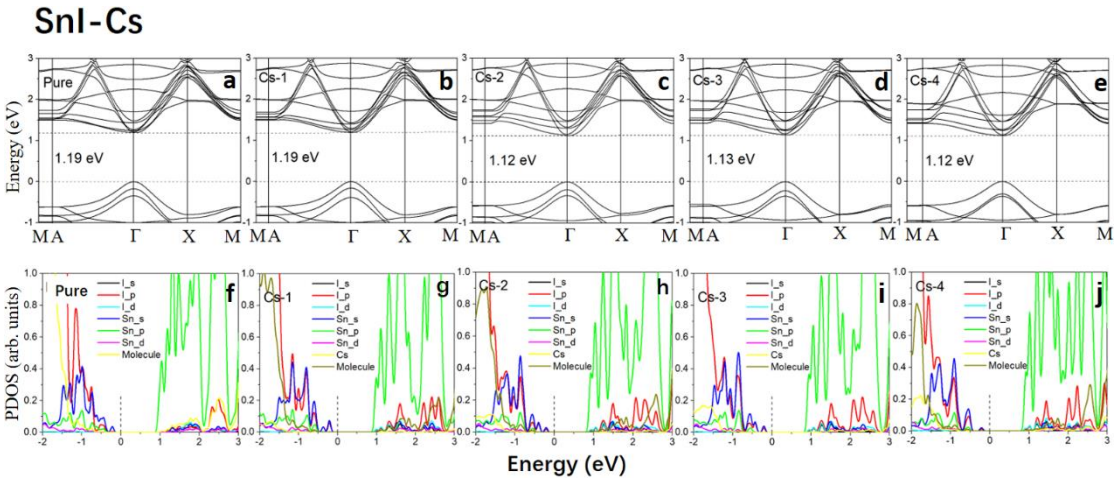

Figure S3. Calculated band structures of SnI-based 2D HOIP by mixing Cs atom with different concentrations: (a) pure, (b) Cs-1, (c) Cs-2, (d) Cs-3, and (e) Cs-4. Calculated partial densities of states (PDOSs) of SnI-based 2D HOIP by mixing Cs atom with different concentrations: (f) pure, (g) Cs-1, (h) Cs-2, (i) Cs-3, and (j) Cs-4.

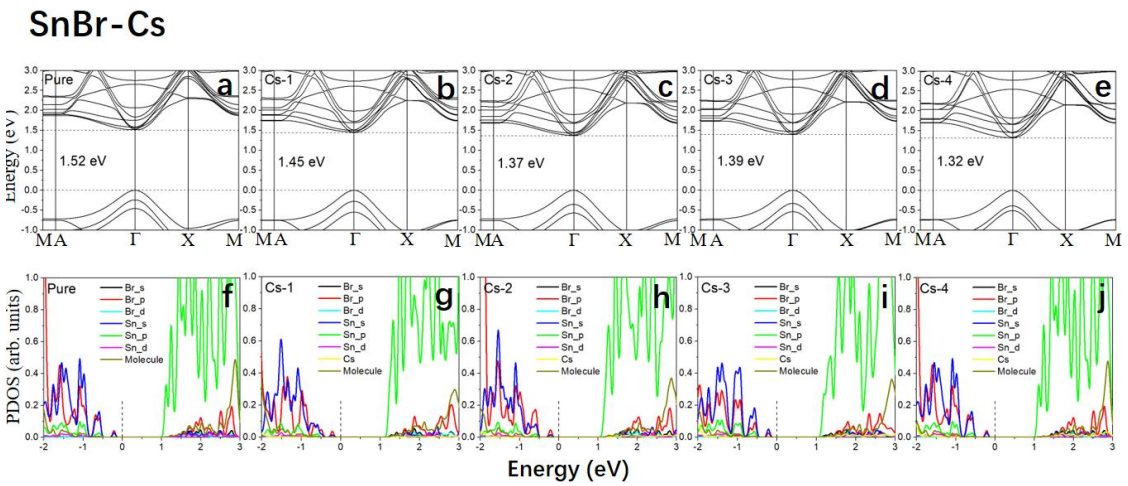

Figure S4. Calculated band structures of SnBr-based 2D HOIP by mixing Cs atom with different concentrations: (a) pure, (b) Cs-1, (c) Cs-2, (d) Cs-3, and (e) Cs-4. Calculated partial densities of states (PDOSs) of SnBr-based 2D HOIP by mixing Cs atom with different concentrations: (f) pure, (g) Cs-1, (h) Cs-2, (i) Cs-3, and (j) Cs-4.

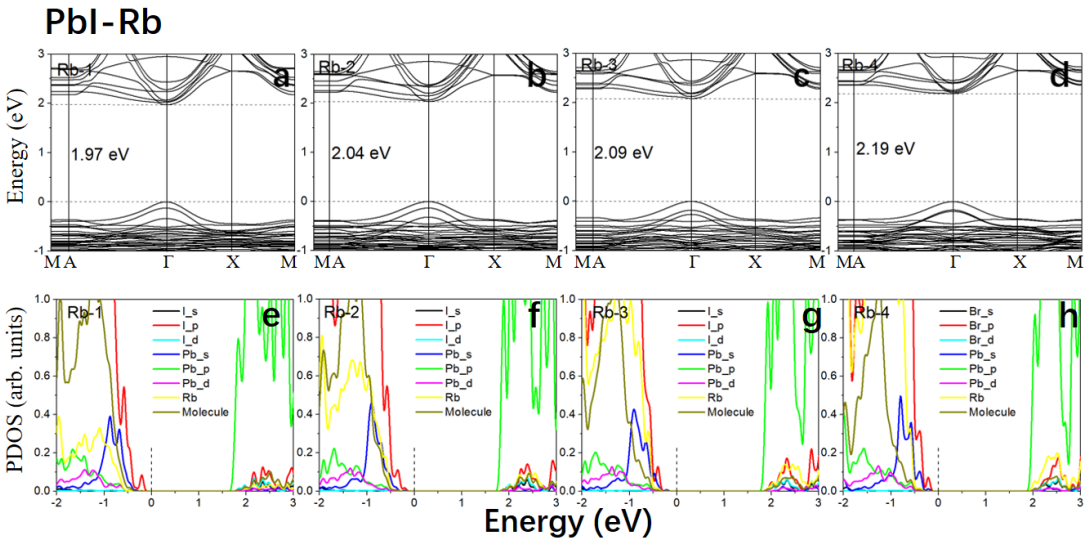

Figure S5. Calculated band structures of PbI-based 2D HOIP by mixing Rb atom with different concentrations: (a) Rb-1 (25%), (b) Rb-2 (50%), (c) Rb-3 (75%), and (d) Rb-4 (100%). Calculated partial densities of states (PDOSs) of PbI-based 2D HOIP by mixing Rb atom with different concentrations: (e) Rb-1, (f) Rb-2, (g) Rb-3, and (h) Rb-4.

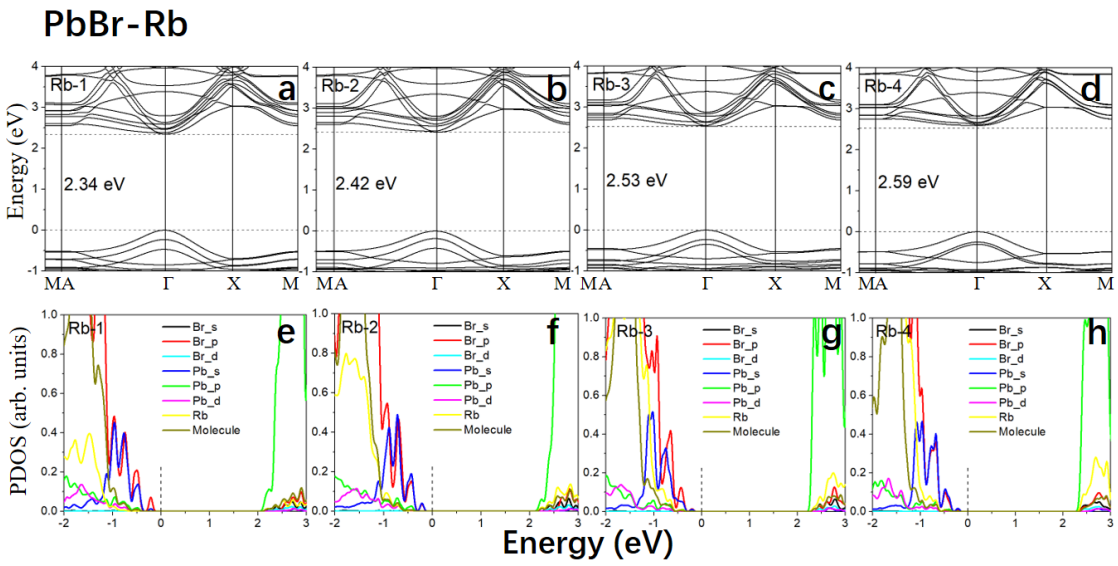

Figure S6. Calculated band structures of PbBr-based 2D HOIP by mixing Rb atom with different concentrations: (a) Rb-1, (b) Rb-2, (c) Rb-3, and (d) Rb-4. Calculated partial densities of states (PDOSs) of PbBr-based 2D HOIP by mixing Rb atom with different concentrations: (e) Rb-1, (f) Rb-2, (g) Rb-3, and (h) Rb-4.

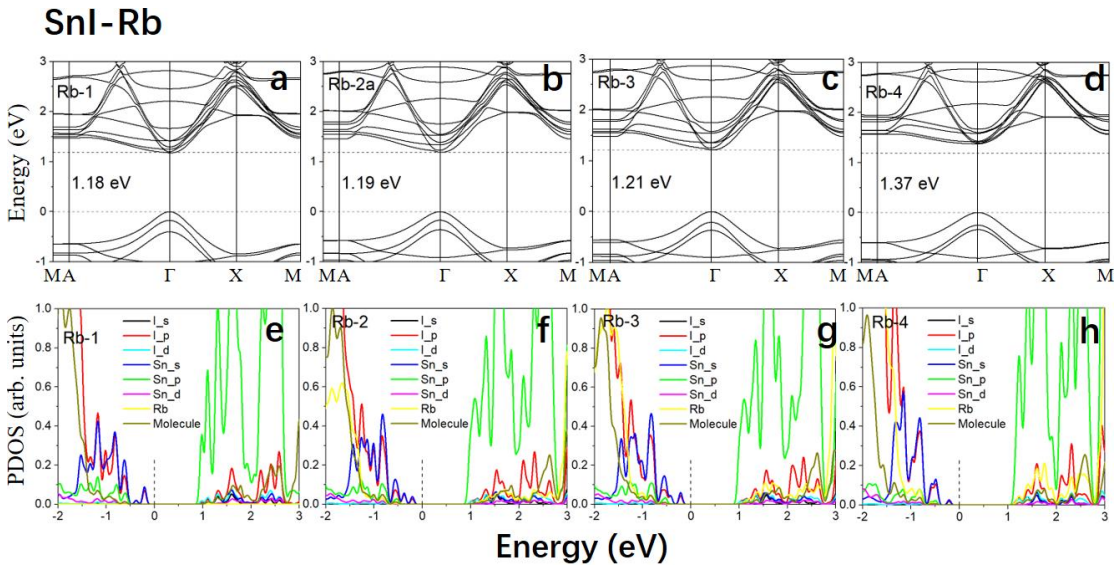

Figure S7. Calculated band structures of SnI-based 2D HOIP by mixing Rb atom with different concentrations: (a) pure, (b) Rb-1, (c) Rb-2, (d) Rb-3. Calculated partial densities of states (PDOSs) of SnI-based 2D HOIP by mixing Rb atom with different concentrations: (e) pure, (f) Rb-1, (g) Rb-2, and (h) Rb-3.

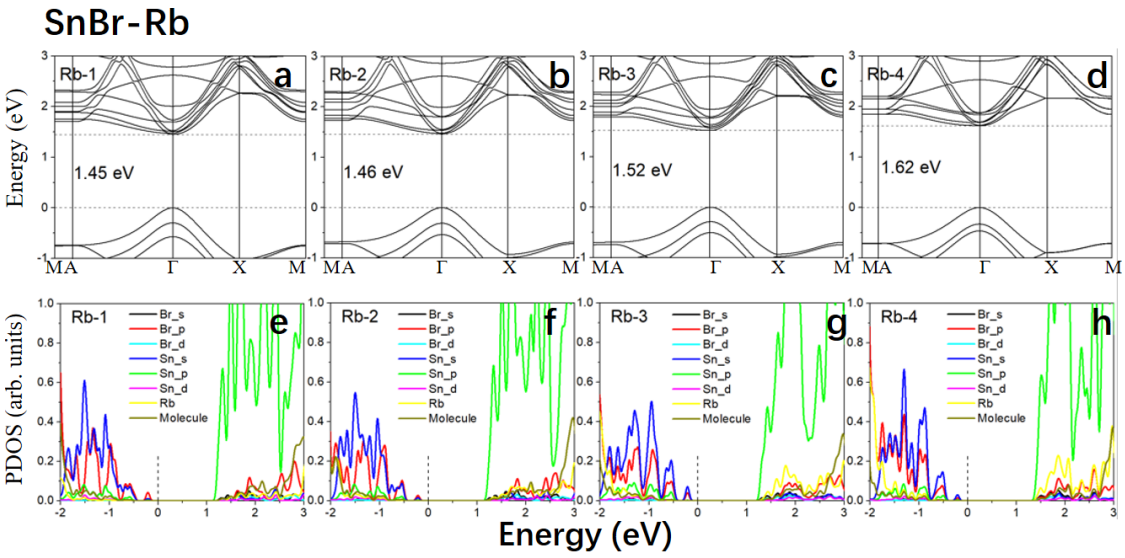

Figure S8. Calculated band structures of SnBr-based 2D HOIP by mixing Rb atom with different concentrations: (a) Rb-1, (b) Rb-2, (c) Rb-3, (d) Rb-4. Calculated partial densities of states (PDOSs) of SnBr-based 2D HOIP by mixing Rb atom with different concentrations: (e) Rb-1, (f) Rb-2, (g) Rb-3, and (h) Rb-4.

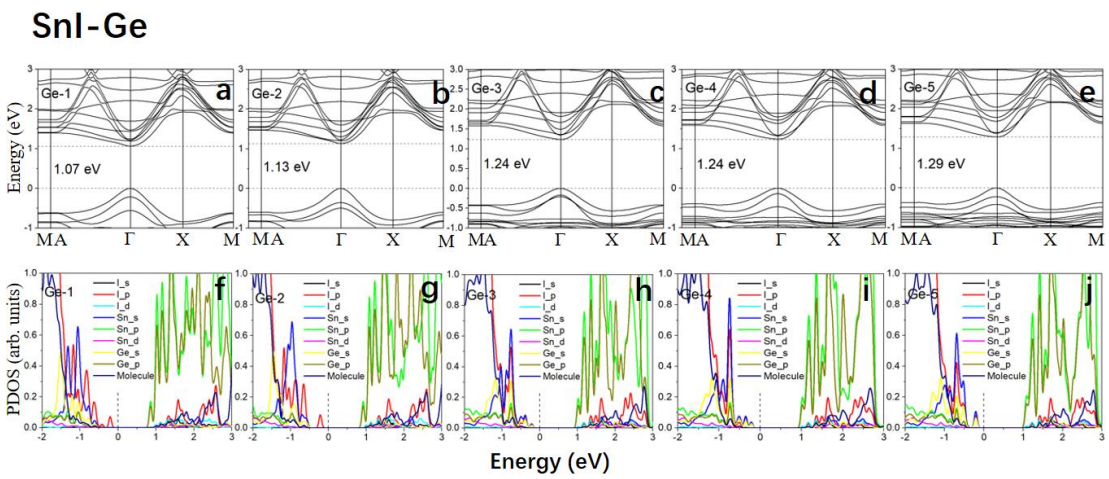

Figure S9. Calculated band structures of SnI-based 2D HOIP by mixing Ge atom with different concentrations: (a) Ge-1 (16.67%), (b) Ge-2 (33.33%), (c) Ge-3 (50%), (d) Ge-4 (66.67%), (e) Ge-5 (83.33%). Calculated partial densities of states (PDOSs) of SnI-based 2D HOIP by mixing Ge atom with different concentrations: (f) Ge-1, (g) Ge-2, (h) Ge-3, (i) Ge-4, and (j) Ge-5.

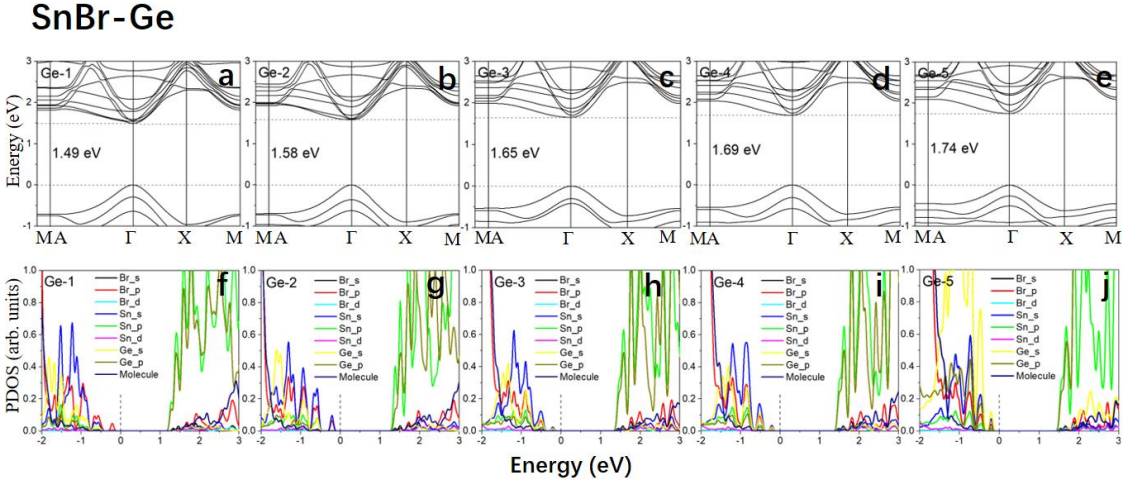

Figure S10. Calculated band structures of SnBr-based 2D HOIP by mixing Ge atom with different concentrations: (a) Ge-1, (b) Ge-2, (c) Ge-3, (d) Ge-4, (e) Ge-5. Calculated partial densities of states (PDOSs) of SnBr-based 2D HOIP by mixing Ge atom with different concentrations: (f) Ge-1, (g) Ge-2, (h) Ge-3, (i) Ge-4, and (j) Ge-5.

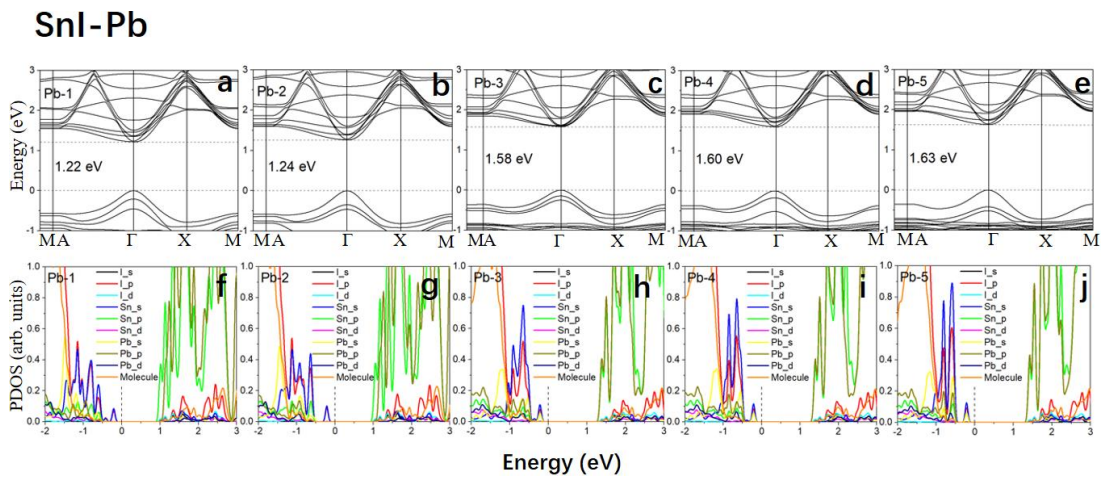

Figure S11. Calculated band structures of SnI-based 2D HOIP by mixing Pb atom with different concentrations: (a) Pb-1 (16.67%), (b) Pb-2 (33.33%), (c) Pb-3 (50%), (d) Pb-4 (66.67%), and (e) Pb-5 (83.33%). Calculated partial densities of states (PDOSs) of SnI-based 2D HOIP by mixing Pb atom with different concentrations: (f) Pb-1, (g) Pb-2, (h) Pb-3, (i) Pb-4, and (j) Pb-5.

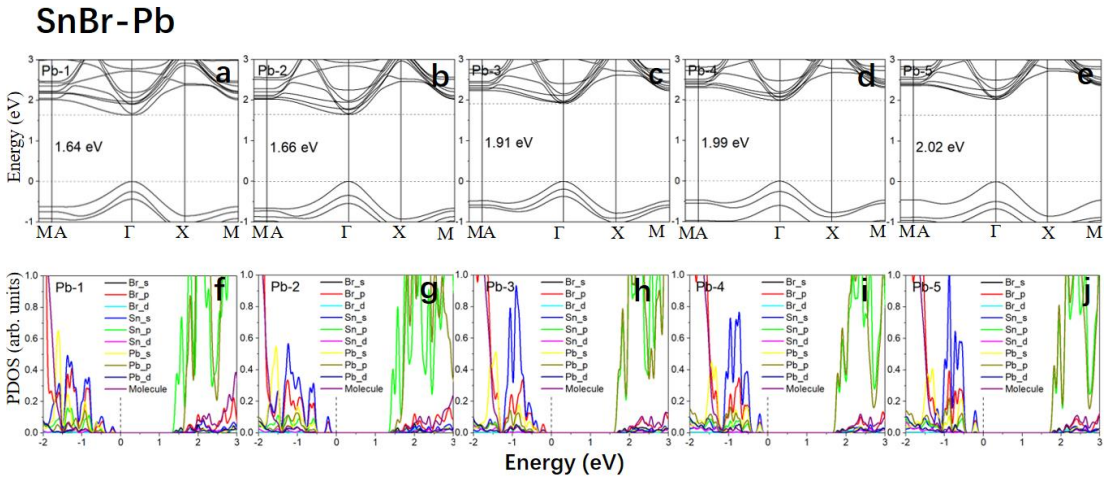

Figure S12. Calculated band structures of SnBr-based 2D HOIP by mixing Pb atom with different concentrations: (a) Pb-1, (b) Pb-2, (c) Pb-3, (d) Pb-4, (e) Pb-5. Calculated partial densities of states (PDOSs) of SnBr-based 2D HOIP by mixing Pb atom with different concentrations: (f) Pb-1, (g) Pb-2, (h) Pb-3, (i) Pb-4, and (j) Pb-5.

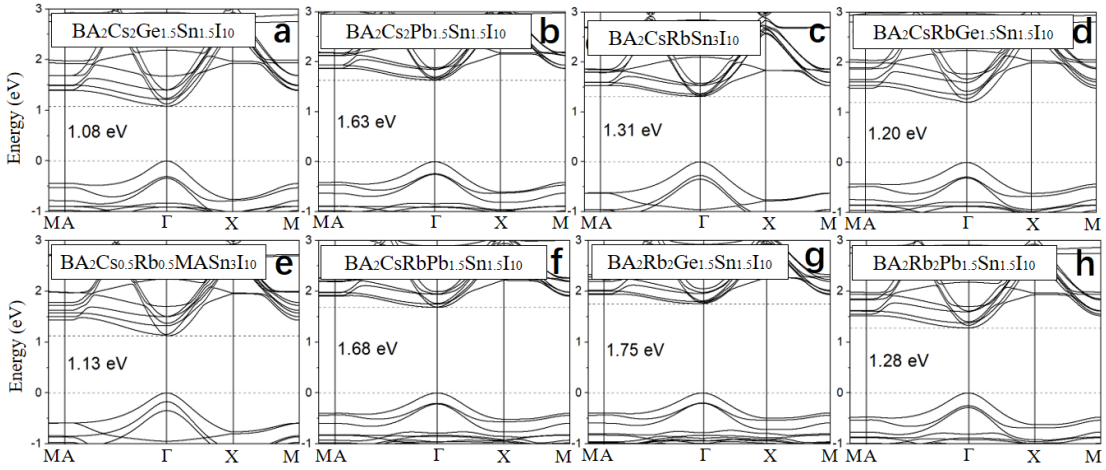

Figure S13. Calculated band structures of the various atoms (Cs, Rb, Ge, and Pb) mixed 2D SnI-based system: (a) $BA_2Cs_2Ge_{1.5}Sn_{1.5}I_{10}$, (b) $BA_2Cs_2Pb_{1.5}Sn_{1.5}I_{10}$, (c) $BA_2CsRbSn_3I_{10}$, (d) $BA_2CsRbGe_{1.5}Sn_{1.5}I_{10}$, (e) $BA_2Cs_{0.5}Rb_{0.5}MASn_3I_{10}$, (f) $BA_2CsRbPb_{1.5}Sn_{1.5}I_{10}$, (g) $BA_2Rb_2Ge_{1.5}Sn_{1.5}I_{10}$, and (h) $BA_2Rb_2Pb_{1.5}Sn_{1.5}I_{10}$.

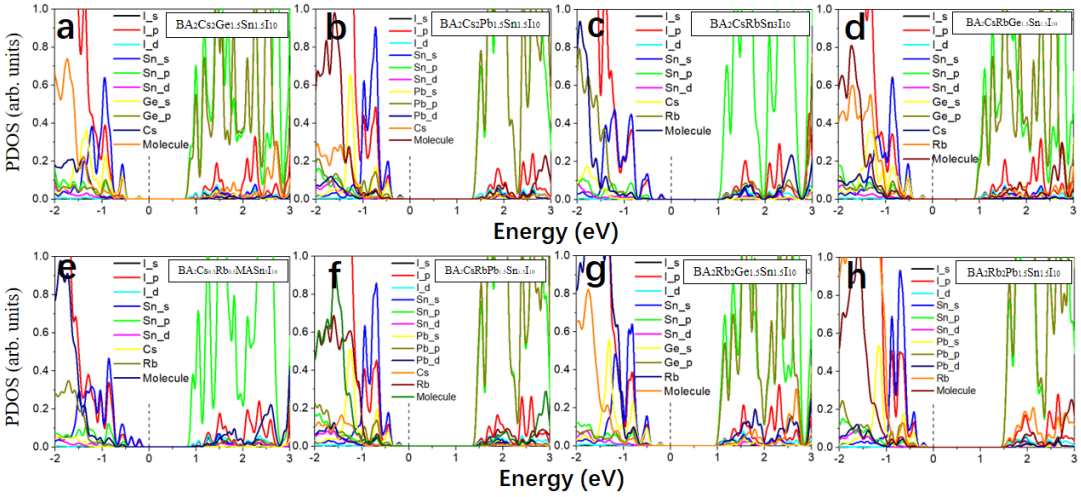

Figure S14. Calculated partial densities of states (PDOSs) of the various atoms (Cs, Rb, Ge, and Pb) mixed 2D SnI-based system: (a) $BA_2Cs_2Ge_{1.5}Sn_{1.5}I_{10}$, (b) $BA_2Cs_2Pb_{1.5}Sn_{1.5}I_{10}$, (c) $BA_2CsRbSn_3I_{10}$, (d) $BA_2CsRbGe_{1.5}Sn_{1.5}I_{10}$, (e) $BA_2Cs_{0.5}Rb_{0.5}MASn_3I_{10}$, (f) $BA_2CsRbPb_{1.5}Sn_{1.5}I_{10}$, (g) $BA_2Rb_2Ge_{1.5}Sn_{1.5}I_{10}$, and (h) $BA_2Rb_2Pb_{1.5}Sn_{1.5}I_{10}$.

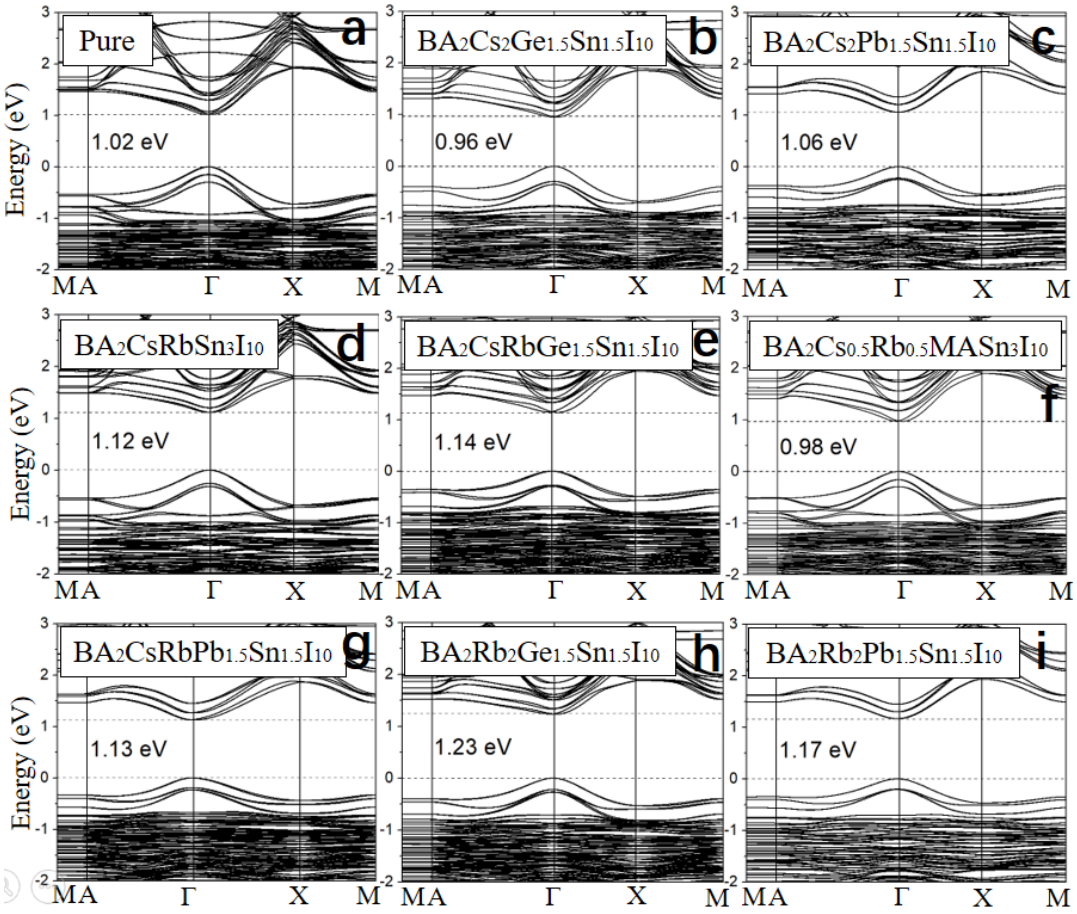

Figure S15. Calculated band structures with SOC effect for the various atoms (Cs, Rb, Ge, and Pb) mixed 2D SnI-based system: (a)Pure, (b) $BA_2Cs_2Ge_{1.5}Sn_{1.5}I_{10}$, (c) $BA_2Cs_2Pb_{1.5}Sn_{1.5}I_{10}$, (d) $BA_2CsRbSn_3I_{10}$, (e) $BA_2CsRbGe_{1.5}Sn_{1.5}I_{10}$, (f) $BA_2Cs_{0.5}Rb_{0.5}MASn_3I_{10}$, (g) $BA_2CsRbPb_{1.5}Sn_{1.5}I_{10}$, (h) $BA_2Rb_2Ge_{1.5}Sn_{1.5}I_{10}$, and (i) $BA_2Rb_2Pb_{1.5}Sn_{1.5}I_{10}$.