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Supporting Information

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Unravelling Alkali-Metal-Assisted Domain Distribution of Quasi-2D Perovskites for Cascade Energy Transfer toward Efficient Blue Light-Emitting Diodes

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Supporting Information

Table S1. Summary of the tri-exponential fitting results for PL lifetime curves of Q-2D $CsPbBr_3$ films with various NaBr concentrations. Time-resolved PL lifetime curves were fitted by the tri-exponential function:

$$I = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + A_3 \exp\left(-\frac{t}{\tau_3}\right) \quad \textbf{(S1)}$$

Where, I is the normalized photoluminescence intensity; A_1, A_2 and A_3 are the fractions of the three decay components and $A_1 + A_2 + A_3 = 1$; τ_1, τ_2 and τ_3 stand for the lifetime constants of a fast

component, a middle component and a slow component, respectively. The average lifetime (τ_{ave}) was calculated with the A_i and τ_i (i= 1, 2, 3) values according to the following equation:

$$\tau_{ave} = \frac{A_1\tau_1 + A_2\tau_2 + A_3\tau_3}{A_1 + A_2 + A_3} \quad \textbf{(S2)}$$

Perovskite Emitter	**Emission [nm]**	**τ_1 [ns]**	***A_1* [%]**	**τ_2 [ns]**	***A_2* [%]**	**τ_3 [ns]**	***A_3* [%]**	**τ_{ave} [ns]**	**χ^2**
0% NaBr	478	2.20	13.21	10.87	38.82	40.51	47.97	34.82	1.07
15% NaBr	485	3.27	30.21	12.01	46.02	55.01	23.77	40.27	1.10
30% NaBr	485	3.64	34.12	12.88	45.40	68.30	20.48	49.11	1.12
45% NaBr	487	4.16	26.82	16.49	46.94	75.97	26.24	57.20	1.15

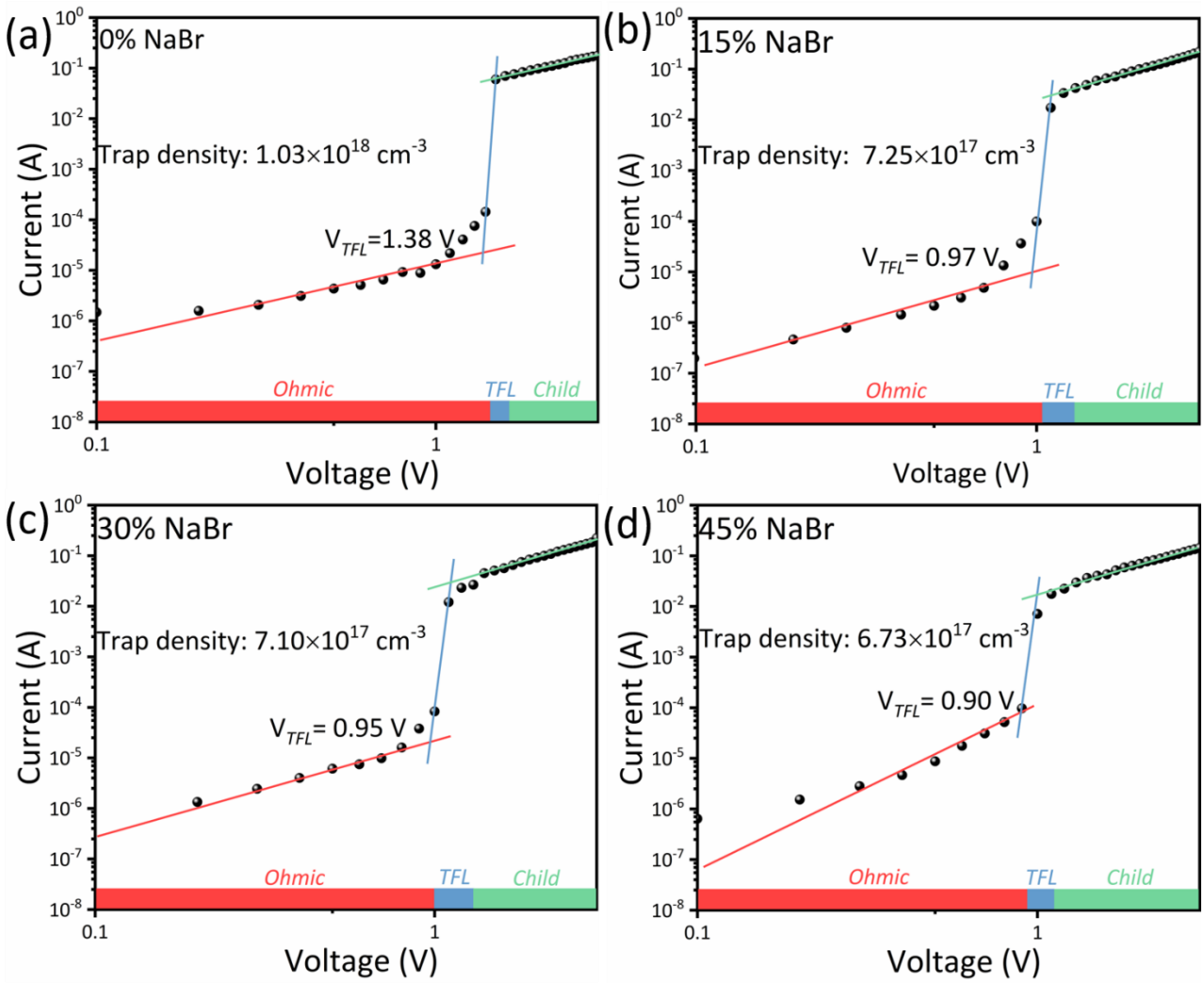

Figure S1. Trap density extraction by dark current-voltage measurement of the hole-only device, with the device structure of ITO/PEDOT/perovskite/MoO_3/Ag. The light-blue lines represent the ohmic regime of each case, and the dark-blue lines indicate the trap-filled limit (TFL) regime with the onset voltage (V_{TFL}). The trap density (N_t) of Q-2D perovskite films with various ratios of NaBr can be extracted by the equation $N_t=\frac{2\varepsilon_0\varepsilon_r VTFL}{qL2}$, where ε_0, q and L represent vacuum permittivity, elementary charge and thickness of the perovskite film, respectively; ε_r is the average relative dielectric constant of Q-2D perovskite films with various ratios of NaBr.

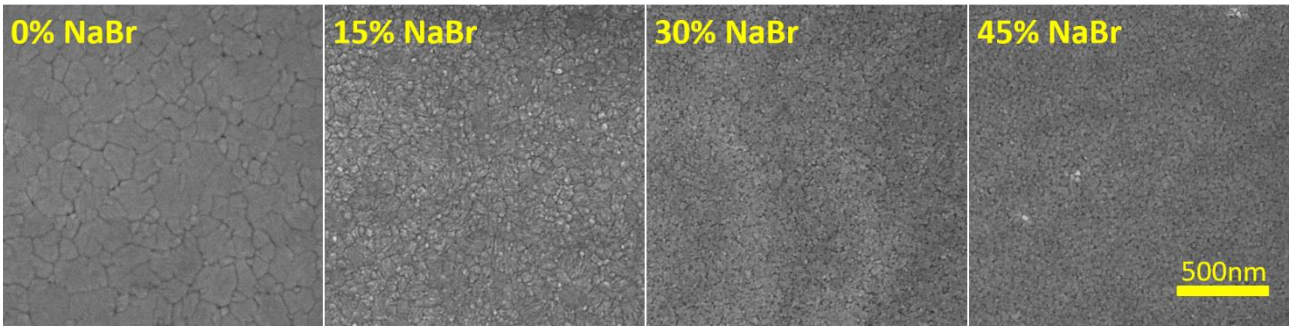

Figure S2. Scanning electron microscope (SEM) images of Q-2D perovskite films with various ratios of NaBr.

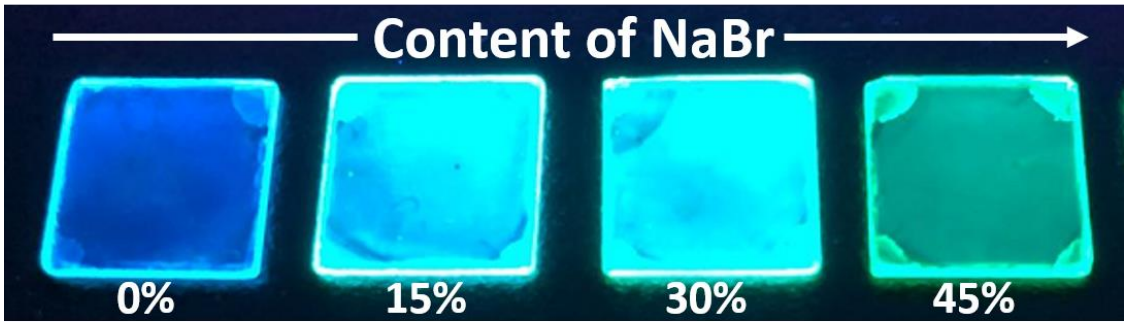

Figure S3. Photoluminescence image of quasi-2D perovskite films with various ratios of NaBr under ultraviolet lamp excitation (365 nm).

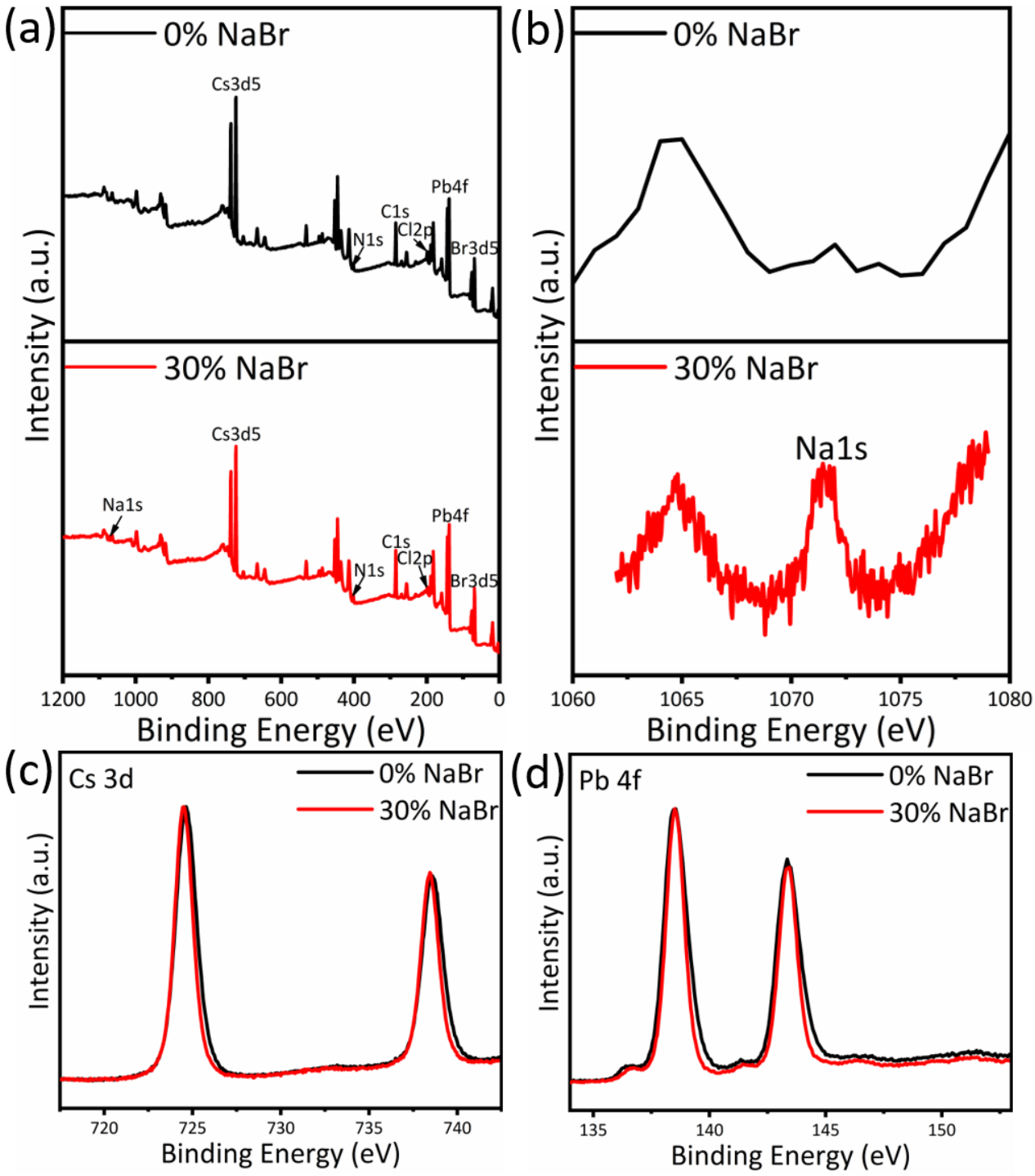

Figure S4. XPS spectra of perovskite films without and with 30% NaBr: (a) Entire spectra, (b) Na 1s spectra, (c) Cs 3d spectra and (d) Pb 4f spectra.

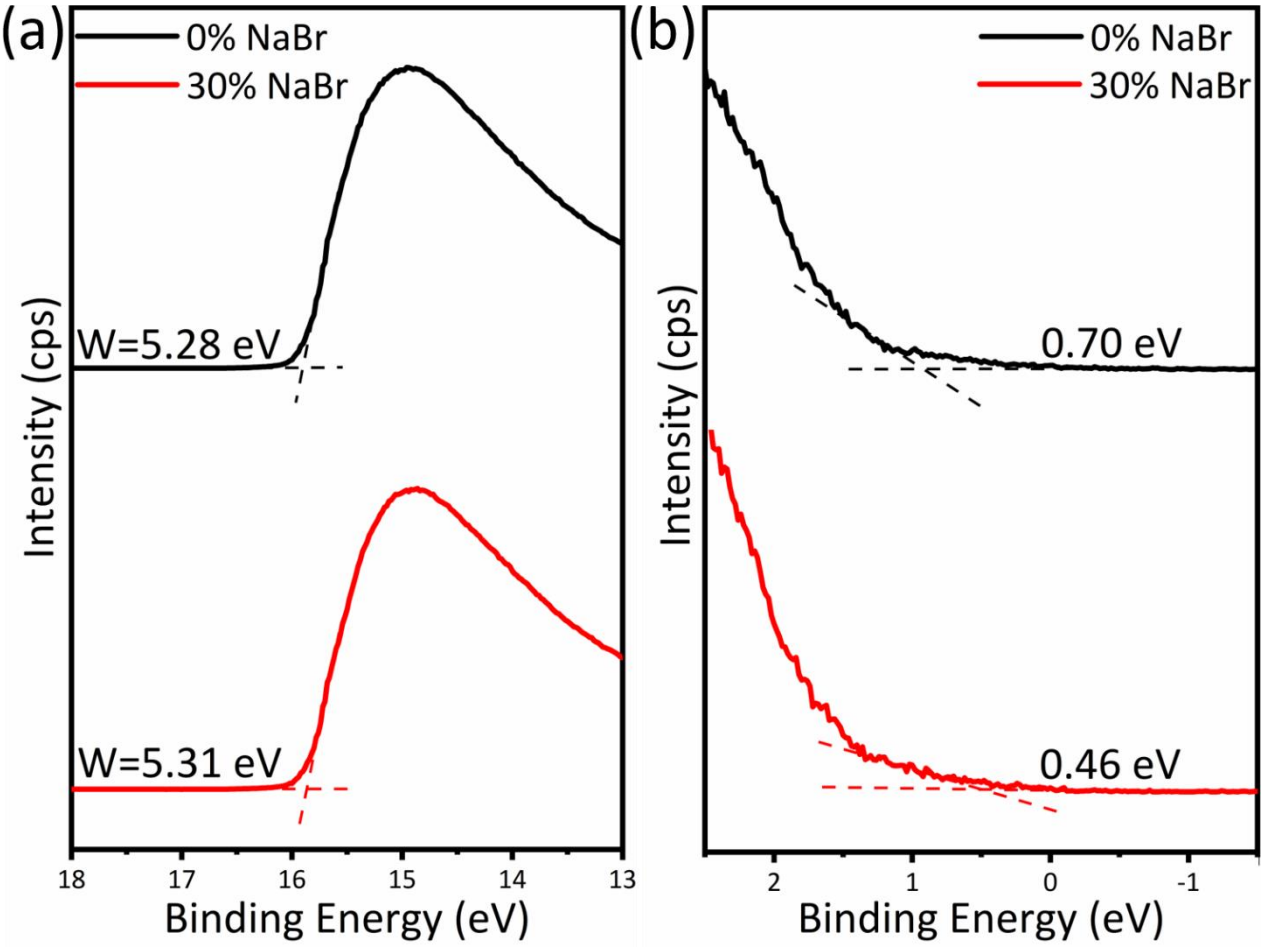

Figure S5. (a) High-binding energy secondary-electron cut-off regions of Q-2D perovskites without and with 30% NaBr. (b) VB-edge regions. The VB is calculated using the equation of VB = 21.2 – (E_{cutoff} – E_{onset}). The calculated VBs were 6.0 and 5.8 for 0% NaBr and 30% NaBr-based perovskite films, respectively.

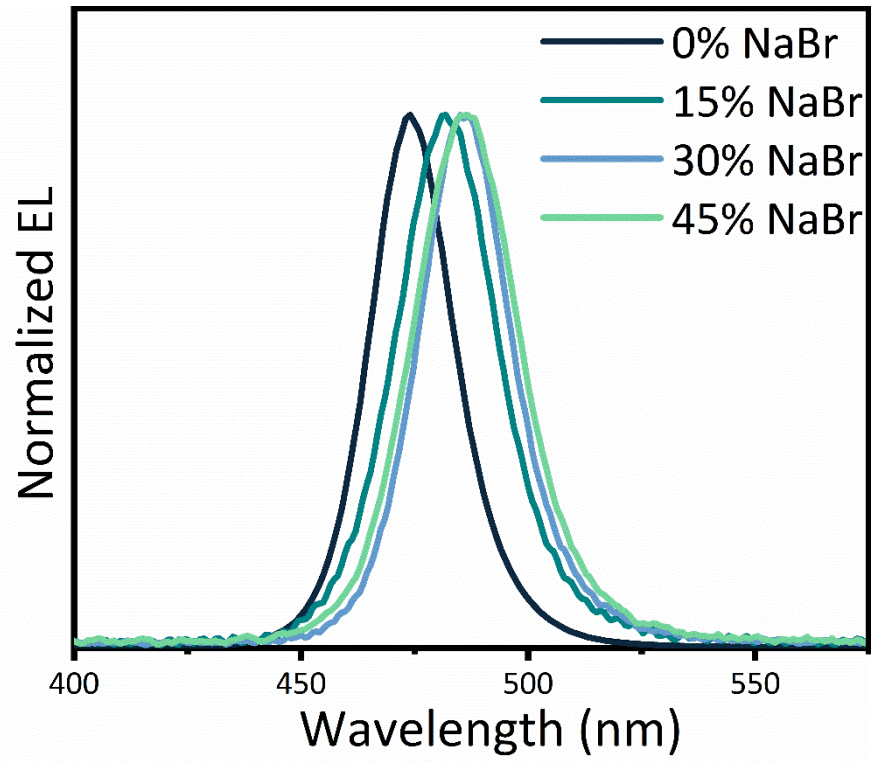

Figure S6. Electroluminescence (EL) spectra of perovskite LEDs.

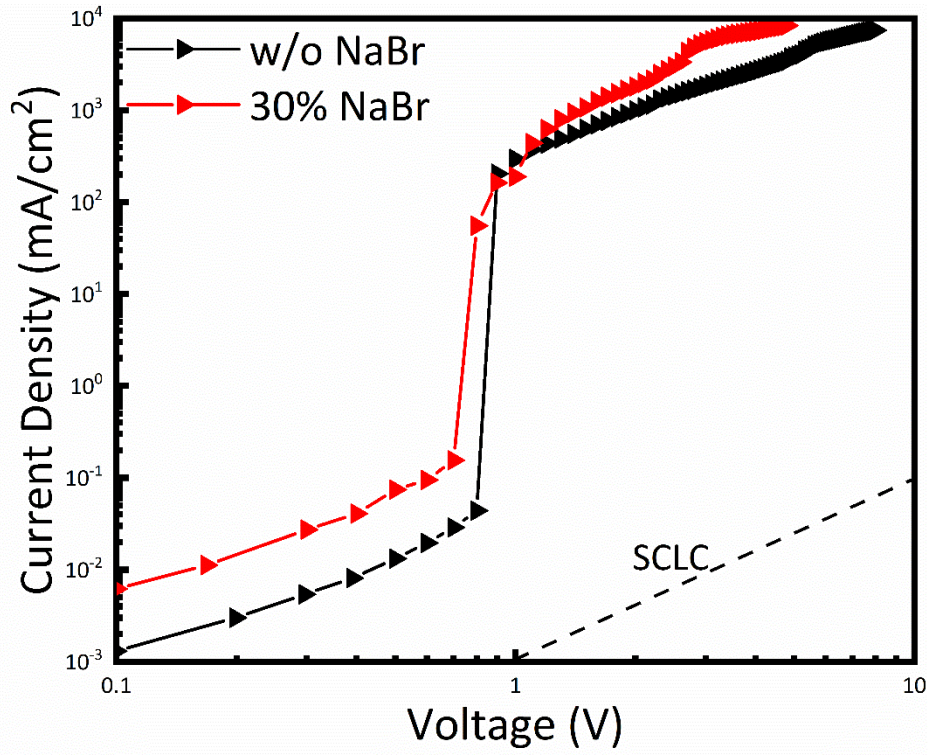

Figure S7. Hole mobility measurement of Q-2D perovskites without and with 30% NaBr by SCLC method, with a device structure of ITO/PEDOT:PSS/perovskite/MoO_3/Ag.

Table S2. Fitting parameters for the kinetics shown in **Figure 5e**. The kinetics are fitted by the multiple-exponential function, $\Delta A(t) = a_1\exp(-t/\tau_1) + a_2\exp(-t/\tau_2) + a_3\exp(-t/\tau_3) - c_1\exp(-t/\tau_{et})$, where a_1, a_2, a_3 and c_1 are the amplitudes; τ_1, τ_2 and τ_3 are the decay time constants and τ_{et} is the electron transfer time constant.

	GSB [nm]	**τ_{et} [ps]**	**τ_1 [ps]**	**τ_2 [ps]**	**τ_3 [ps]**	**χ^2**
n=2	414		5.1	45.6	8190.7	0.99
n=3	440		20.3	387.1	100000	0.97
n≥4	475	30.4	290.5	20715.7		0.99

Table S3. Fitting parameters for the kinetics shown in **Figure 5f**. The kinetics are fitted by the multiple-exponential function, $\Delta A(t) = a_1\exp(-t/\tau_1) + a_2\exp(-t/\tau_2) + a_3\exp(-t/\tau_3) - c_1\exp(-t/\tau_{et})$, where a_1, a_2, a_3 and c_1 are the amplitudes; τ_1, τ_2 and τ_3 are the decay time constants and τ_{et} is the electron transfer time constant.

	GSB [nm]	**τ_{et} [ps]**	**τ_1 [ps]**	**τ_2 [ps]**	**τ_3 [ps]**	**χ^2**
n=2	418		4.8	43.0	8163.3	0.98
n=3	445		2.5	55.5	12707.5	0.99
n≥4	488	22.2	229.4	16319.5		0.99

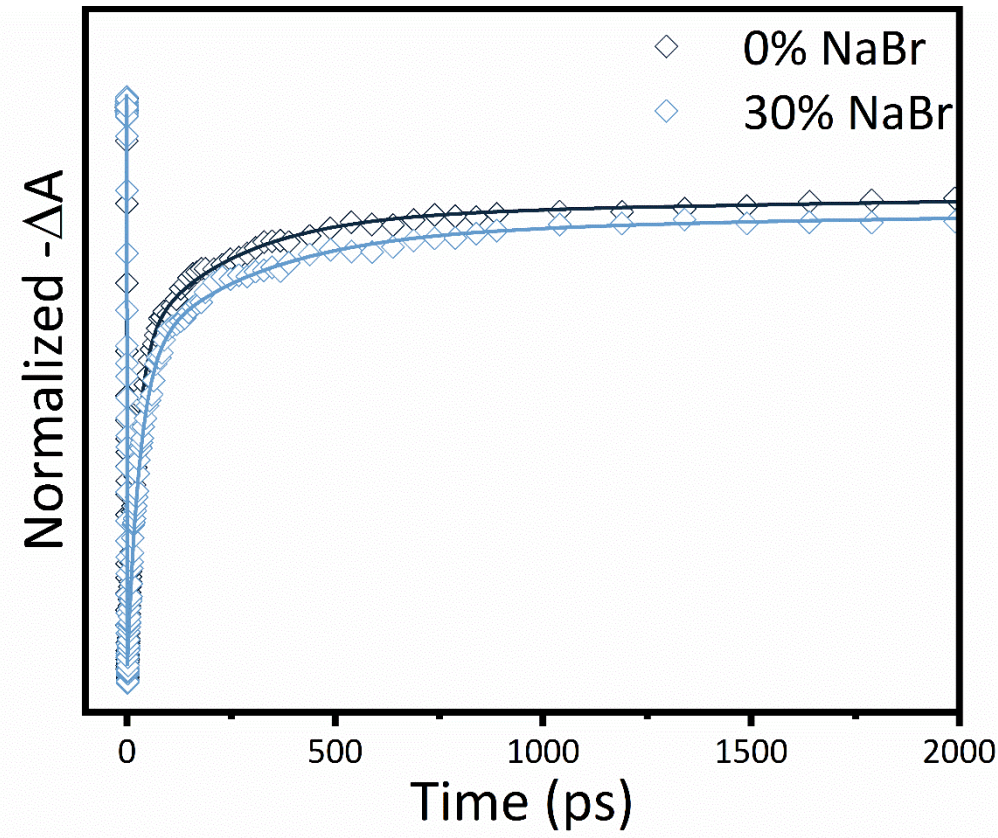

Figure S8. Bleach recovery dynamics for pristine and 30% NaBr-incorporated perovskite films. Solid lines are the fits of the kinetics by exponential function.

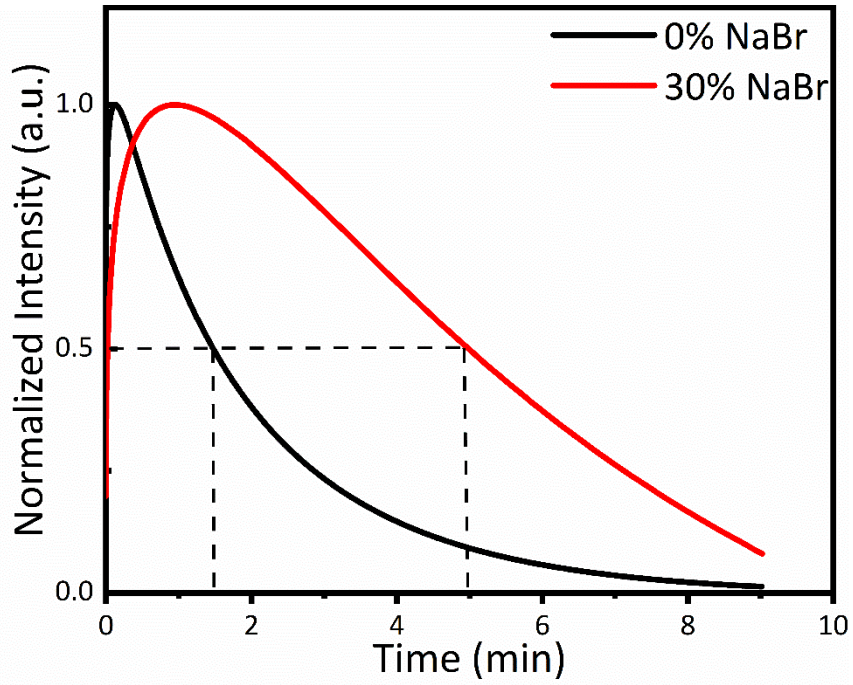

Figure S9. Stability measurement of the PeLEDs with an initial luminance of 100 cd/m^2.