Supporting Information

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Manipulating Color Emission in 2D Hybrid Perovskites by Fine Tuning Halide Segregation: A Transparent Green Emitter

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**Figure S1.** Photoluminescence (PL) spectra of the materials analyzed in this work by halides composition changes and organic cations used. a,b) PL spectra of TMA-based LDP materials for iodide-bromide, iodide-chloride halides mixtures respectively. c,d) PL spectra of TEA-based LDP materials for iodide-bromide, iodide-chloride halides mixtures respectively. The data have been collected upon an incident light excitation of 300 nm or 405 nm with respect to the position of the main absorption peak of the material; iodide-chloride mixtures have been all analyzed through 300 nm light excitation.
Figure S2. XRD patterns of the materials analyzed in this work by halides composition changes and organic cations used. a,b,c) XRD patterns of TMA-based LDP materials for iodide-bromide, iodide-chloride, chlorine-bromide halides mixtures respectively; and (d,e) XRD patterns of TEA-based LDP materials for iodide-bromide and iodide-chloride mixtures. The superposition of the patterns for each series is represented with a zoom on the first diffraction peak, in order to highlight their shift upon halides composition modification: f,g,h)
TMA-based iodide-bromide, iodide-chloride and bromide-chloride mixtures; and i,l) TEA-based iodide-bromide and iodide-chloride halides mixtures.

**Figure S3.** Raman spectra for TEA$_2$PbI$_4$ and TMA$_2$PbI$_4$ LPD materials. Structurally TEA and TMA molecules differ by single carbon atom so that TMA molecule is more rigid than TEA molecule. Therefore, 2D perovskite with TMA molecule, TMA$_2$PbI$_4$ shows Raman bands shift towards lower wavenumber with less band filling, on contrary 2D perovskite with TEA molecules, TEA$_2$PbI$_4$ shifts higher wavenumber with stronger band filling. Rigid organic molecules make the structure more rigid, in this system all the lattice phonon oscillating at higher frequencies, and favors stronger phonon-light interaction, consequently all the Raman active mode appears at lower wavenumbers, compared to the less rigid TEA-system.
**Figure S4.** Hyperspectral photoluminescence (PL), Reflectance (R), and Transmittance (T) maps for a) TMA$_2$PbI$_4$, b) TMA$_2$PbI$_2$Br$_2$, c) TMA$_2$PbI$_2$Br$_2$, d) TMA$_2$PbI$_2$Cl$_3$ perovskite thin films. All the PL maps are taken using 405 nm laser excitation with ≈200 mW cm$^{-2}$ intensity. A calibrated white light source with an intensity of 100 mW cm$^{-2}$ was used for measuring transmittance. PL maps are normalised to the maximum value of the map. The dark shapes in the maps represent gold nanoparticles that we used for marking the samples. All measurements were performed in an ambient atmosphere.
Figure S5. Hyperspectral photoluminescence (PL), Reflectance (R), and Transmittance (T) maps for a) TEA$_2$PbI$_4$, b) TEA$_2$PbI$_2$Br$_2$, c) TEA$_2$PbI$_2$Br$_2$, d) TEA$_2$PbI$_2$Cl$_3$ perovskite thin films. All the PL maps are taken using 405 nm laser excitation with $\approx$200 mW cm$^{-2}$ intensity. A calibrated white light source with an intensity of 100 mWcm$^{-2}$ was used for measuring transmittance. PL maps are normalised to the maximum value of the map. The dark shapes in the maps represent gold nanoparticles that we used for marking the samples. All measurements were performed in an ambient atmosphere.
Figure S6. Photoluminescence spectra spatially integrated over the PL mapped regions of Figures SI4 and SI5, for the 2D perovskite thin film with the cations of: a) TMA and b) TEA.

Figure S7. Energy-dispersive X-ray spectroscopy (EDX) maps of a) chlorine, b) iodine, c) iodine/chlorine (I:Cl) peak intensity ratio, and d) lead for the \( \text{TEA}_2\text{PbICl}_3 \) film.
Figure S8. EDX maps of a) chlorine, b) iodine, c) iodine/chlorine (I:Cl) ratio, and d) lead for the TMA$_2$PbICl$_3$ film.

Figure S9. Top view scanning electron microscopy (SEM) images of the same regions as shown in Figure 5 for a) TEA$_2$PbICl$_3$ and b) TMA$_2$PbICl$_3$. 
Figure S10. Top view SEM images of the same PL mapped regions (dashed line areas) for a) TEA$_2$PbI$_4$, b) TEA$_2$PbI$_2$Br$_2$, c) TEA$_2$PbI$_2$Cl$_2$, d) TEA$_2$PbICl$_3$, e) TMA$_2$PbI$_4$, f) TMA$_2$PbI$_2$Br$_2$, g) TMA$_2$PbI$_2$Cl$_2$, h) TMA$_2$PbICl$_3$. 
Figure S11. Peak PL intensity normalized to the initial intensity value, and b. Peak wavelength of TEA₂PbI₃ and TMA₂PbI₃ perovskite films over time in ambient air conditions under laser illumination (405 nm laser, \( \approx 100 \text{ mW cm}^{-2} \)).
Figure S12. Hyperspectral photoluminescence maps for a) TEA2PbICl3, and b) TMA2PbICl3 perovskite thin films at emission wavelength of 500 nm and 516 nm, respectively. (c,d) The mean photoluminescence spectra (green and purple curves) and 1-T spectra (black curves) spatially integrated from the High PL (solid lines) and Low PL (dotted lines) areas of the PL maps in panels a and b (The insets show the normalized PL spectra from bright, solid, and dark areas, dashed).
Figure S13. SEM–EDX maps of iodine:chlorine (I:Cl) peak intensity ratio overlaid with the regions of relatively high photoluminescence intensity (green areas) at peak emission wavelengths for a) TEA$_2$PbICl$_3$ (500 nm) and b) TMA$_2$PbICl$_3$ (516 nm). The yellow shapes in the maps are gold particles deposited as fiducial markers to guide spatial correlation between PL imaging and elemental mapping. The PL maps are taken using 405 nm laser excitation with ≈200 mW cm$^{-2}$ intensity and the measurements were performed in an ambient atmosphere.

Figure S14. The photoluminescence emission (PLE), PL, and Absorption spectra for (a) TEA$_2$PbICl$_3$, and (b) TMA$_2$PbICl$_3$ compositions. The PLE data was acquired at emission wavelengths of 500 nm for TEA$_2$PbICl$_3$ and 520 nm for TMA$_2$PbICl$_3$. 

Figure S15. Photoluminescence quantum efficiency (PLQE) of the TEA$_2$PbI$_4$, TEA$_2$PbICl$_3$, TMA$_2$PbI$_4$, and TMA$_2$PbICl$_3$ perovskite thin films measured with illumination intensity of 100 mW/cm$^2$ with 405 nm excitation.

| Sample         | x    | y    |
|----------------|------|------|
| TEA$_2$PbI$_4$ | 0,2885 | 0,6743 |
| TMA$_2$PbI$_4$ | 0,2046 | 0,7412 |
| TEA$_2$PbICl$_3$ | 0,1056 | 0,4931 |
| TMA$_2$PbICl$_3$ | 0,1240 | 0,7765 |

Figure S16. CIE color coordinates for the PL emission of the relevant samples and reference ones. In the table are reported the calculated values of the (x, y) coordinates reported in the graph.