

Electronic Supplementary Information for:

Detailed-Balance Analysis of $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{1-x}\text{Br}_x)_3$ Quantum-Cutting Layers for High-Efficiency Photovoltaics under Real-World Conditions

Matthew J. Crane, Daniel M. Kroupa, Daniel R. Gamelin *

Department of Chemistry, University of Washington, Seattle, WA 98195-1700

*Email: gamelin@chem.washington.edu

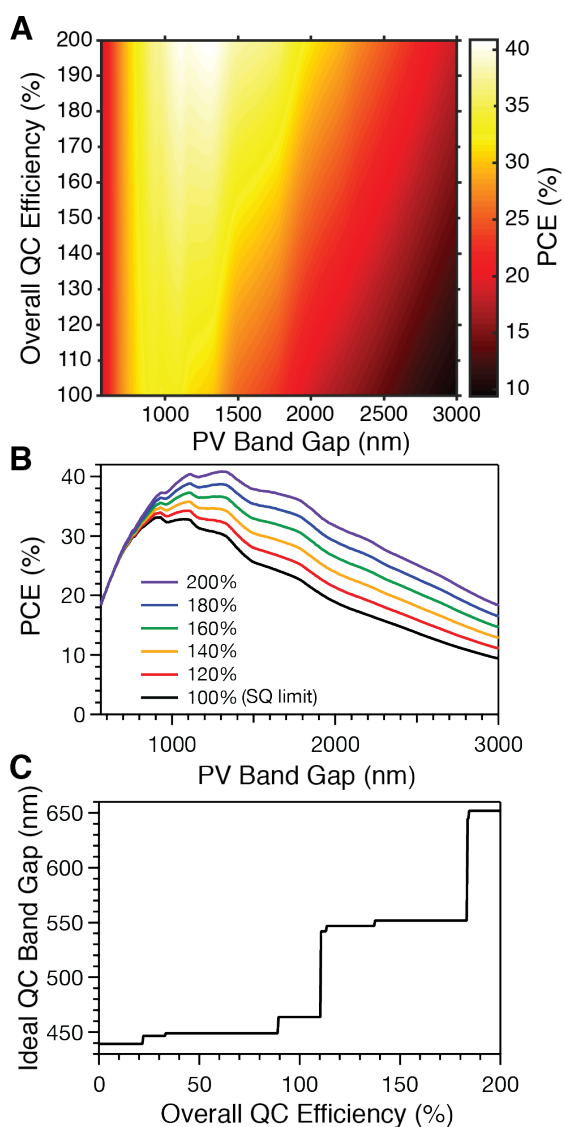


Figure S1. The data from Figure 2 converted to nm for convenience.

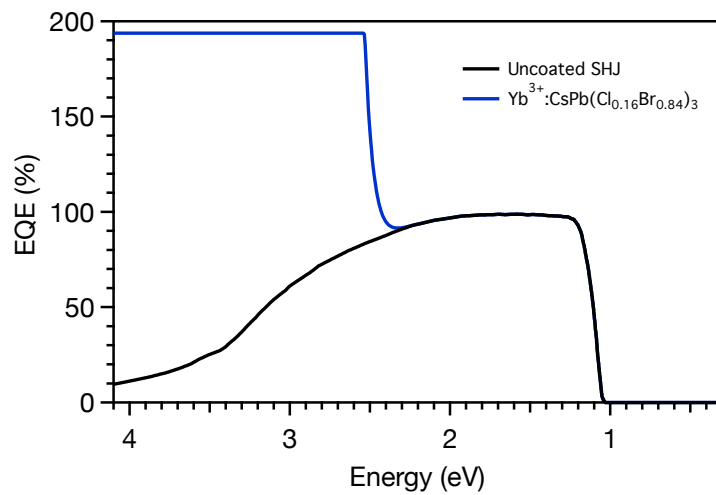


Figure S2. The predicted EQE of a Yb³⁺:CsPb(Cl_{0.16}Br_{0.84})₃/SHJ QC/PV device. Here, η is 200%. This provides a visual representation of the integrand of eq 4.

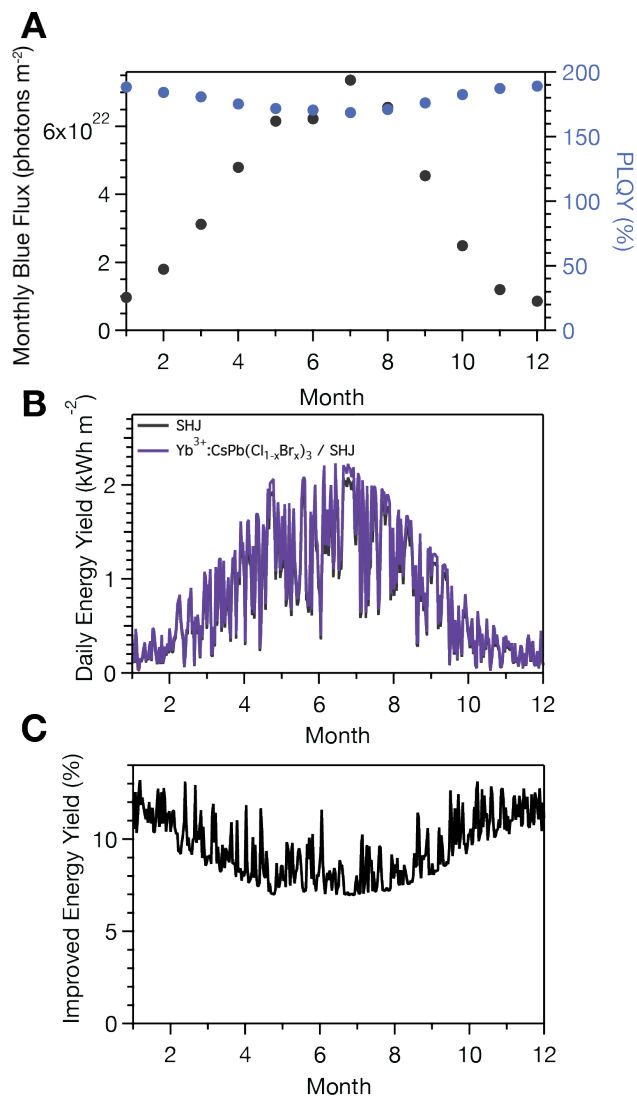


Figure S3. The effect of saturation on Yb³⁺:CsPb(Cl_{0.16}B_{0.84})₃ PLQY and energy yield on Yb³⁺:CsPb(Cl_{0.16}B_{0.84})₃/SHJ device averaged on a day time scale for a TMD in Seattle, WA. **(A)** The total monthly flux blue flux integrated from 4.133 to 2.531 eV (300 to 490 nm), and the average monthly PLQY during hours of power generation. **(B)** A comparison of the daily energy yield produced by a SHJ device and a Yb³⁺:CsPb(Cl_{0.16}B_{0.84})₃/SHJ device and **(C)** the improved daily energy yield.

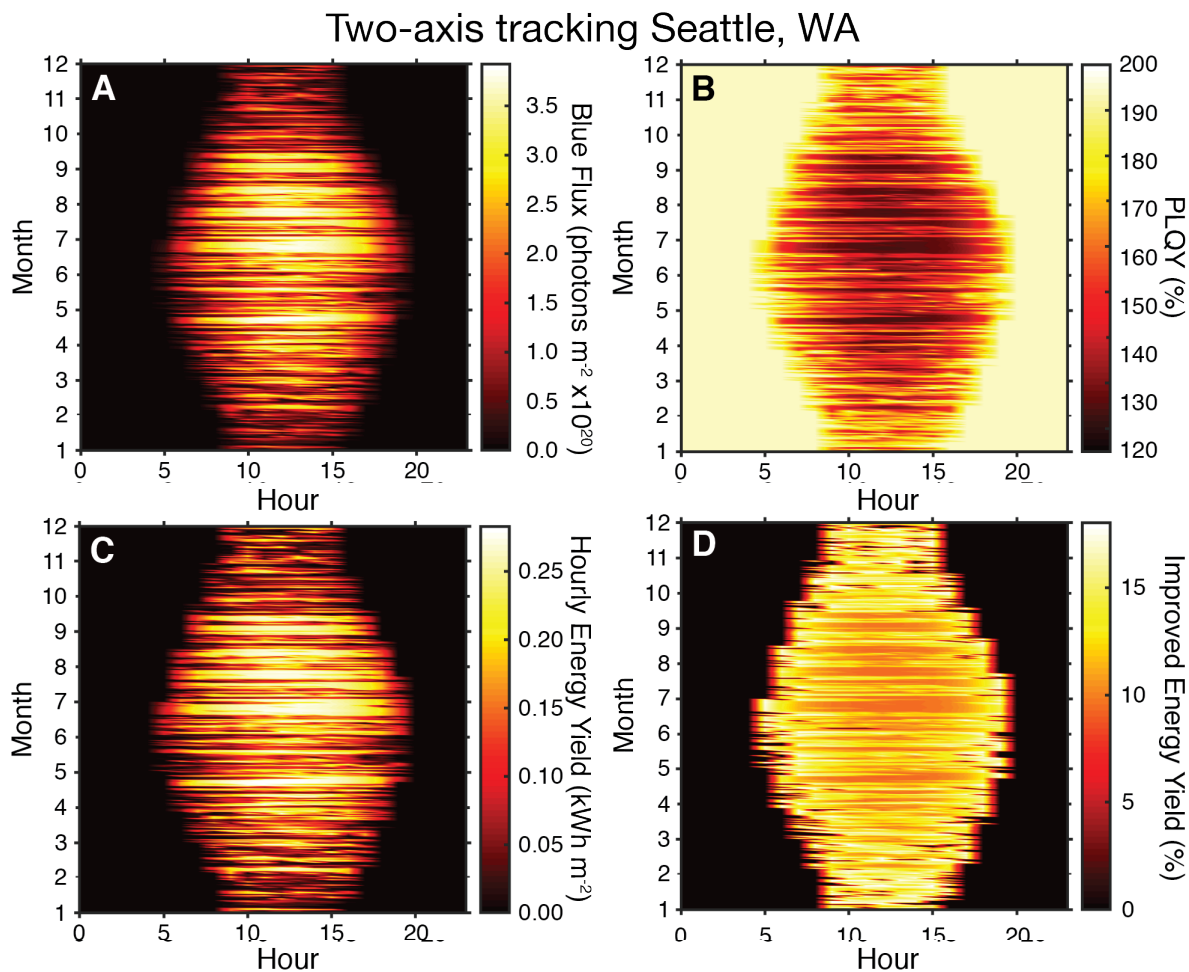


Figure S4. The impact of flux-dependent quantum yields for a $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{0.16}\text{Br}_{0.84})_3/\text{SHJ}$ QC/PV device, represented for a typical meteorological year in Seattle, WA, USA, using two-axis tracking.¹ **(A)** The average hourly, global horizontal irradiance (GHI) photon flux from 300 to 490 nm absorbed by an optimized $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{1-x}\text{Br}_x)_3$ quantum cutter. **(B)** The hourly PLQY of the QC material using the data from panel (A) and experimental PL saturation results from ref. 2. **(C)** The areal hourly energy yield of the QC/PV device, calculated using the model from Figure 4 and assuming 100% optical coupling of the QC PL. **(D)** The corresponding percent increase in areal hourly energy yield relative to a standard SHJ PV without a QC layer, in Seattle with two-axis tracking.

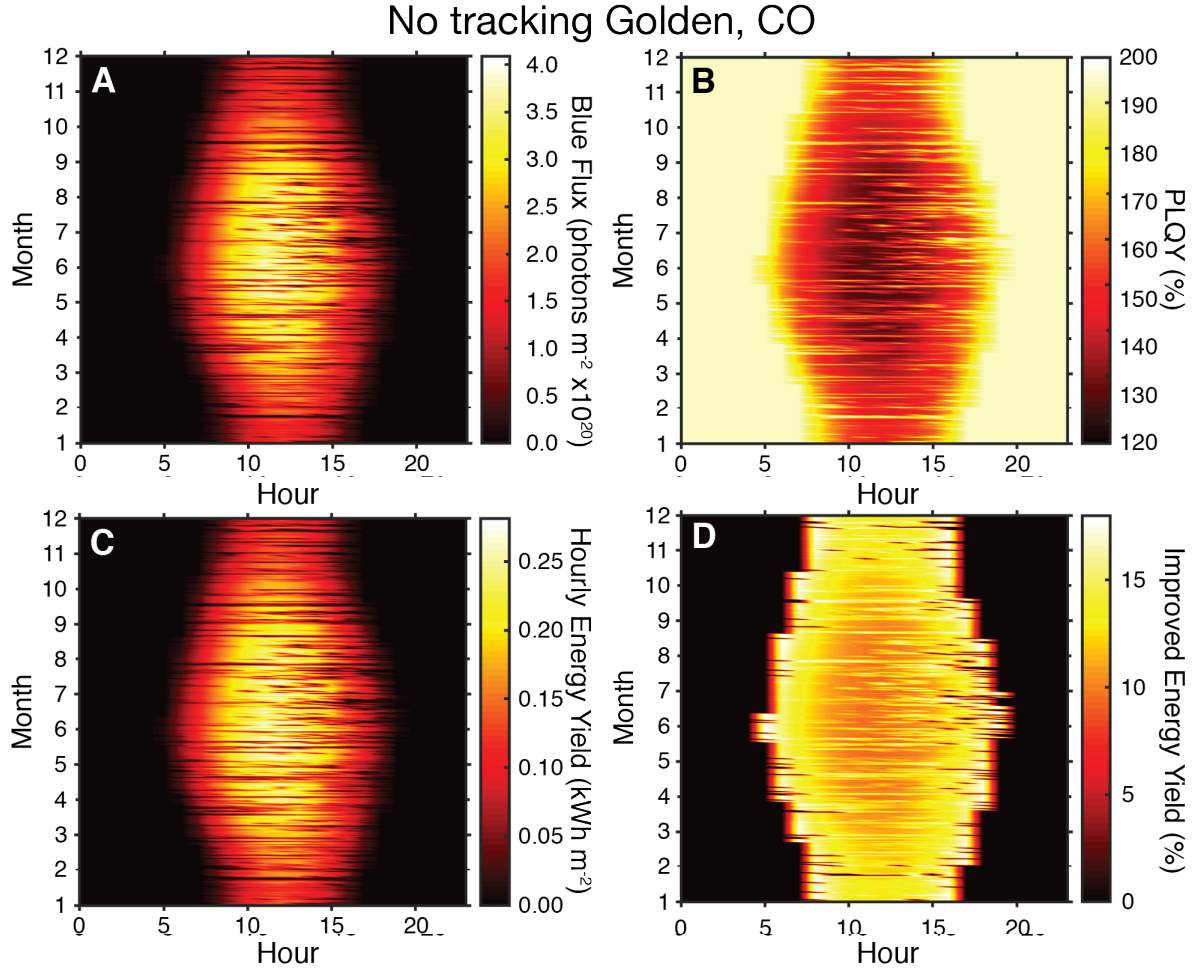


Figure S5. The impact of flux-dependent quantum yields for a $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{0.16}\text{Br}_{0.84})_3/\text{SHJ}$ QC/PV device, represented for a typical meteorological year in Golden, CO, USA, without tracking.¹ **(A)** The average hourly, global horizontal irradiance (GHI) photon flux from 300 to 490 nm absorbed by an optimized $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{1-x}\text{Br}_x)_3$ quantum cutter. **(B)** The hourly PLQY of the QC material using the data from panel (A) and experimental PL saturation results from ref. 2. **(C)** The areal hourly energy yield of the QC/PV device, calculated using the model from Figure 4 of the main text and assuming 100% optical coupling of the QC PL. **(D)** The corresponding percent increase in areal hourly energy yield relative to a standard SHJ PV without a QC layer, in Colorado.

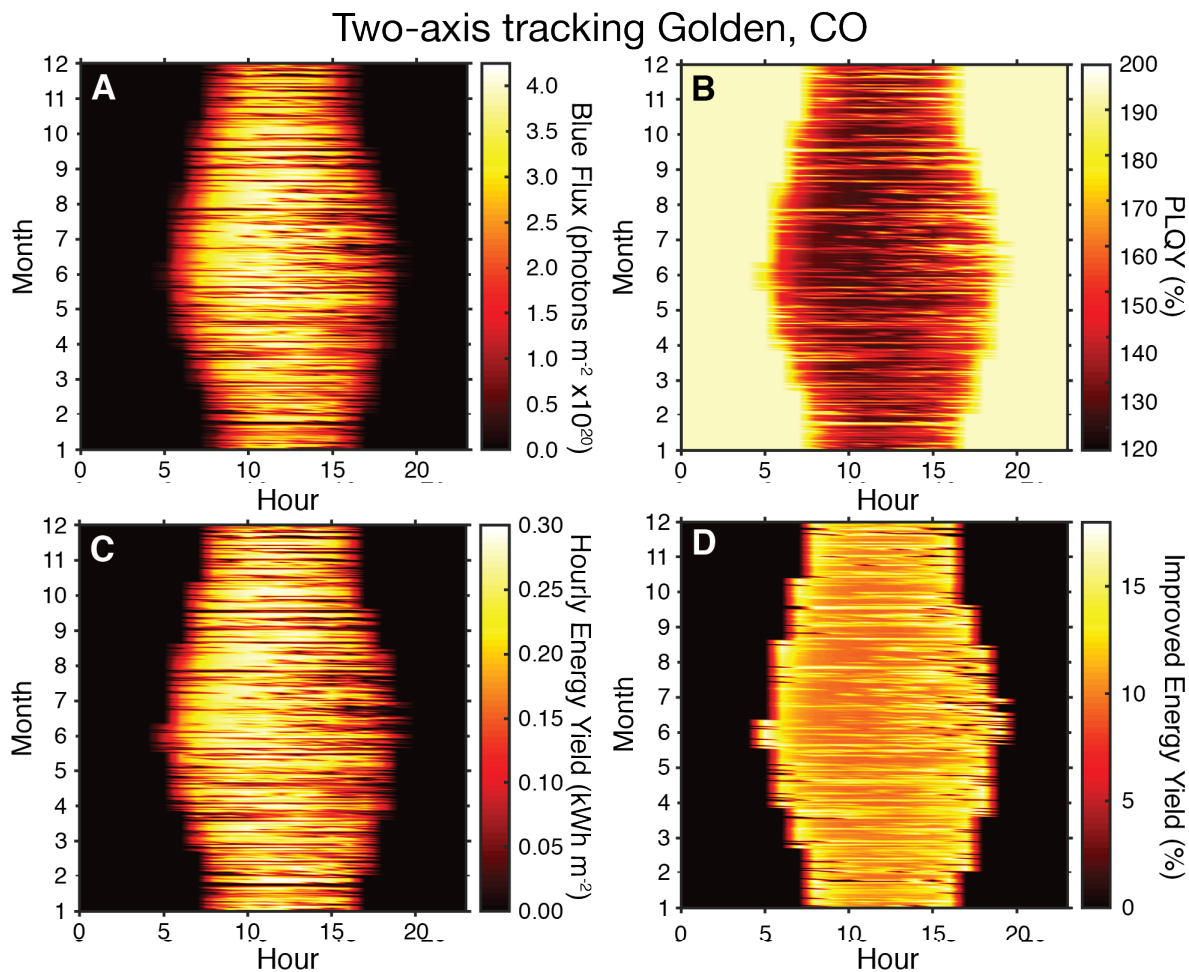


Figure S6. The impact of flux-dependent quantum yields for a $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{0.16}\text{Br}_{0.84})_3/\text{SHJ}$ QC/PV device, represented for a typical meteorological year in Golden, CO, USA, using two-axis tracking.¹ **(A)** The average hourly, global horizontal irradiance (GHI) photon flux from 300 to 490 nm absorbed by an optimized $\text{Yb}^{3+}:\text{CsPb}(\text{Cl}_{1-x}\text{Br}_x)_3$ quantum cutter. **(B)** The hourly PLQY of the QC material using the data from panel (A) and experimental PL saturation results from ref. 2. **(C)** The areal hourly energy yield of the QC/PV device, calculated using the model from Figure 4 of the main text and assuming 100% optical coupling of the QC PL. **(D)** The corresponding percent increase in areal hourly energy yield relative to a standard SHJ PV without a QC layer, in Colorado with two-axis tracking.

Table S1. Collection of symbols, their meanings, and their units used in the calculation.

Symbol	Meaning	Unit
PCE	Device power conversion efficiency	%
P_{device}	Areal power generated by a device	$W\ m^{-2}$
P_{sun}	Areal power from the sun incident on a device	$W\ m^{-2}$
J_{op}	Operating current density	$A\ m^{-2}$
V_{op}	Operating voltage	V
$\Phi_{AM1.5G}$	One-sun solar spectral irradiance	$W\ m^{-2}\ eV^{-1}$
E	Energy, typically of photons	eV
n	Refractive index	-
h	Planck's constant	J s
c	Speed of light	$M\ s^{-1}$
q	Elementary charge of an electron	C
V	Voltage	V
T	Temperature	K
α_{PV}	Photovoltaic absorption probability	-
α_{QC}	Quantum cutter absorption probability	-
$\bar{\alpha}_{PV}$	Photovoltaic absorption probability weighted by quantum cutter photoluminescence linewidth	-
E_{QC}	Energy of quantum cutter emission	eV
ϕ	Photoluminescence quantum yield (PLQY)	-
ξ	Efficiency of optical coupling between the quantum cutter and photovoltaic	-
η	Overall quantum cutting efficiency	-
I_{QC}	Normalized spectra line shape of the quantum cutter's photoluminescence	-

- 1 M. Sengupta, Y. Xie, A. Lopez, A. Habte, G. Maclaurin and J. Shelby, *Renewable and Sustainable Energy Reviews*, 2018, **89**, 51–60.
- 2 C. S. Erickson, M. J. Crane, T. J. Milstein and D. R. Gamelin, *J. Phys. Chem. C*, DOI:10.1021/acs.jpcc.9b01296.