Supporting Information for

Charge Carrier Lifetimes Exceeding 15 Microseconds in Methylammonium Lead Iodide Single Crystals

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KINETIC MODEL:

The following set of coupled differential equations was used to model the concentrations of electrons in the conduction band (n_e), holes in the valence band (n_h) and trapped electrons (N_T) as function of time after photo-excitation of the perovskite single crystal:

$$\frac{dn_e}{dt} = G_C - k_2 n_e(n_h + p_0) - k_T n_e(N_T - n_t) \quad (1)$$

$$\frac{dn_h}{dt} = G_C - k_2 n_e(n_h + p_0) - k_R n_t(n_h + p_0) \quad (2)$$

$$\frac{dn_t}{dt} = k_T n_e(N_T - n_t) - k_R n_t(n_h + p_0) \quad (3)$$

For a full description of this kinetic model, see Hutter *et al.*, *J. Phys. Chem. Lett.* **2015**, *6*, 3082–3090 (Ref. (*29*) in the main text).

Table S1 lists the kinetic parameters that were used to model both the PL and TRMC lifetimes.

k_2 (cm^3s^{-1})	5.5×10^{-9}
k_T (cm^3s^{-1})	9×10^{-6}
k_R (cm^3s^{-1})	2×10^{-9}
N_T (cm^{-3})	1.5×10^{13}
p_0 (cm^{-3})	4×10^{13}
$\Sigma\mu_h$ *(cm^2/Vs)	105
$\Sigma\mu_e$ *(cm^2/Vs)	25

Table S1. Kinetic parameters used to model the TRMC measurements (see **Fig. 3** in the main text). Here, k_2, k_T and k_R are the rate constants for band-to-band electron-hole recombination, trap filling and trap emptying, respectively. N_T denotes the concentration of trap states, p_0 is the background hole concentration at thermal equilibrium. Finally, μ_e and μ_h are the mobilities of electrons (e) and holes (h), obtained from Ref. (*20*) in the main text.

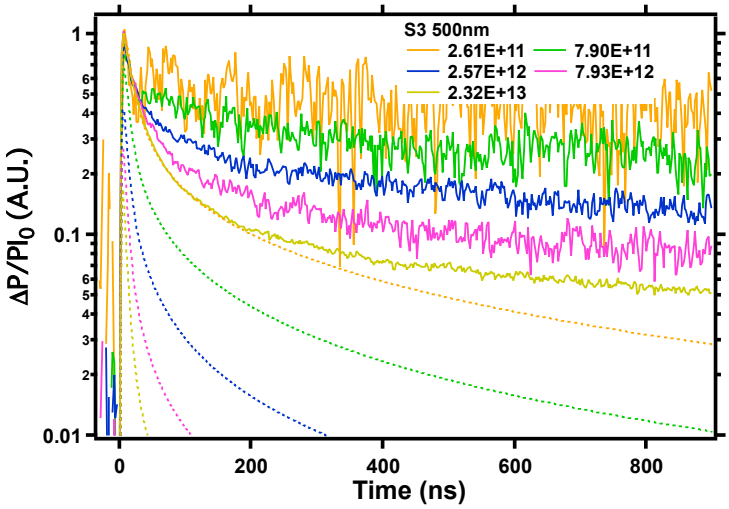

Figure S1. Calculated TRMC traces for excitation at 500 nm using same parameters as excitation at 845 nm. (When we excite our single crystal at 500 nm, it yields an extremely non-uniform charge carrier distribution in the single crystal)

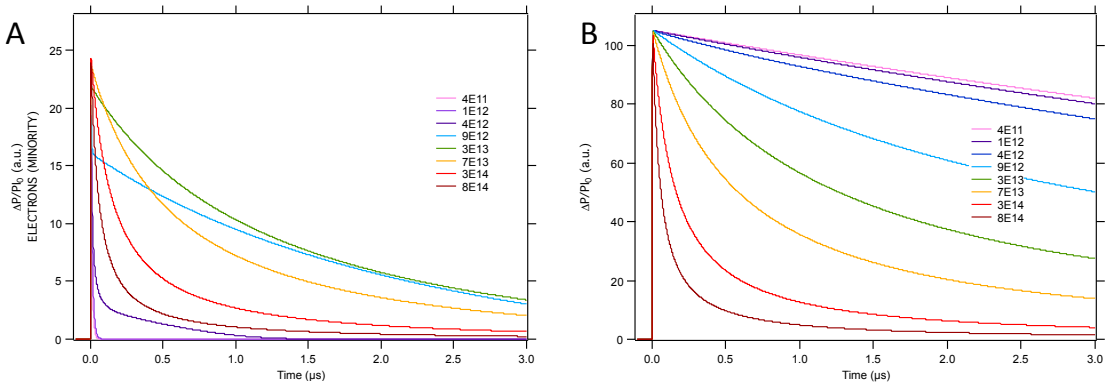

Figure S2. Calculated TRMC traces for charge carriers at different excitation intensities. (**A**) Conduction band electrons and (**B**) valence band holes (photons/cm^2, λ = 845 nm). Note that the experimental TRMC traces are always the sum of A and B.