**Supporting Information** 

## Closing the Green Gap of Photosystem I with Synthetic Fluorophores in Photobiocathodes

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Fig. S1: Light emission intensity of the light source used for photoelectrochemical measurements.



Fig. S2: UV/Vis absorption spectra of the utilized fluorophores in HEPES buffer (50 mM HEPES, pH 7, 50 mM MgSO<sub>4</sub>, 0.03 % DDM).



Fig. S3: Fluorescence measurements of native PSI in comparison with the PSI-ATTO constructs in HEPES buffer (50 mM HEPES, pH 7, 50 mM MgSO<sub>4</sub>, 0.03 % DDM). PSI concentrations were adjusted to 0.25  $\mu$ M. (A) Excitation at 532 nm; (B) Excitation at 590 nm.



Fig. S4: SEM image of the artificial 3D ITO electrode structure. Pore size is ca. 800 nm corresponding to the diameter of the applied polymeric template. (A) Side view at an angle of 30°, (B) top view.



Fig. S5: Comparison of the photo action spectra and the UV/Vis absorbance of the 4 applied constructs. (A) PSI-pure, (B) PSI-ATTO590, (C) PSI-ATTO532, and (D) PSI-ATTO-mix.



Fig. S6: Comparison of the wavelength-dependent performance of the 4 constructs. All photobioelectrodes were measured in 100 mM MES pH 6 and 400 mM KCl at an applied potential of -100 mV vs. Ag/AgCl in the absence of a mediator.



Fig. S7: Photocurrent output for measurements with the dyes on 3D ITO without PSI (n=3). Incubation for 3 min of ATTO 532 (48  $\mu$ M), ATTO 590 (44  $\mu$ M) or ATTO-mix (44  $\mu$ M ATTO 532 & 32  $\mu$ M ATTO 590). Electrodes were measured in 100 mM MES pH 6 and 400 mM KCl at an applied potential of -100 mV vs. Ag/AgCl at 100 mW/cm<sup>2</sup> illumination intensity. For better visibility, the curves were shifted by +1  $\mu$ A/cm<sup>2</sup> for ATTO 532 and -1  $\mu$ A/cm<sup>2</sup> for ATTO-mix.



Fig. S8: Photocurrent output including standard deviation (n=3) for the measurements shown in Figure 6B. Photobiocathodes were prepared with PSI-ATTO532 and measured in 100 mM MES pH 6 and 400 mM KCl at an applied potential of -100 mV vs. Ag/AgCl at 100 mW/cm<sup>2</sup> illumination intensity.