Electronic Supplementary Information for:

Circular management of perovskite solar cells by green solvents: from recycling and reuse of critical components to life cycle assessment

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Supplementary figures

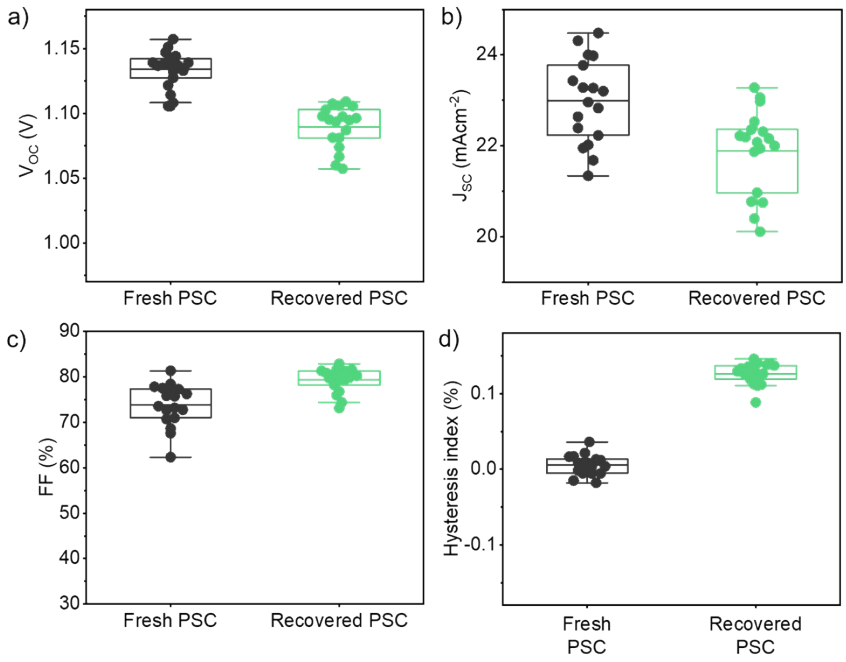

Fig. S1

a) Open-circuit voltage (V_{OC}), b) short-circuit current density (J_{SC}), c) fill factor (FF) and d) hysteresis index box charts for perovskite solar cells (PSCs) fabricated with fresh components and recovered indium tin oxide (ITO)/ tin oxide (SnO_2), lead iodide (PbI_2) and $N^2,N^2,N^{2'},N^{2'},N^7,N^7,N^{7'},N^{7'}$-octakis(4-methoxyphenyl)−9,9'-spirobi[9H-fluorene]−2,2',7,7'-tetramine (Spiro-OMeTAD). Sample size is 18. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value.

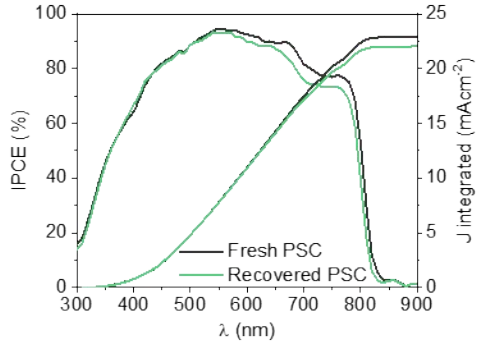

Fig. S2

Incident photon-to-current efficiency (IPCE) and integrated current density curves of PSC fabricated with fresh and recovered ITO/SnO_2, PbI_2 and Spiro-OMeTAD.

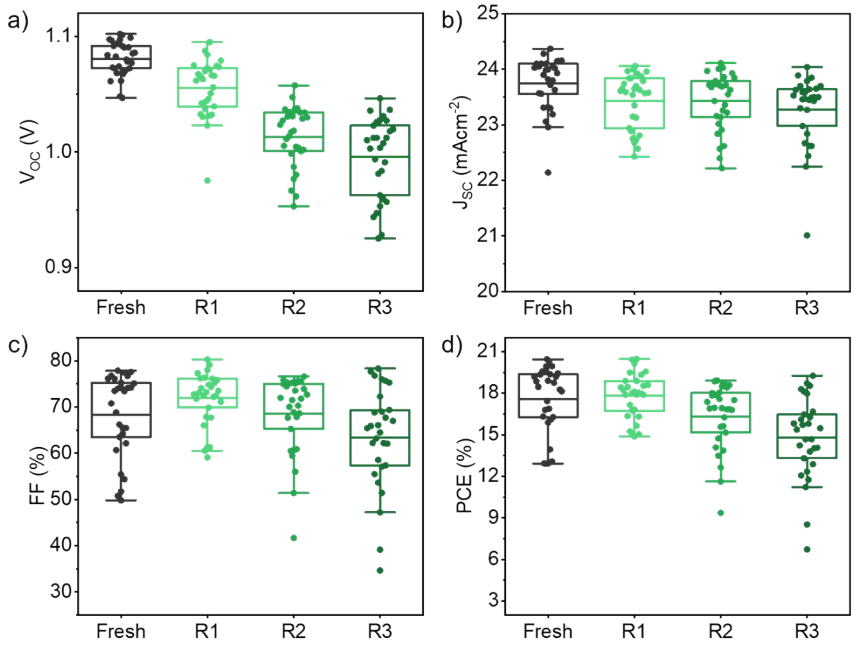

Fig. S3

a) V_{OC}, b) J_{SC}, c) FF and d) PCE box charts for PSCs fabricated with fresh components and recovered ITO/SnO_2, PbI_2 and Spiro-OMeTAD, subjected to multiple recovery iteration processes. Sample size is 30. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value.

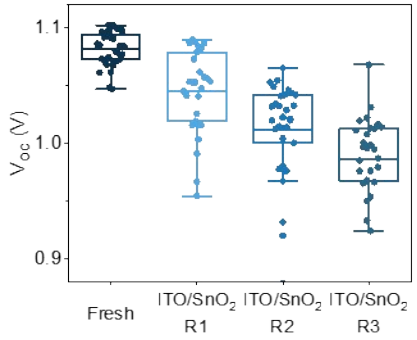

Fig. S4

V_{OC} box charts for PSCs fabricated with fresh and first, second and third-time iterated restoration of ITO/SnO_2 substrates. Sample size is 30. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value.

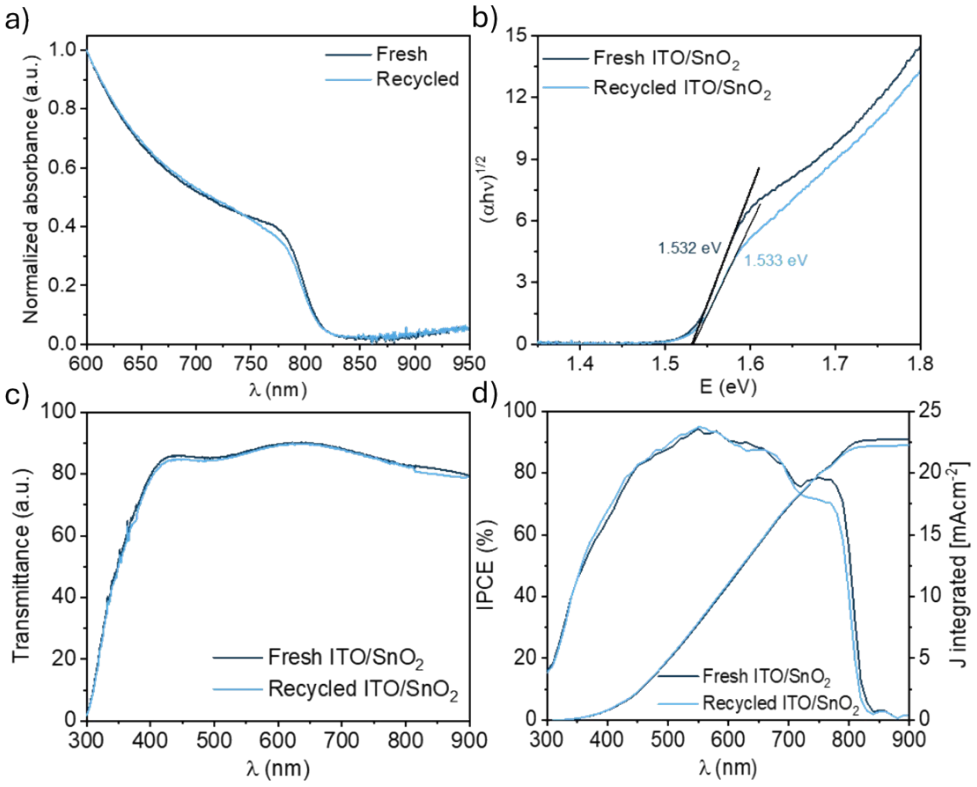

Fig. S5

a) Normalized absorption spectra of Fresh and recycled samples, b) Tauc Plot of Fresh and recycled samples, c) Transmittance spectra of Fresh and recycled samples and d) IPCE spectra of Fresh and recycled samples

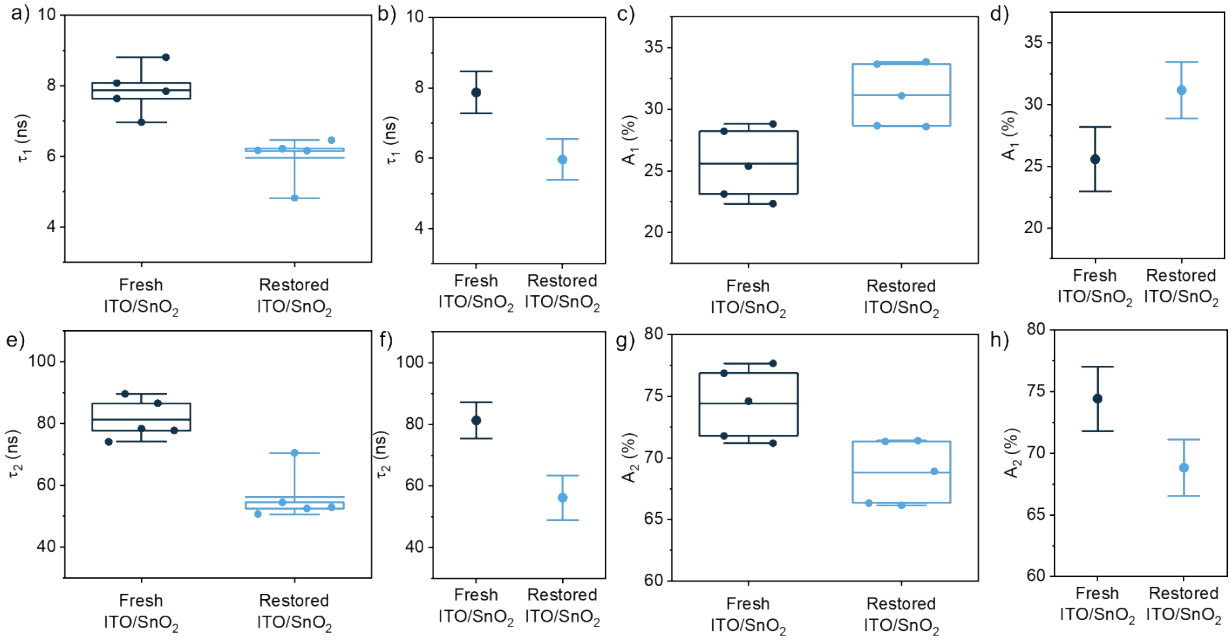

Fig. S6

a) τ_1, c) A_1 e) τ_2, and g) A_2 box charts for PSCs fabricated with fresh and restored ITO/SnO_2. Sample size is 5. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value. Average values with error bars of b) τ_1, d) A_1 f) τ_2, and h) A_2.

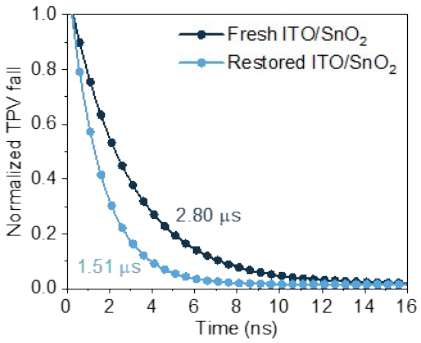

Fig. S7

Transient photovoltage (TPV) decay of PSCs fabricated with fresh and restored ITO/SnO_2 substrates.

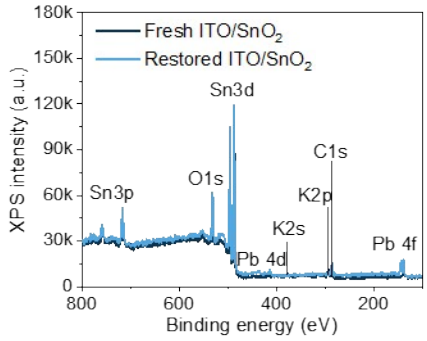

Fig. S8

X-ray photoemission spectroscopy (XPS) of fresh and restored ITO/SnO_2 substrates.

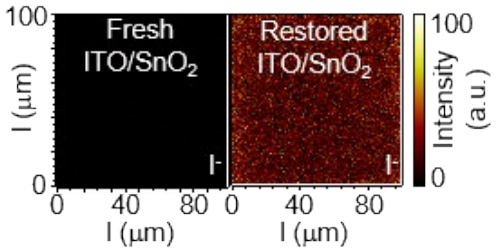

Fig. S9

Time-of-flight secondary ion mass spectroscopy (ToF-SIMS) surface maps of fresh and restored ITO/SnO_2 substrates for I^- ions.

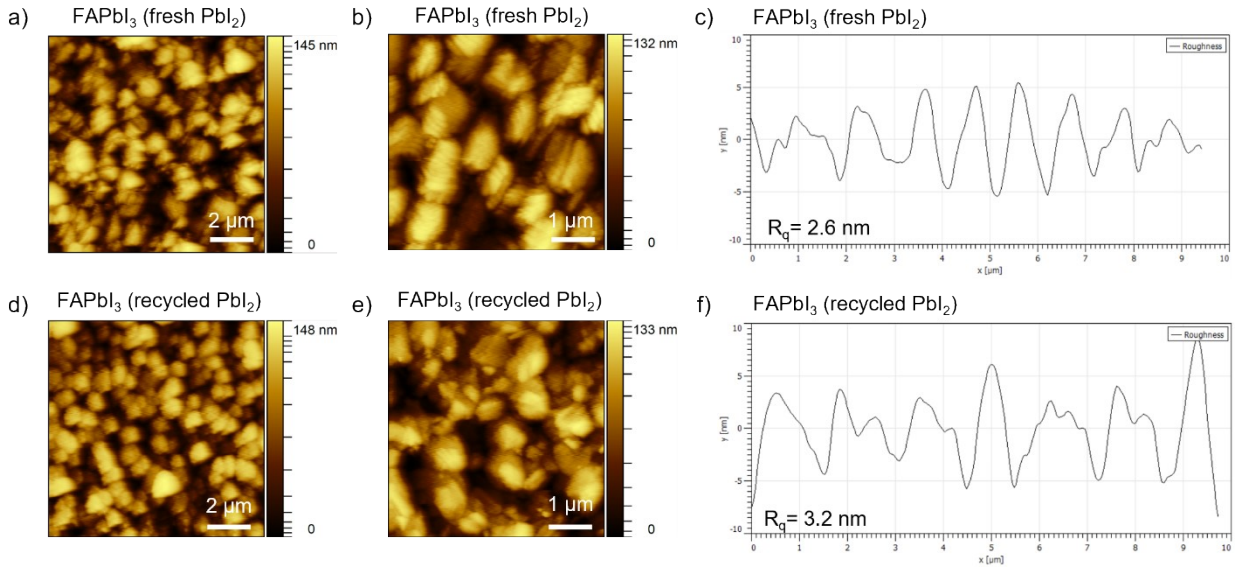

Fig. S10

a) 10µm x 10µm and b) 5µm x 5µm AFM images and c) root mean square roughness (R_q) analysis, averaged on the entire 10µm x 10µm investigated surface, of formamidinium lead iodide ($FAPbI_3$) fabricated with fresh PbI_2. d) 10µm x 10µm and e) 5µm x 5µm AFM images and f) root mean square roughness (R_q) analysis, averaged on the entire 10µm x 10µm investigated surface, of $FAPbI_3$ fabricated with recycled PbI_2.

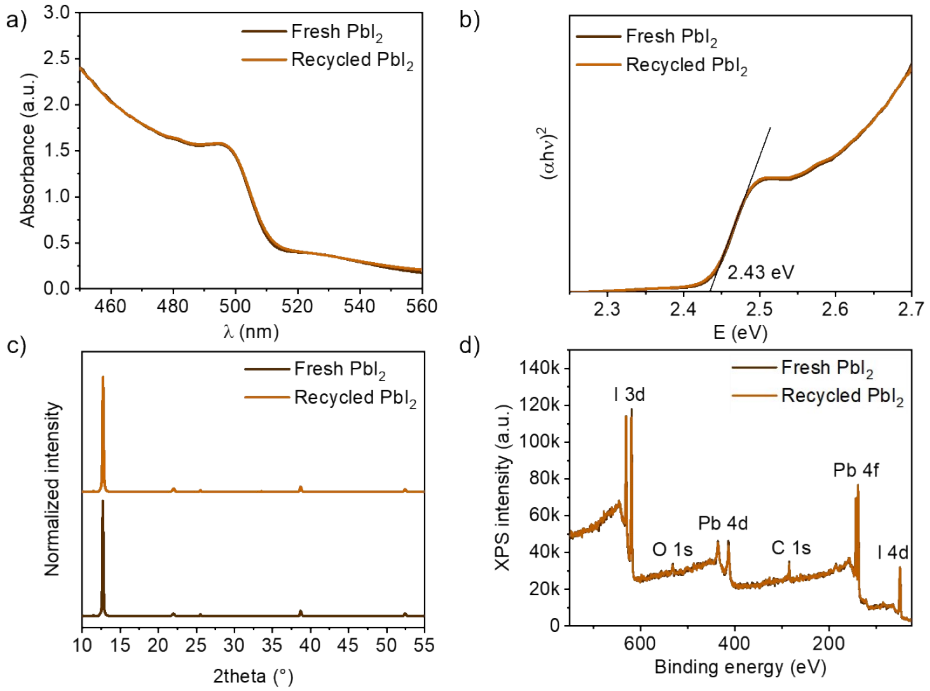

Fig. S11

a) UV-vis absorbance spectra, b) Tauc plot analysis, c) XRD patterns and d) XPS measurement of fresh and recycled PbI_2.

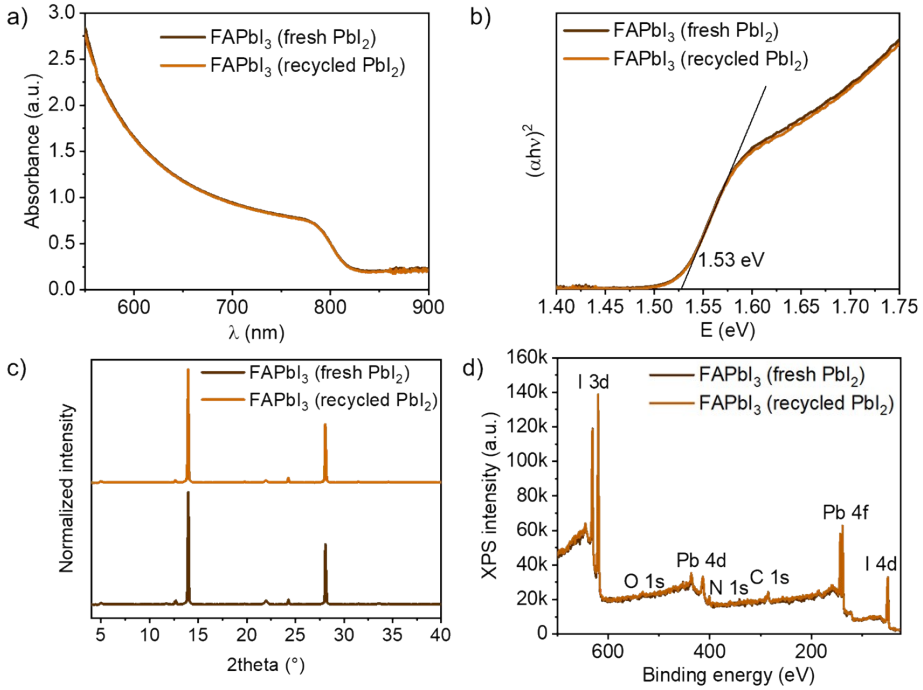

Fig. S12

a) UV-vis absorbance spectra, b) Tauc plot analysis, c) XRD patterns and d) XPS measurement of $FAPbI_3$ fabricated with fresh and recycled PbI_2.

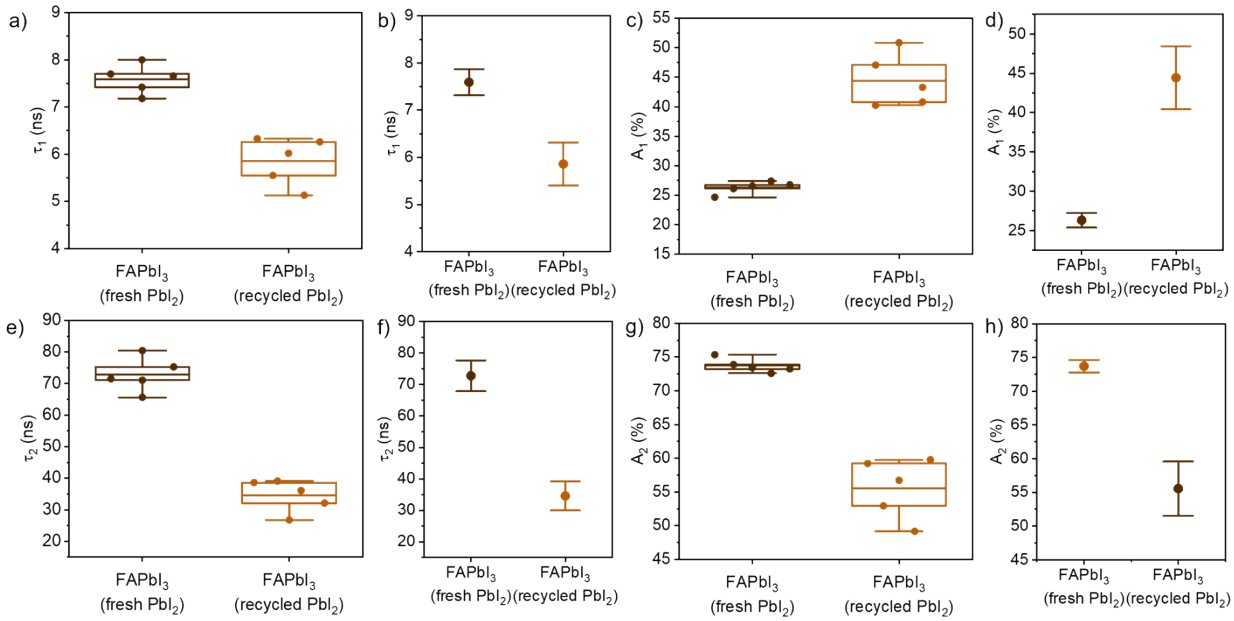

Fig. S13

a) τ_1, c) A_1 e) τ_2, and g) A_2 box charts for PSCs fabricated with fresh and recycled PbI_2. Sample size is 5. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value. Average values with error bars of b) τ_1, d) A_1 f) τ_2, and h) A_2.

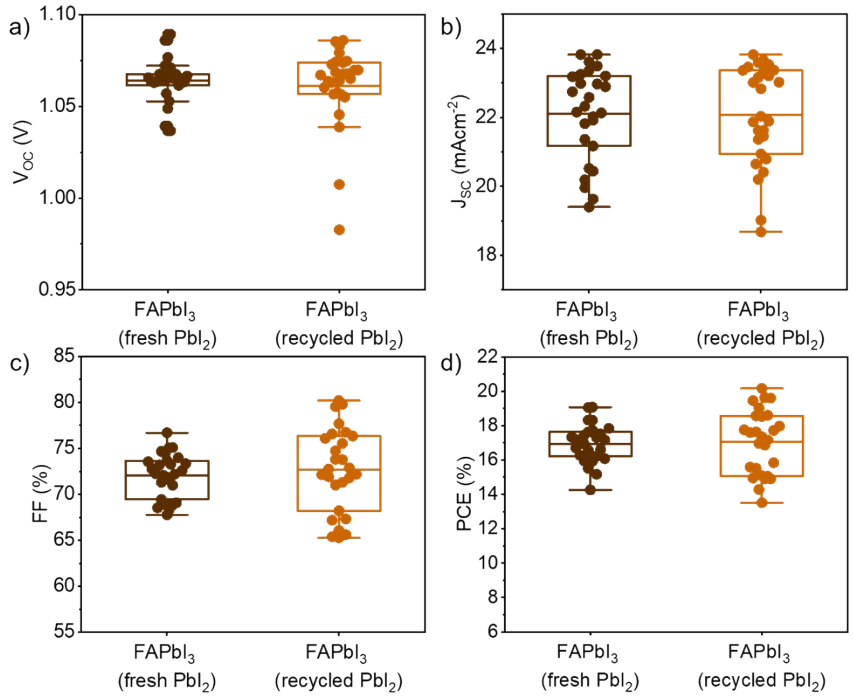

Fig. S14

a) V_{OC}, b) J_{SC}, c) FF and d) PCE box charts for perovskite solar cells (PSCs) fabricated with fresh components and recycled PbI_2. Sample size is 27. Whiskers limit the 1.5 interquartile range, the box identifies the 25th and 75th percentile and the horizontal line represents the average value.

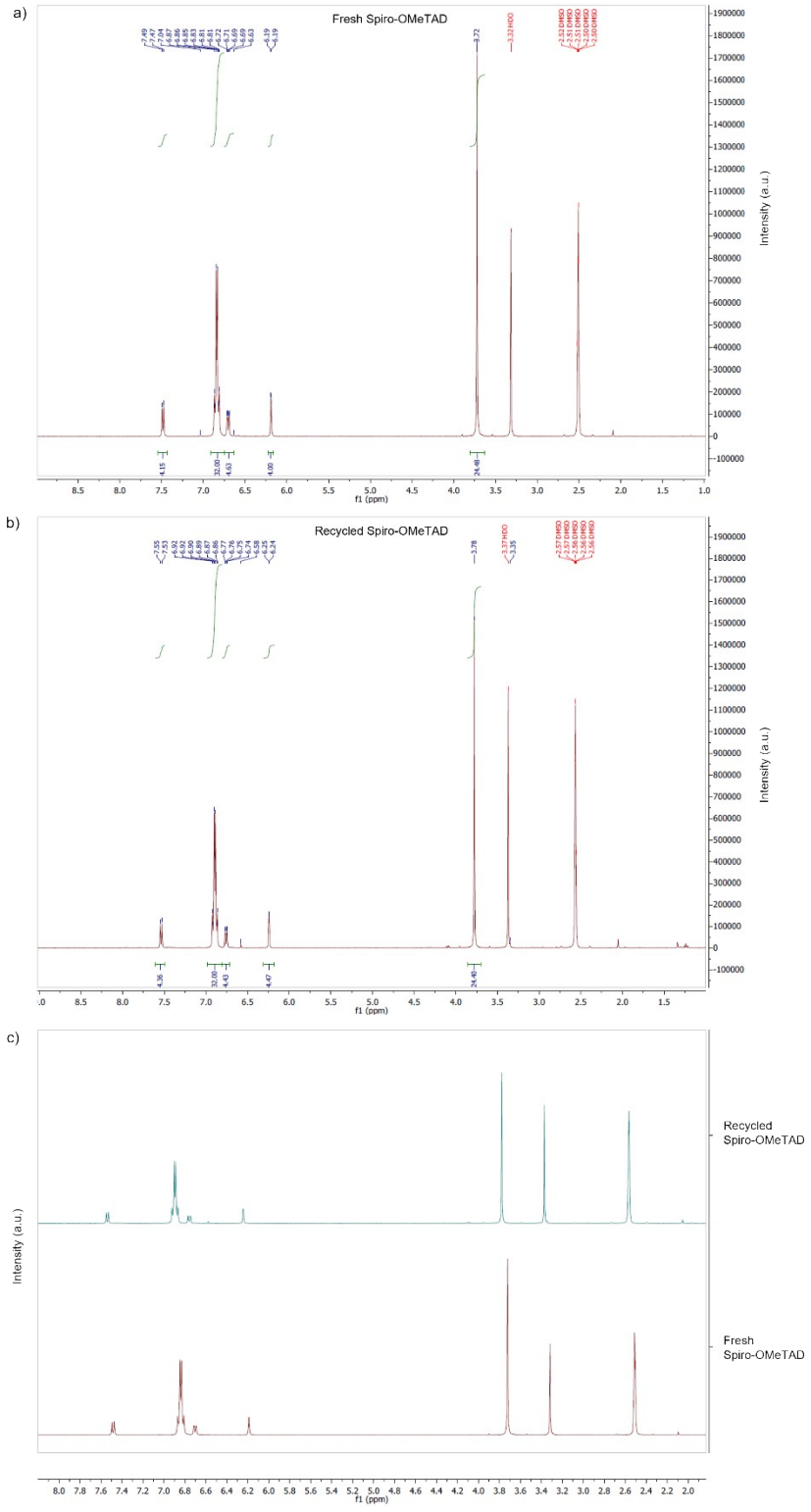

Fig. S15

Proton nuclear magnetic resonance (^{1}H-NMR) spectra of a) fresh and b) recycled Spiro-OMeTAD and c) comparison of the two spectra.

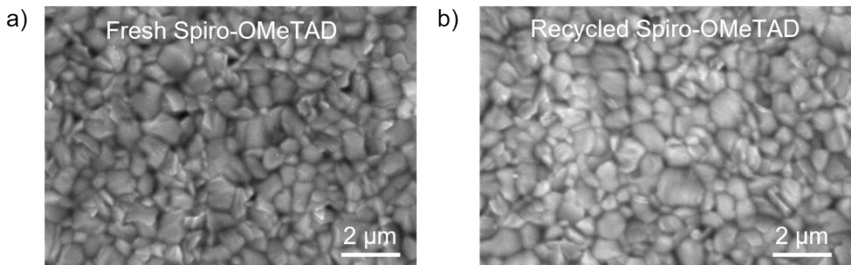

Fig. S16

SEM images of a) fresh and b) recycled Spiro-OMeTAD deposited onto fresh $FAPbI_3$.

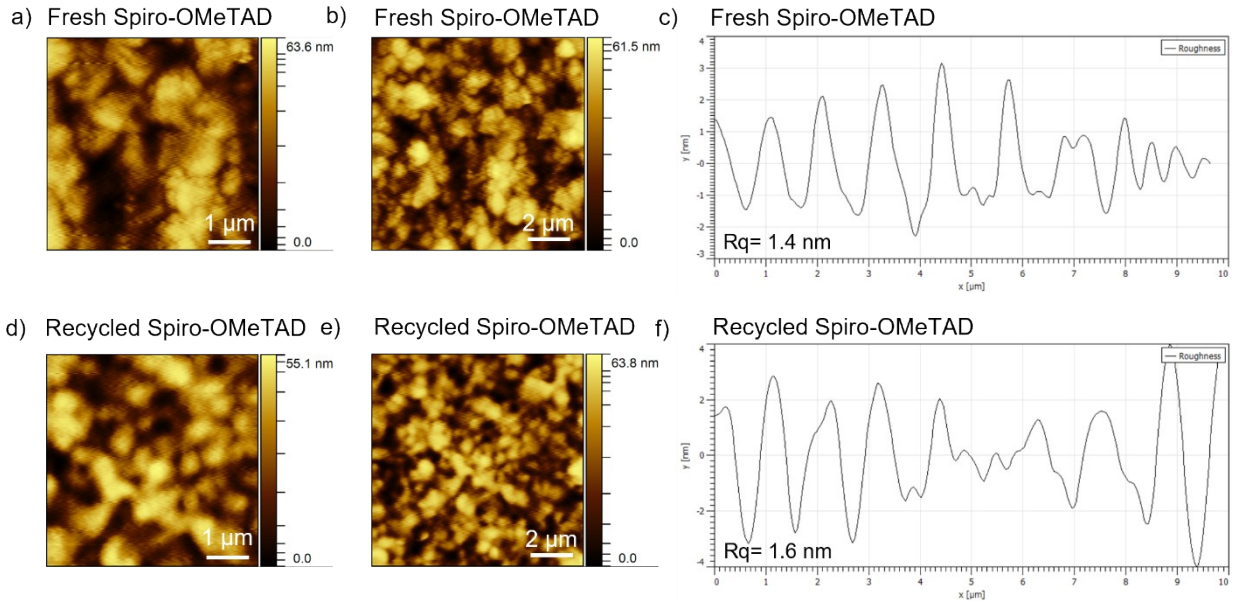

Fig. S17

a) 5µm x 5µm and b) 10 x 10µm AFM images and c) root mean square roughness (R_q) analysis, averaged on the entire 10µm x 10µm investigated surface, of fresh Spiro-OMeTAD deposited onto fresh $FAPbI_3$. d) 5µm x 5µm and e) 10 x 10µm AFM images and f) root mean square roughness (R_q) analysis, averaged on the entire 10µm x 10µm investigated surface, of recycled Spiro-OMeTAD deposited onto fresh $FAPbI_3$.

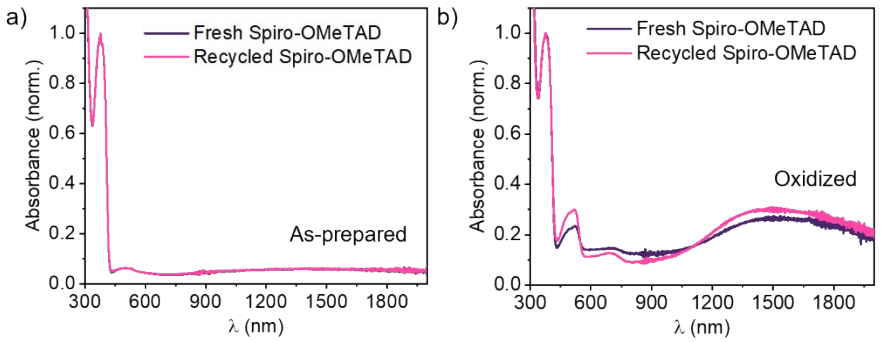

Fig. S18

a) UV-vis absorbance spectra of as-prepared fresh and recycled Spiro-OMeTAD, deposited on glass. b) UV-vis absorbance spectra of fresh and recycled Spiro-OMeTAD subjected to oxidizing atmosphere for 6 days.

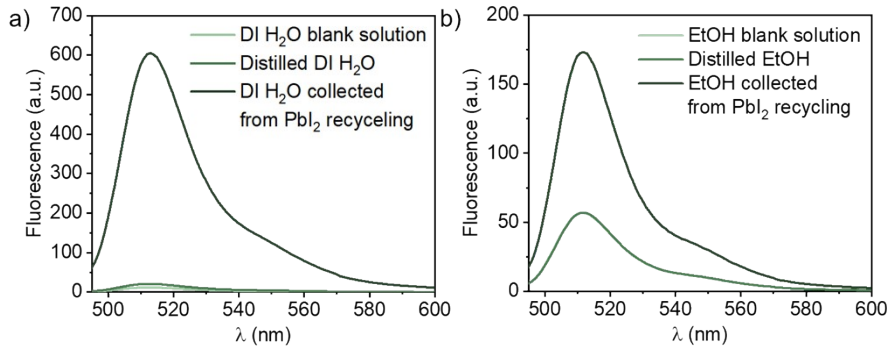

Fig. S19

a) Fluorescence spectra of deionized waster (DI H_2O) blank solution, distilled DI H_2O and DI H_2O collected from PbI_2 recycling. b) Fluorescence spectra of ethanol (EtOH) blank solution, EtOH and EtOH collected from PbI_2 recycling.

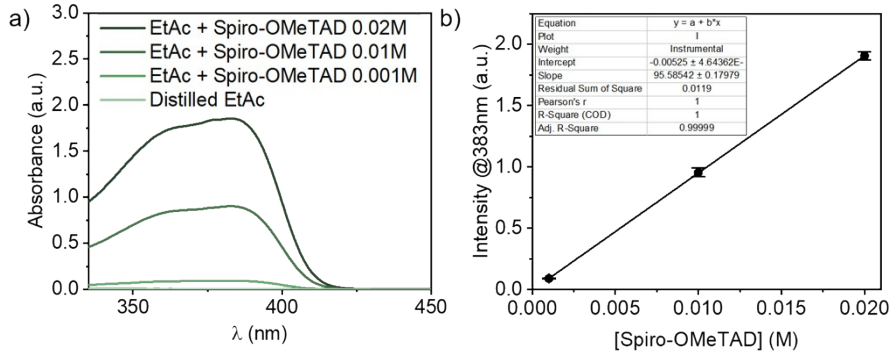

Fig. S20

a) UV-vis absorbance spectra of Spiro-OMeTAD in ethyl acetate (EtAc) 0.02 M, 0.01 M and 0.001 M solutions and of distilled EtAc. b) Calibration line generated for the 383 nm peak.

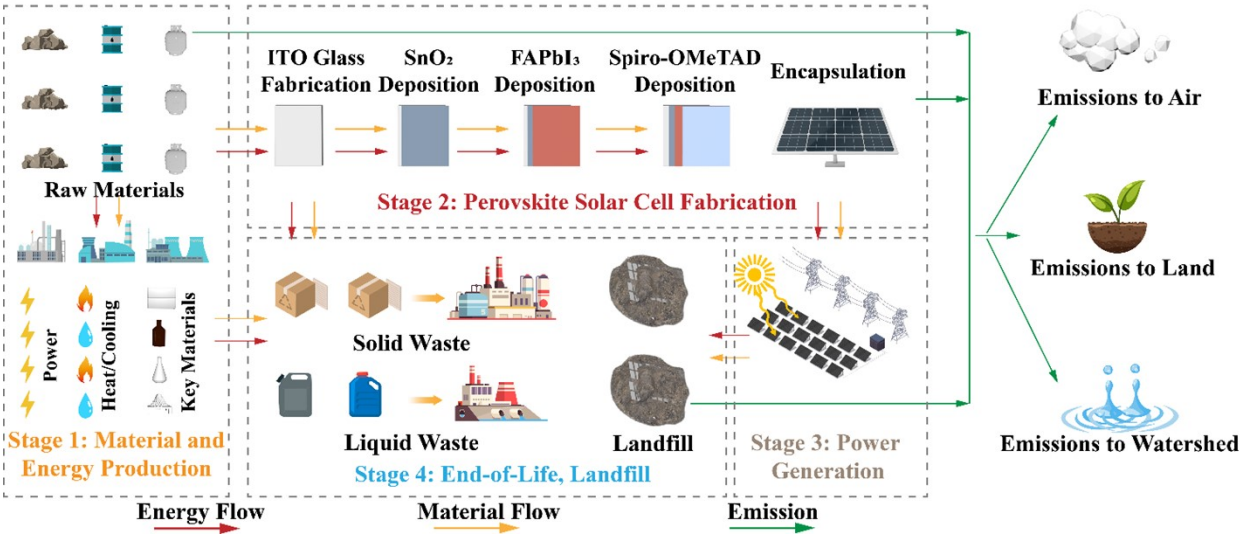

Fig. S21

System boundary of manufacturing perovskite solar modules with landfill as the end-of-life scenario, which is used to compare with Fig. S21. The main difference is that after stages 1, 2, and 3, the degraded perovskite cells will be landfilled directly rather than recycling.

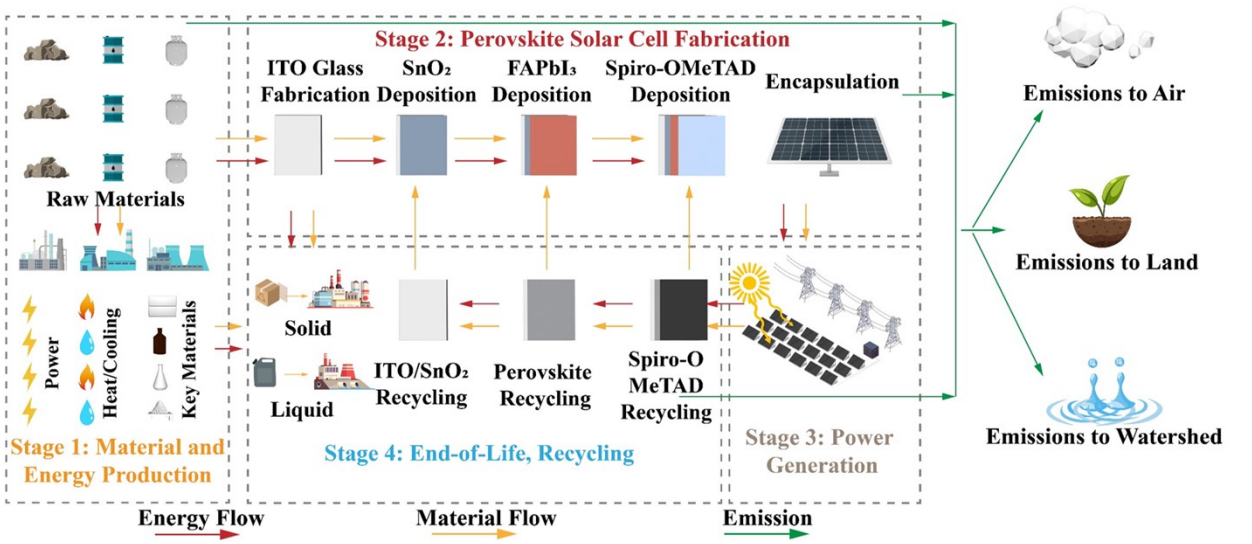

Fig. S22

System boundary of manufacturing perovskite solar modules with recycling as the end-of-life scenario.

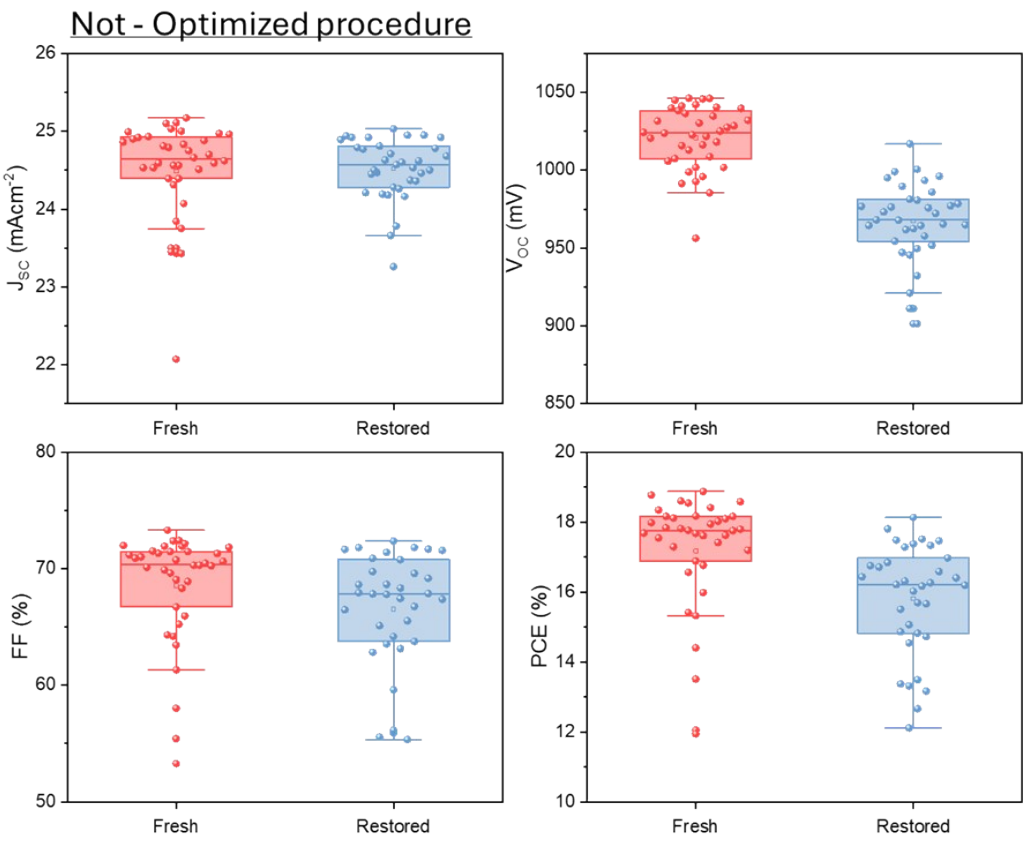

Fig. S23 PV parameters distribution of ITO/SnO_2 recycled without initial wash with detergent solution.

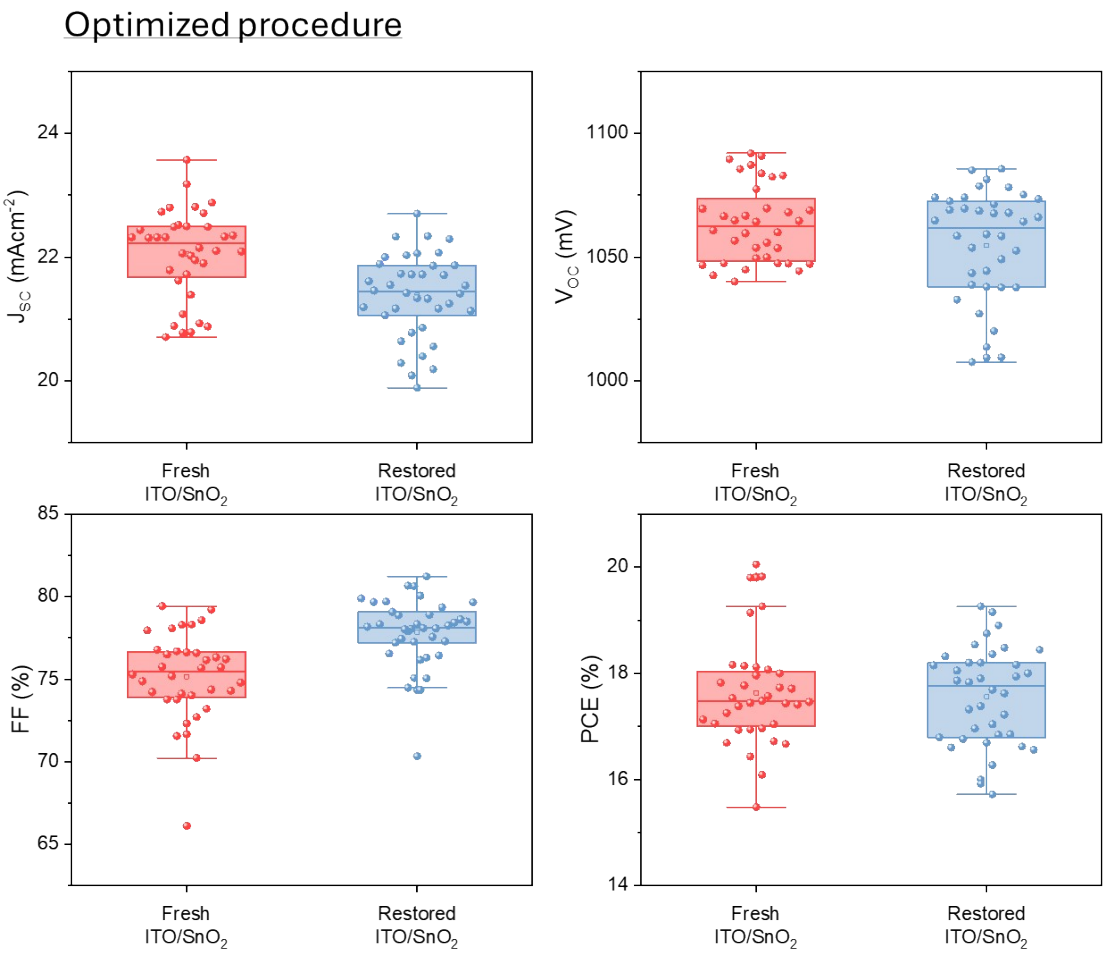

Fig. S24 PV parameters distribution of ITO/SnO_2 recycled with initial wash with hellmanex detergent solution.

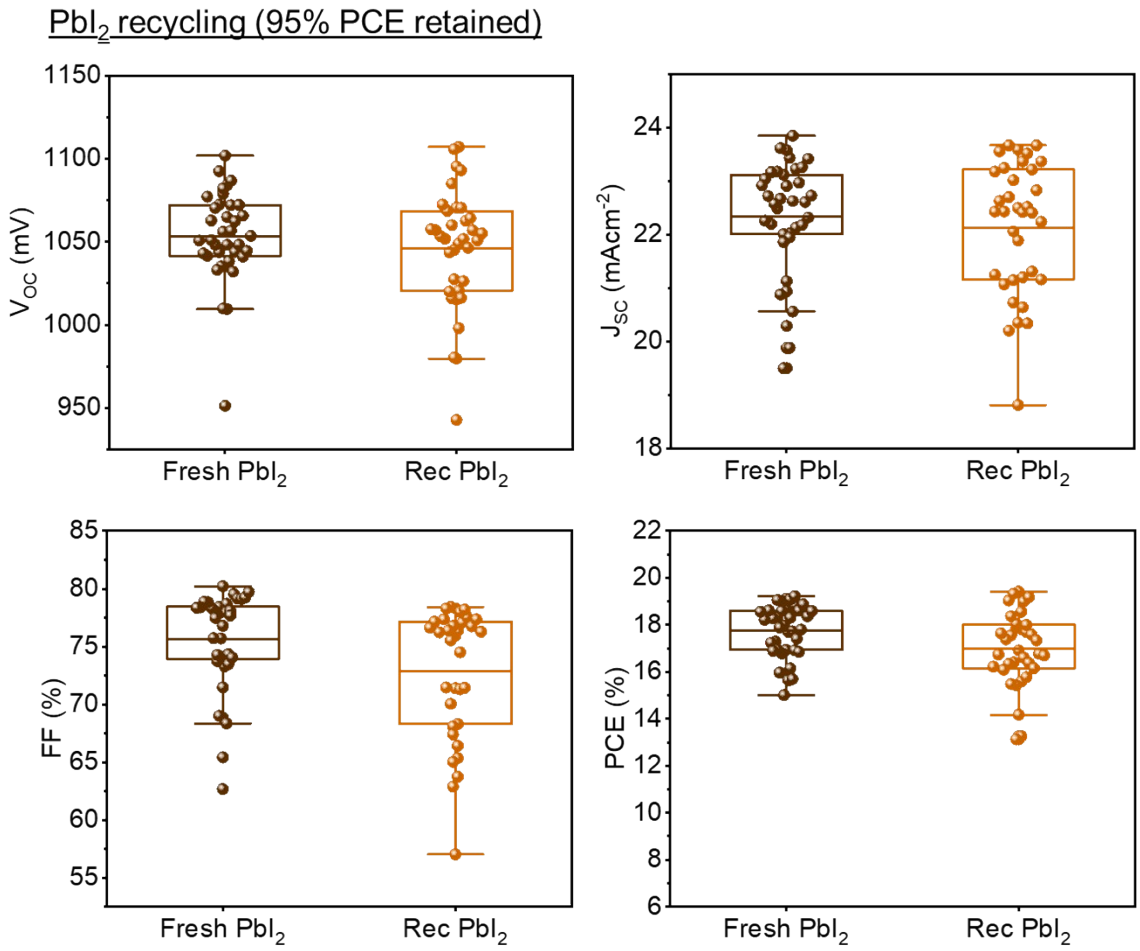

Fig. S25 PV parameters distribution of PbI_2 recycled with two washing cycles with ethanol.

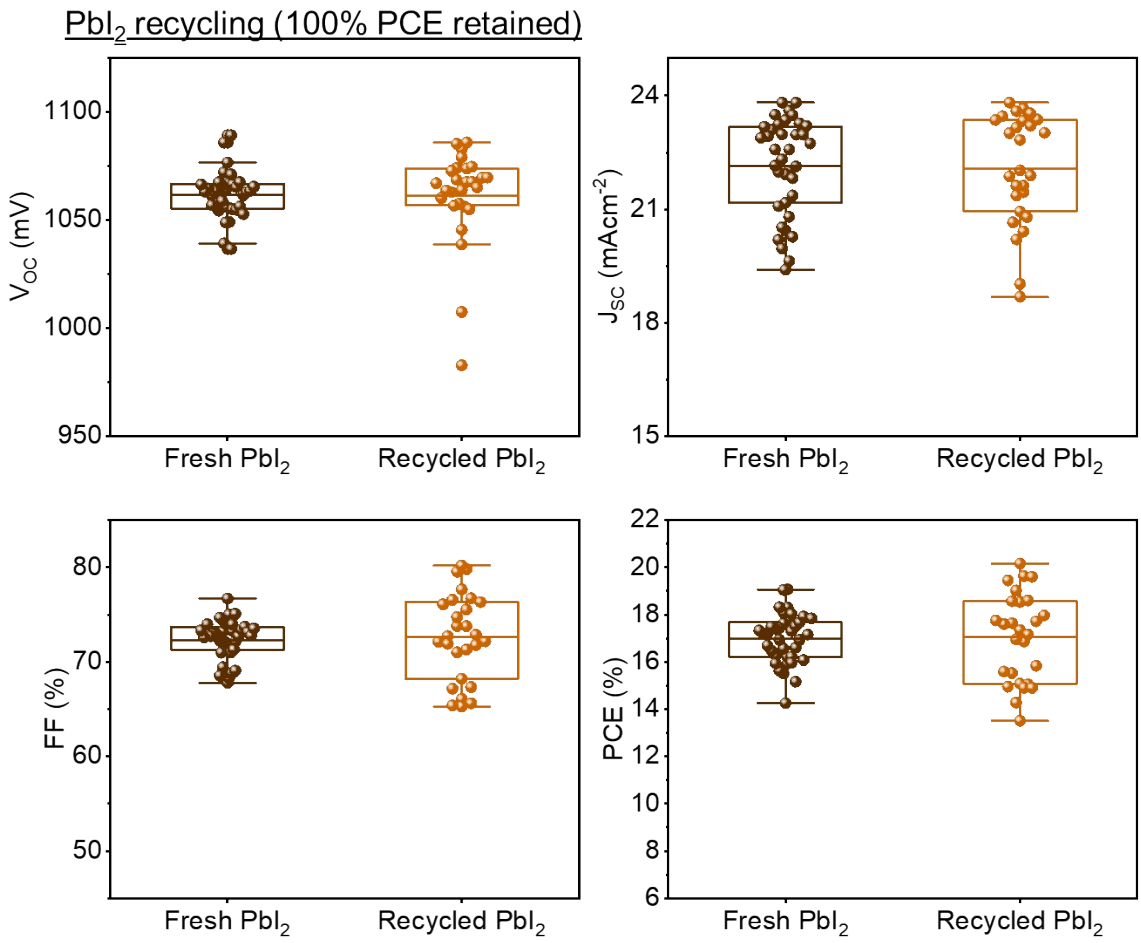

Fig. S26 PV parameters distribution of PbI2 recycled with four washing cycles with ethanol.

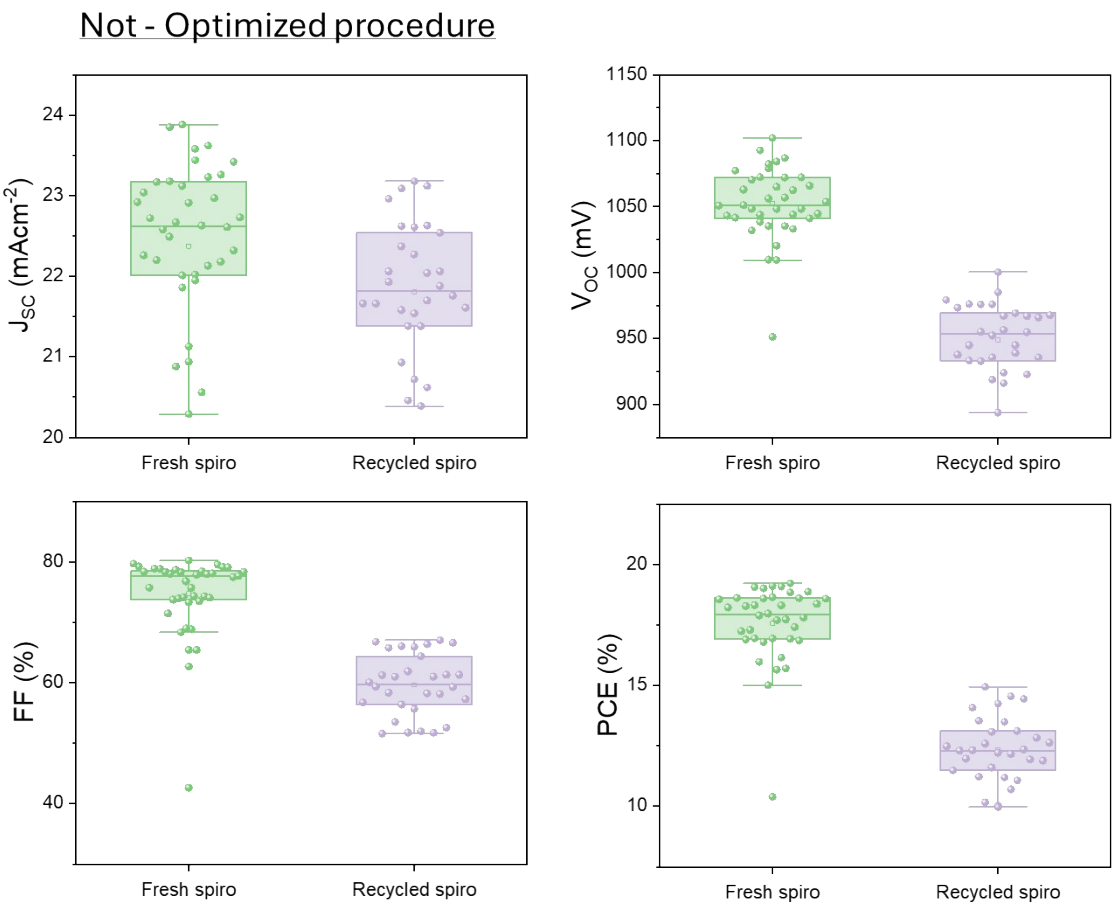

Fig. S27 PV parameters distribution of devices fabricated with Spiro recycled by using H_2O distilled in the procedure and with two steps of washing with Ethyl Acetate.

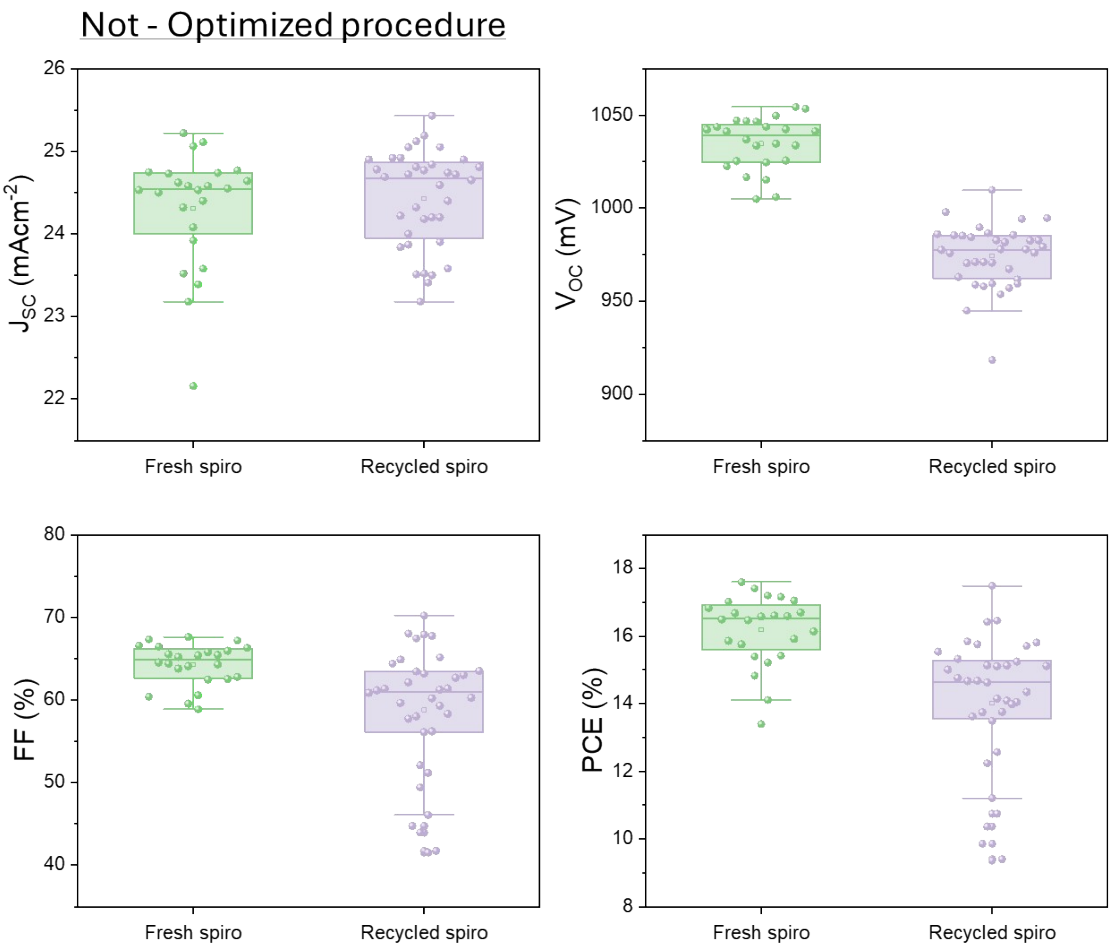

Fig. S28 PV parameters distribution of devices fabricated with Spiro recycled by using H_2O milliQ in the procedure and with two steps of washing with Ethyl Acetate.

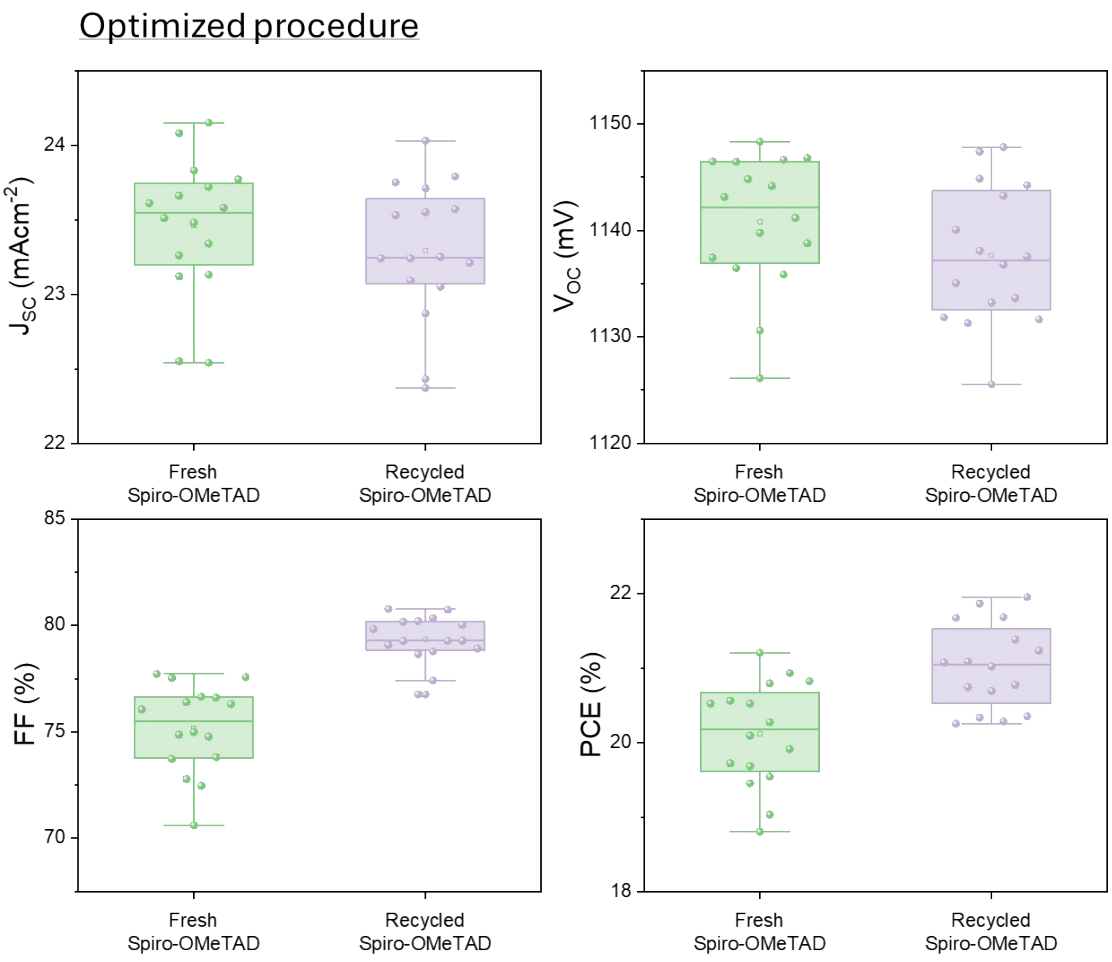

Fig. S29 PV parameters distribution of devices fabricated with Spiro recycled by using H_2O milliQ in the procedure and by passing from two to four washing with Ethyl Acetate.

Life cycle inventories

Tables S1-S4 exhibit the relevant life cycle inventories of perovskite production and recycling. Tables S1 and S2 show the materials and energy consumption for 1 m^2 perovskite solar cell production. Tables S3 and S4 describe the energy and materials inventories of 1 m^2 perovskite solar cell recycling process.

Table S1
Material inventory of 1 m^2 of the perovskite solar cell fabrication.

Process	Unit	Values
Acetone for cleaning	kg	8.00E-01
IPA for cleaning	kg	8.00E-01
ITO Glass	m^2	1.00E+00
SnO_2	kg	4.73E-05
Deionized H_2O	kg	3.47E-01
PbI_2	kg	1.24E-03
FAI	kg	4.76E-04
MACl	kg	6.12E-05
Anhydrous DMF	kg	1.31E-01
Anhydrous DMSO	kg	3.82E-02
Anhydrous chlorobenzene	kg	1.15E+00
MePEACl	kg	1.52E-05
Anhydrous IPA	kg	2.73E-01
Spiro-OMeTAD	kg	1.02E-04
Li TFSI	kg	4.72E-02
4-tert-butylpiridine	kg	1.38E-01
Cu	kg	7.17E-04
EVA	kg	1.00E-02
Solvent Treatment	kg	3.19E+00
Landfill	kg	2.94E+00

Table S2

Energy inventory of 1 m^2 of the perovskite solar cell fabrication.

Process	Electricity (kWh)
ITO/glass sonication during cleaning steps	3.38E+00
ITO/glass oxygen-plasma cleaning	6.04E+00
SnO_2 spin coating	2.92E-01
Spin coating vacuum pump	4.58E-02
SnO_2 annealing	2.50E+01
UV-ozone cleaner	5.75E+00
PbI_2 dissolution on hot plate	1.14E+00
Glovebox consumption during solution preparation	2.25E+00
Perovskite spin coating	3.40E-01
Spin coating vacuum pump	5.35E-02
Perovskite annealing	6.81E+00
Passivation layer spin coating	2.92E-01
Spin coating vacuum pump	4.58E-02
Spiro-OMeTAD spin coating	2.92E-01
Spin coating vacuum pump	4.58E-02
Glovebox consumption during fabrication	1.28E+00
Evaporation of metal contact	3.03E+00
Rotary vacuum pump for evaporation	6.25E+00
Turbomolecular vacuum pump for evaporation	6.00E+00
Total	6.83E+01

Table S3

Material inventory for the recycling process of 1 m^2 of the perovskite solar cell.

Process	Unit	Values
Acetone for cleaning	kg	8.00E-01
IPA for cleaning	kg	8.00E-01
Deionized H_2O	kg	1.74E+00
Cleaning concentrate	kg	2.05E-04
Ethanol	kg	1.83E+00
FAI	kg	4.76E-04
MACl	kg	6.12E-05
MePEACl	kg	1.52E-05
Cu	kg	7.17E-04
Ethyl acetate	kg	5.22E-01
Ultrapure water	kg	2.31E+00
Li TFSI	kg	4.72E-02
4-tert-butylpiridine	kg	1.38E-01
Solvent treatment	kg	3.95E+00
EVA	kg	1.00E-02

Table S4

Energy inventory for the recycling process of 1 m^2 of the perovskite solar cell.

Process	Electricity (kWh)
Energy for recycling of 1m^2 Perovskite Layer	
Sonicator	1.13E+00
Centrifuge	1.73E+01
Rotavapor	6.25E-03
Rotavapor vacuum pump	1.56E-02
Rotavapor heating bath	1.77E-01
Energy for recycling of 1m^2 ITO Glass	
Sonicator	6.75E+00
Energy for recycling of 1m^2 Spiro-OMeTAD	
Rotavapor	6.25E-03
Rotavapor vacuum pump	1.56E-02
Rotavapor heating bath	1.77E-01
Cu	3.03E+00
Energy for purifying solvents	
Hot plate	2.50E+01
Total	5.36E+01

LCA contribution analysis

Fig. S22 and Fig. S23 show the environmental profiles of the perovskite landfill and recycling for the explored midpoint impact categories according to the PEF method, respectively. These two figures exhibit the key factors contributing to the environmental impacts and guide future perovskite solar cell production and recovery to further reduce the impacts.

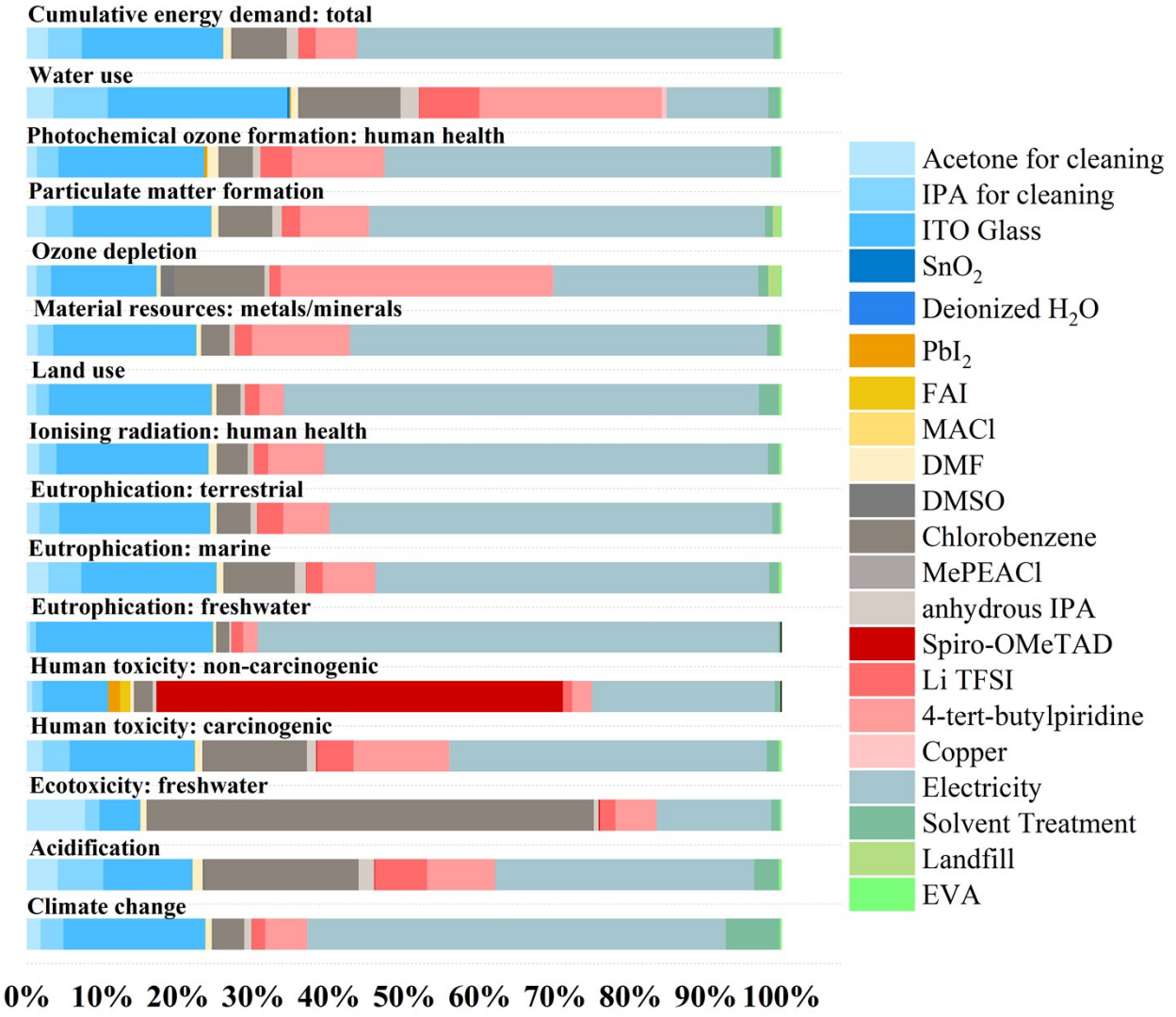

Fig. S22

Contribution analysis results for the perovskite solar cell with landfill as the end-of-life scenario.

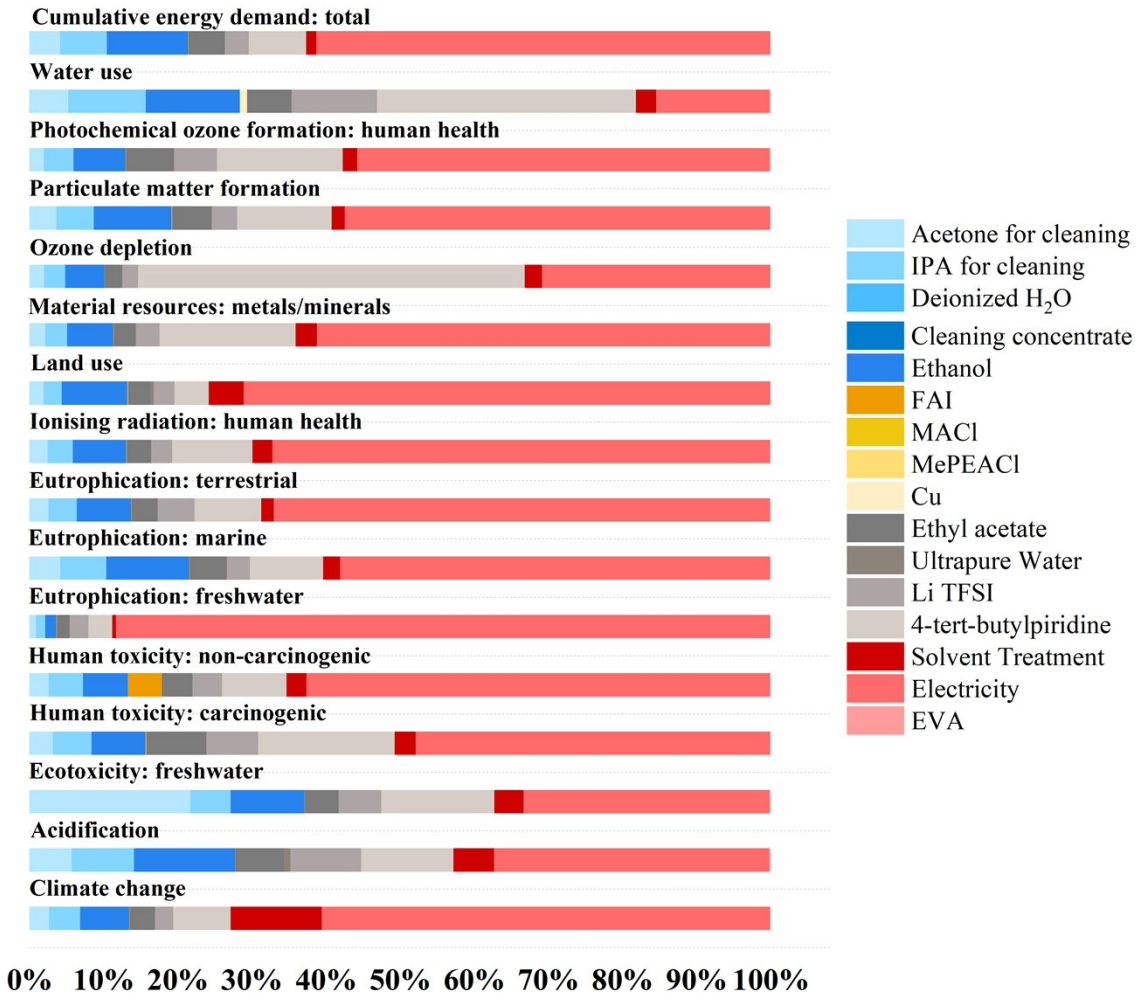

Fig. S23

Contribution analysis results for the perovskite solar cell with recycling as the end-of-life scenario.