Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

SUPPORTING INFORMATION

Evaluation of multiple cation/anion perovskite solar cells through life cycle assessment

Jaume-Adrià Alberola-Borràs,[a,b] Rosario Vidal,[a,]* Iván Mora-Seró[b,]*

[a] Grupo de Ingeniería de Diseño (GID), Departamentd'enginyeriamecànica i construcció, Universitat Jaume I, Av. SosBaynat s/n, 12071 Castelló, Spain

[b]Institute of Advanced Materials (INAM), UniversitatJaume I, Av. SosBaynat, s/n, 12006 Castelló, Spain

*Corresponding author. E-mail: vidal@uji.es; sero@uji.es. Tel: +34 964387552

1 Inventory

The inventory of triple cation perovskites was extracted from the work in which all their compositions are reported.[1] Meanwhile, the canonical perovskite inventory was modelled from the manuscript of Noh et al.[2] Inventories of the five perovskites are shown in Table S1.

Table S1 Inventory of flows used for the perovskites analysed.

Name	Value for 0% caesium perovskite	Value for 5% caesium perovskite	Value for 10% caesium perovskite	Value for 15% caesium perovskite	Value for canonical perovskite	Unit	Observations
Reagents							
CsI	0.00E+00	4.38E-06	8.76E-06	1.31E-05		g/cm^2	Detailed inventory in Table S2
FAI	4.81E-05	4.57E-05	4.33E-05	4.09E-05		g/cm^2	Detailed inventory in Table S3
MABr	6.42E-06	6.09E-06	5.77E-06	5.45E-06		g/cm^2	Detailed inventory in Table S4
MAI					5.24E-05	g/cm^2	Detailed inventory in Table S5
PbI_2	1.29E-04	1.28E-04	1.28E-04	1.27E-04	1.52E-04	g/cm^2	Detailed inventory in Table S6
$PbBr_2$	2.10E-05	2.16E-05	2.21E-05	2.26E-05		g/cm^2	Detailed inventory in Table S7
Solvents							
DMF	7.99E-04	7.91E-04	7.83E-04	7.74E-04		g/cm^2	DMF solvent from

							Ecoinvent database[3]
DMSO	2.33E-04	2.30E-04	2.28E-04	2.26E-04		g/cm^2	DMSO solvent from Ecoinvent database[3]
CB	4.44E-03					g/cm^2	CB solvent from Ecoinvent database[3]
GBL					3.07E-04	g/cm^2	GBL solvent from Ecoinvent database[3]
Outputs							
Emissions - DMF	7.99E-04	7.91E-04	7.83E-04	7.74E-04		g/cm^2	Emissions to air due to evaporation of DMF solvent
Emissions - DMSO	2.33E-04	2.30E-04	2.28E-04	2.26E-04		g/cm^2	Emissions to air due to evaporation of DMSO solvent
Emissions - CB	4.44E-03					g/cm^2	Emissions to air due to evaporation of CB solvent
Emissions - GBL					3.07E-04	g/cm^2	Emissions to air due to evaporation of GBL solvent
Amount of transportation							
Transport, lorry > 16 t	2.71E-06	2.70E-06	2.70E-06	2.69E-06	2.20E-07	tkm/cm^2	Distances considered from suppliers to Castelló (Spain)
Use of energy							
Electricity, low voltage	2.12E-03				2.09E-03	MJ/cm^2	European electricity mix from ecoinvent[3]

1.1 Inventory of caesium iodide model

The recovery of caesium from pollucite ore was modelled according to a method in which the raw pollucite is digested with sulphuric acid.[4] Its inventory is shown in Table S2. The caesium iodide supplier was abcr GmBH in Pontevedra (Spain). Its distance to Castelló (Spain) is 974 km.

Table S2 Inventory for 1 kg of caesium iodide.

Name	Value	Unit
Materials/fuels		
Sulphuric acid	754.989	g
Lime	431.677	g
Iodine	488.434	g
Hydrogen	3.849	g
Water, deionised	10271.291	g
Pollucite Ore	1694.725	g
Electricity/heat		
Heat, natural gas, <100kW	3.58	kWh
Electricity, low voltage	0.027	kWh
Crushing, rock	1694.725	g
Chemical plant, organics	4E-10	p
Emissions to air		
Water	9924.884	g
Emissions to water		
Aluminium oxide	196.222	g
Silicon dioxide	925.045	g
Sodium hydroxide	303.988	g
Aluminium oxide	196.222	g
Waste to treatment		
Waste gypsum (waste treatment)	1325.342	g

1.2 Inventory of formamidinium iodide model

The inventory of formamidinium iodide was modelled from three reactions in which hydrogen cyanide, hydroxylamine, acetic acid and hydroiodic acid are involved.[5–7] It is shown in Table S3. The formamidinium iodide supplier was Dyesol Limited in Manchester (United Kingdom). Its distance to Castelló (Spain) is 2013 km.

Table S3 Inventory for 1 kg of formamidinium iodide.

Name	**Value**	**Unit**
Materials/fuels		
Hydrogen cyanide	157.153	g
Hydroxylamine	192.068	g
Acetic anhydride	296.825	g
Hydrogen	5.815	g
Iodine	737.919	g
Water, deionised	423.965	g
Diethyl ether	101.588	g
Methanol	838.438	g
Chemical plant, organics	4E-10	p
Electricity/heat		
Electricity, low voltage	197.79	kWh
Emissions to air		
Diethyl ether	101.588	g
Methanol	838.438	g
Emissions to water		
Water	476.3	g
Acetic acid	349.189	g

1.3 Inventory of methylammonium bromide model

In Table S4, the methylammonium bromide inventory was modelled according to a reaction of methylamine and hydrobromic acid.[8] The methylammonium bromide supplier was Dyesol Limited in Manchester (United Kingdom). Its distance to Castelló (Spain) is 2013 km.

Table S4 Inventory for 1 kg of methylammonium bromide.

Name	Value	Unit
Materials/fuels		
Methylamine	277.023	g
Hydrogen	8.936	g
Bromine	714.041	g
Methanol	415.535	g
Water, deionised	783.225	g
Ethanol	11.103	g
Diethyl ether	10.033	g
Chemical plant, organics	4E-10	p
Electricity/heat		
Electricity, low voltage	143.05	kWh
Emissions to air		
Methanol	415.535	g
Water	783.225	g
Ethanol	11.103	g
Diethyl ether	10.033	g

1.4 Inventory of methylammonium iodide model

In Table S5, the methylammonium iodide inventory was modelled according to a reaction of methylamine and hydrobromic acid.[2] The formamidinium iodide supplier was Sigma Aldrich in Madrid (Spain). Its distance to Castelló (Spain) is 430 km.

Table S5 Inventory for 1 kg of methylammonium iodide.

Name	Value	Unit
Materials/fuels		
Methylamine	195.086	g
Hydrogen	6.293	g
Iodine	798.621	g
Methanol	292.629	g
Water, deionised	607.216	g
Ethanol	8.733	g
Diethyl ether	7.892	g
Chemical plant, organics	4E-10	p
Electricity/heat		
Electricity, low voltage	90.87	kWh
Emissions to air		
Methanol	292.629	g
Water	607.216	g
Ethanol	8.733	g
Diethyl ether	7.892	g

1.5 Inventory of lead iodide model

In Table S6, the lead iodide inventory was modelled according to a reaction of lead nitrate and potassium iodide.[9] The lead iodide supplier was TCI Europe N.V. in Zwijndrecht (Belgium). Its distance to Castelló (Spain) is 1645 km.

Table S6 Inventory for 1 kg of lead iodide.

Name	Value	Unit
Materials/fuels		
Lead, primary	449.449	g
Nitric acid, 50% in H_2O	546.627	g
Potassium hydroxide	243.372	g
Iodine	550.551	g
Chemical plant, organics	4E-10	p
Electricity/heat		
Electricity, low voltage	114.4	kWh
Emissions to air		
Hydrogen	6.507	g
Emissions to water		
Water	39.045	g
Nitric acid	273.314	g
Potassium bicarbonate	434.257	g

1.6 Inventory of lead bromide model

In Table S7, the lead bromide inventory was modelled according to a reaction of lead nitrate and potassium bromide.[9] The lead bromide supplier was TCI Europe N.V. in Zwijndrecht (Belgium). Its distance to Castelló (Spain) is 1645 km.

Table S7 Inventory for 1 kg of lead bromide.

Name	Value	Unit
Materials/fuels		
Lead, primary	564.565	g
Nitric acid, 50% in H_2O	686.634	g
Potassium hydroxide	305.706	g
Bromine	435.435	g
Chemical plant, organics	4E-10	p
Electricity/heat		
Electricity, low voltage	143.7	kWh
Emissions to air		
Hydrogen	8.174	g
Emissions to water		
Water	49.045	g
Nitric acid	545.483	g
Potassium bicarbonate	343.317	g

1.7 Economic indicators

The economic analysis was performed to support the information provided by the environmental analysis. In this analysis, prices of reagents and solvents were obtained from the most economical retail prices found in October 2017, which are described in Table S8.

Table S8 Economic indicators of the reagents, solvents and electricity.

Economic indicator (reference unit)	Price (€)	Supplier
Electricity (1 MJ)	0.036	Spanish network cost
Caesium iodide (1 kg)	480.00	abcr
Formamidinium iodide (1 kg)	2299.49	Dyesol
Methylammonium Bromide (1 kg)	879.76	Dyesol
Methylammonium Iodide (1 kg)	763.86	Dyesol
Lead Bromide (1 kg)	199.80	Sigma Aldrich
Lead Iodide (1 kg)	1246.00	Sigma Aldrich
N,N-dimethylformamide (1 kg)	18.05	Right Price Chemicals
Dimethyl sulfoxide (1 kg)	42.60	Sigma Aldrich
Chlorobenzene (1 kg)	7.00	Noah Technologies Corporation
Butyrolactone (1 kg)	23.85	Sigma Aldrich

2 Results

2.1 Impact results

Absolute results sorted into types of flow for 0% caesium perovskite (Table S9), 5% caesium perovskite (Table S10), 10% caesium perovskite (Table S11), 15% caesium perovskite (Table S12) and canonical perovskite (Table S13).

Table S9 Impact results of 0% caesium perovskite sorted by flows.

	ADP (kg Sb eq)	ADPF (MJ)	GWP (kg CO_2eq)	ODP (kg CFC-11 eq)	POP (kg C_2H_4eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)	CED (MJ)	HTC (CTUh)	HTNC (CTUh)	FET (CTUe)
Total	3.83E-09	4.49E-03	3.41E-04	2.10E-11	9.16E-08	1.64E-06	1.18E-06	7.88E-03	3.20E-11	1.32E-10	3.16E-03
Reagents	3.23E-09	2.10E-04	1.64E-05	9.14E-13	1.16E-08	8.37E-08	5.57E-08	3.65E-04	1.49E-12	7.38E-12	1.51E-04
CsI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
FAI	8.99E-10	7.37E-05	5.57E-06	3.27E-13	9.03E-09	2.91E-08	1.76E-08	1.26E-04	4.93E-13	2.04E-12	4.89E-05
MABr	2.10E-11	6.77E-06	5.18E-07	2.92E-14	5.31E-10	2.48E-09	1.60E-09	1.18E-05	4.66E-14	1.91E-13	4.49E-06
PbI_2	2.19E-09	1.08E-04	8.61E-06	4.66E-13	1.73E-09	4.34E-08	3.03E-08	1.89E-04	7.86E-13	4.28E-12	8.07E-05
$PbBr_2$	1.24E-10	2.17E-05	1.74E-06	9.11E-14	3.48E-10	8.77E-09	6.18E-09	3.83E-05	1.60E-13	8.72E-13	1.64E-05
Solvents	5.91E-11	2.74E-04	1.08E-05	4.47E-12	1.96E-08	4.52E-08	1.19E-07	3.19E-04	7.25E-13	3.99E-12	1.99E-04
DMF	5.85E-12	3.87E-05	1.48E-06	2.45E-13	4.53E-10	6.57E-09	7.10E-08	4.49E-05	8.52E-14	4.17E-13	9.34E-06
DMSO	2.50E-11	9.80E-06	2.99E-07	4.42E-14	6.36E-11	7.75E-10	4.44E-10	1.07E-05	1.47E-14	7.21E-14	1.58E-06
CB	2.82E-11	2.26E-04	8.99E-06	4.18E-12	1.91E-08	3.78E-08	4.74E-08	2.63E-04	6.26E-13	3.50E-12	1.88E-04
Process outputs	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	1.93E-12	4.00E-06
Emissions - DMF	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.45E-12	2.37E-07
Emissions - DMSO	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.08E-09
Emissions - CB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	4.84E-13	3.76E-06
Transport, lorry >16t	9.75E-13	5.31E-06	3.60E-07	5.82E-14	5.86E-11	1.96E-09	5.23E-10	5.72E-06	2.21E-14	6.59E-14	1.46E-06
Electricity, low voltage	5.40E-10	4.00E-03	3.13E-04	1.55E-11	6.03E-08	1.51E-06	1.00E-06	7.19E-03	2.93E-11	1.18E-10	2.81E-03

Table S10 Impact results of 5% caesium perovskite sorted by flows.

	ADP (kg Sb eq)	ADPF (MJ)	GWP (kg CO_2eq)	ODP (kg CFC-11 eq)	POP (kg C_2H_4eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)	CED (MJ)	HTC (CTUh)	HTNC (CTUh)	FET (CTUe)
Total	3.83E-09	4.49E-03	3.40E-04	2.10E-11	9.11E-08	1.64E-06	1.18E-06	7.87E-03	3.20E-11	1.32E-10	3.16E-03
Reagents	3.23E-09	2.06E-04	1.61E-05	8.98E-13	1.12E-08	8.24E-08	5.47E-08	3.58E-04	1.46E-12	7.27E-12	1.48E-04
CsI	5.36E-11	2.21E-07	1.66E-08	2.57E-15	9.78E-12	2.27E-10	1.39E-11	2.38E-07	4.35E-16	3.24E-15	5.94E-08
FAI	8.54E-10	7.00E-05	5.29E-06	3.11E-13	8.58E-09	2.76E-08	1.67E-08	1.20E-04	4.69E-13	1.94E-12	4.65E-05
MABr	2.00E-11	6.43E-06	4.92E-07	2.78E-14	5.05E-10	2.35E-09	1.52E-09	1.12E-05	4.43E-14	1.81E-13	4.27E-06
PbI_2	2.17E-09	1.07E-04	8.56E-06	4.64E-13	1.72E-09	4.32E-08	3.02E-08	1.88E-04	7.82E-13	4.26E-12	8.03E-05
$PbBr_2$	1.27E-10	2.23E-05	1.78E-06	9.34E-14	3.57E-10	8.99E-09	6.34E-09	3.93E-05	1.64E-13	8.94E-13	1.68E-05
Solvents	5.87E-11	2.74E-04	1.08E-05	4.47E-12	1.96E-08	4.51E-08	1.18E-07	3.18E-04	7.24E-13	3.98E-12	1.99E-04
DMF	5.79E-12	3.83E-05	1.47E-06	2.43E-13	4.49E-10	6.50E-09	7.03E-08	4.44E-05	8.43E-14	4.13E-13	9.25E-06
DMSO	2.47E-11	9.70E-06	2.96E-07	4.38E-14	6.30E-11	7.67E-10	4.39E-10	1.06E-05	1.46E-14	7.14E-14	1.56E-06
CB	2.82E-11	2.26E-04	8.99E-06	4.18E-12	1.91E-08	3.78E-08	4.74E-08	2.63E-04	6.26E-13	3.50E-12	1.88E-04
Process outputs	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	1.92E-12	4.00E-06
Emissions - DMF	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.43E-12	2.34E-07
Emissions - DMSO	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.98E-09
Emissions - CB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	4.84E-13	3.76E-06
Transport, lorry >16t	9.73E-13	5.30E-06	3.59E-07	5.81E-14	5.85E-11	1.95E-09	5.21E-10	5.71E-06	2.21E-14	6.58E-14	1.46E-06
Electricity, low voltage	5.40E-10	4.00E-03	3.13E-04	1.55E-11	6.03E-08	1.51E-06	1.00E-06	7.19E-03	2.93E-11	1.18E-10	2.81E-03

Table S11 Impact results of 10% caesium perovskite sorted by flows.

	ADP (kg Sb eq)	ADPF (MJ)	GWP (kg CO_2eq)	ODP (kg CFC-11 eq)	POP (kg C_2H_4eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)	CED (MJ)	HTC (CTUh)	HTNC (CTUh)	FET (CTUe)
Total	3.83E-09	4.48E-03	3.40E-04	2.09E-11	9.07E-08	1.64E-06	1.18E-06	7.86E-03	3.19E-11	1.32E-10	3.16E-03
Reagents	3.23E-09	2.02E-04	1.59E-05	8.83E-13	1.07E-08	8.10E-08	5.38E-08	3.52E-04	1.43E-12	7.17E-12	1.45E-04
CsI	1.07E-10	4.41E-07	3.33E-08	5.15E-15	1.96E-11	4.54E-10	2.78E-11	4.76E-07	8.70E-16	6.48E-15	1.19E-07
FAI	8.09E-10	6.64E-05	5.01E-06	2.95E-13	8.13E-09	2.62E-08	1.58E-08	1.14E-04	4.44E-13	1.83E-12	4.40E-05
MABr	1.89E-11	6.09E-06	4.66E-07	2.63E-14	4.78E-10	2.23E-09	1.44E-09	1.06E-05	4.19E-14	1.72E-13	4.05E-06
PbI_2	2.16E-09	1.07E-04	8.52E-06	4.61E-13	1.71E-09	4.30E-08	3.00E-08	1.87E-04	7.78E-13	4.24E-12	7.99E-05
$PbBr_2$	1.30E-10	2.28E-05	1.82E-06	9.56E-14	3.66E-10	9.21E-09	6.49E-09	4.02E-05	1.68E-13	9.15E-13	1.73E-05
Solvents	5.84E-11	2.73E-04	1.07E-05	4.46E-12	1.96E-08	4.50E-08	1.17E-07	3.18E-04	7.23E-13	3.98E-12	1.99E-04
DMF	5.73E-12	3.79E-05	1.45E-06	2.40E-13	4.44E-10	6.43E-09	6.95E-08	4.40E-05	8.34E-14	4.09E-13	9.15E-06
DMSO	2.45E-11	9.60E-06	2.93E-07	4.33E-14	6.23E-11	7.59E-10	4.35E-10	1.05E-05	1.44E-14	7.07E-14	1.55E-06
CB	2.82E-11	2.26E-04	8.99E-06	4.18E-12	1.91E-08	3.78E-08	4.74E-08	2.63E-04	6.26E-13	3.50E-12	1.88E-04
Process outputs	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	1.90E-12	4.00E-06
Emissions - DMF	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.42E-12	2.32E-07
Emissions - DMSO	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.89E-09
Emissions - CB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.66E-13	4.84E-13	3.76E-06
Transport, lorry >16t	9.71E-13	5.28E-06	3.58E-07	5.79E-14	5.83E-11	1.95E-09	5.20E-10	5.69E-06	2.20E-14	6.56E-14	1.45E-06
Electricity, low voltage	5.40E-10	4.00E-03	3.13E-04	1.55E-11	6.03E-08	1.51E-06	1.00E-06	7.19E-03	2.93E-11	1.18E-10	2.81E-03

Table S12 Impact results of 15% caesium perovskite sorted by flows.

	ADP (kg Sb eq)	ADPF (MJ)	GWP (kg CO_2eq)	ODP (kg CFC-11 eq)	POP (kg C_2H_4eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)	CED (MJ)	HTC (CTUh)	HTNC (CTUh)	FET (CTUe)
Total	3.83E-09	4.48E-03	3.40E-04	2.09E-11	9.02E-08	1.63E-06	1.17E-06	7.86E-03	3.19E-11	1.31E-10	3.15E-03
Reagents	3.23E-09	1.99E-04	1.56E-05	8.68E-13	1.02E-08	7.97E-08	5.29E-08	3.45E-04	1.41E-12	7.06E-12	1.43E-04
CsI	1.61E-10	6.62E-07	4.99E-08	7.72E-15	2.93E-11	6.80E-10	4.16E-11	7.14E-07	1.30E-15	9.72E-15	1.78E-07
FAI	7.64E-10	6.27E-05	4.74E-06	2.78E-13	7.68E-09	2.47E-08	1.49E-08	1.07E-04	4.19E-13	1.73E-12	4.16E-05
MABr	1.79E-11	5.75E-06	4.40E-07	2.49E-14	4.52E-10	2.11E-09	1.36E-09	1.00E-05	3.96E-14	1.62E-13	3.82E-06
PbI_2	2.15E-09	1.06E-04	8.47E-06	4.59E-13	1.70E-09	4.27E-08	2.99E-08	1.86E-04	7.74E-13	4.22E-12	7.95E-05
$PbBr_2$	1.33E-10	2.34E-05	1.87E-06	9.79E-14	3.74E-10	9.43E-09	6.65E-09	4.12E-05	1.72E-13	9.37E-13	1.77E-05
Solvents	5.81E-11	2.73E-04	1.07E-05	4.46E-12	1.96E-08	4.50E-08	1.17E-07	3.17E-04	7.22E-13	3.97E-12	1.99E-04
DMF	5.67E-12	3.75E-05	1.44E-06	2.38E-13	4.39E-10	6.36E-09	6.88E-08	4.35E-05	8.25E-14	4.04E-13	9.05E-06
DMSO	2.42E-11	9.50E-06	2.90E-07	4.29E-14	6.17E-11	7.51E-10	4.30E-10	1.03E-05	1.43E-14	6.99E-14	1.53E-06
CB	2.82E-11	2.26E-04	8.99E-06	4.18E-12	1.91E-08	3.78E-08	4.74E-08	2.63E-04	6.26E-13	3.50E-12	1.88E-04
Process outputs	5.67E-12	3.75E-05	1.44E-06	2.38E-13	4.39E-10	6.36E-09	6.88E-08	4.35E-05	8.25E-14	4.04E-13	9.05E-06
Emissions - DMF	2.42E-11	9.50E-06	2.90E-07	4.29E-14	6.17E-11	7.51E-10	4.30E-10	1.03E-05	1.43E-14	6.99E-14	1.53E-06
Emissions - DMSO	2.82E-11	2.26E-04	8.99E-06	4.18E-12	1.91E-08	3.78E-08	4.74E-08	2.63E-04	6.26E-13	3.50E-12	1.88E-04
Emissions - CB	5.67E-12	3.75E-05	1.44E-06	2.38E-13	4.39E-10	6.36E-09	6.88E-08	4.35E-05	8.25E-14	4.04E-13	9.05E-06
Transport, lorry >16t	2.42E-11	9.50E-06	2.90E-07	4.29E-14	6.17E-11	7.51E-10	4.30E-10	1.03E-05	1.43E-14	6.99E-14	1.53E-06
Electricity, low voltage	5.40E-10	4.00E-03	3.13E-04	1.55E-11	6.03E-08	1.51E-06	1.00E-06	7.19E-03	2.93E-11	1.18E-10	2.81E-03

Table S13 Impact results of canonical perovskite sorted by flows.

	ADP (kg Sb eq)	ADPF (MJ)	GWP (kg CO_2eq)	ODP (kg CFC-11 eq)	POP (kg C_2H_4eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)	CED (MJ)	HTC (CTUh)	HTNC (CTUh)	FET (CTUe)
Total	3.75E-09	1.06E-03	8.24E-05	4.23E-12	2.25E-08	3.97E-07	2.65E-07	1.88E-03	7.62E-12	3.21E-11	7.46E-04
Reagents	3.63E-09	1.63E-04	1.29E-05	7.17E-13	9.07E-09	6.44E-08	4.42E-08	2.85E-04	1.17E-12	6.06E-12	1.19E-04
MAI	1.05E-09	3.64E-05	2.78E-06	1.67E-13	7.04E-09	1.33E-08	8.40E-09	6.25E-05	2.45E-13	1.01E-12	2.37E-05
PbI_2	2.58E-09	1.27E-04	1.01E-05	5.50E-13	2.04E-09	5.12E-08	3.58E-08	2.23E-04	9.27E-13	5.05E-12	9.52E-05
Solvents (GBL)	1.81E-12	1.88E-05	9.99E-07	1.07E-13	2.39E-10	3.09E-09	1.29E-09	2.17E-05	3.92E-14	1.85E-13	3.82E-06
Process outputs (Emissions - GBL)	1.81E-12	1.88E-05	9.99E-07	1.07E-13	2.39E-10	3.09E-09	1.29E-09	2.17E-05	3.92E-14	1.85E-13	3.82E-06
Transport, lorry >16t	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.18E-06
Electricity, low voltage	1.18E-10	8.75E-04	6.85E-05	3.40E-12	1.32E-08	3.30E-07	2.20E-07	1.57E-03	6.41E-12	2.59E-11	6.14E-04

For the sake of providing assistance to find the most responsible type of flow of the impacts of perovskites, impacts are distributed per types of flow and compared in a diagram. Figure S1 presents the percentage of impact generated per type of inventory flow. It depicts the categories ADP, GWP, CED, HTC and HTNC, which represent the most crucial categories for the assessment of PSCs.

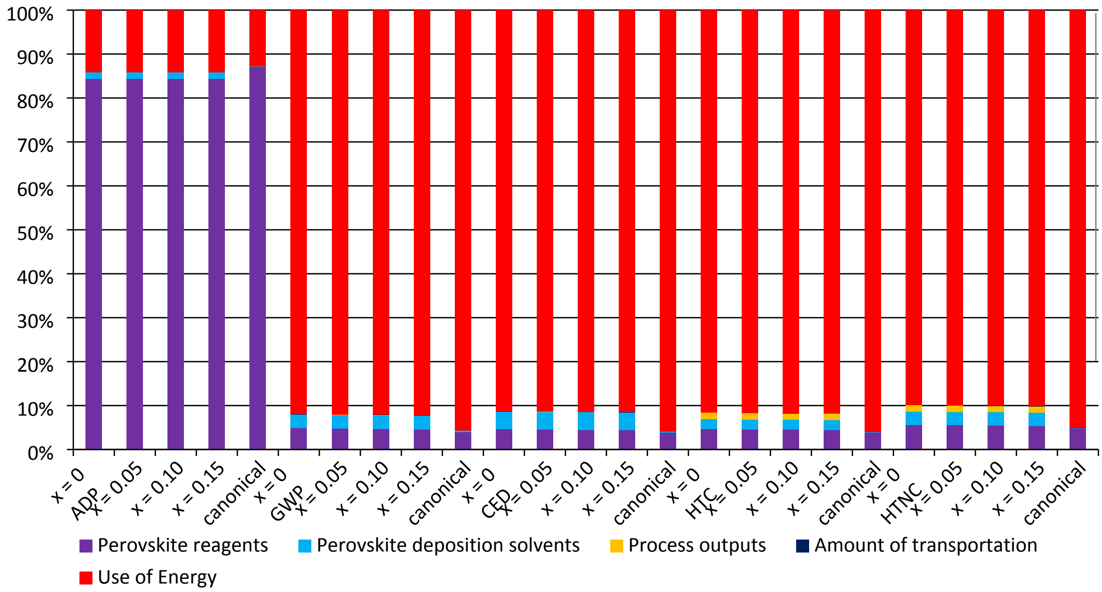

Figure S1. Distribution of impacts per type of flow for the most representative categories to assess PSCs.

From Figure S1, a general trend in which the energy used is the most harmful flow is observed, supposing more than 80% of the total. This circumstance occurs for all categories displayed, except for ADP category. Thus, for ADP perovskite reagents contribution exceeds an 80% and energy is the second most harmful type of flow. For the canonical perovskite, the contribution of energy to every category is superior. In respect to the contributions of triple cation perovskite, contribution of both perovskite reagents and solvents is little.

2.2 Sensitivity analysis

2.2.1 Chlorobenzene

Chlorobenzene mass is constant for multiple cation/anion perovskite compositions, because its amount does not depend on the composition. From the analysis in Figure 3 from the manuscript, its impact is notable in respect to the amounts of the rest of solvents, as chlorobenzene mass, which is one order of magnitude higher, added during the spin-coating step is not optimised. Furthermore, its use is specific of the anti-solvent method. Therefore, three scenarios were established to analyse the consequences of the usage of chlorobenzene for a proper removal of solvents during the spin-coating step. The three scenarios comprise a reference scenario where the amount of chlorobenzene used is not modified (100% scenario), a scenario where the amount of chlorobenzene is reduced to a 50% (50% scenario), and a scenario where the amount of chlorobenzene is completely eliminated (0% scenario). The purpose of the 50% scenario is to provide information about the benefits of reducing the dose of chlorobenzene used. At the same time, the purpose of the 0% scenario is to display the benefits of the conventional deposition method in comparison to the anti-solvent method, as the main difference of both methods is the usage of chlorobenzene to assist the removal of solvents. These three scenarios impact are contrasted with the impact of the canonical perovskite, thus showing if improvements in multiple cation/anion perovskite impact lead to their fall below those of canonical perovskite. In Figure S2, the total impact of the perovskites for each scenario is shown divided by the total impact of the most detrimental scenario for the most determining categories for PSCs. As the most adverse outcomes belong to the perovskite without caesium content, only results of this perovskite are treated in this analysis.

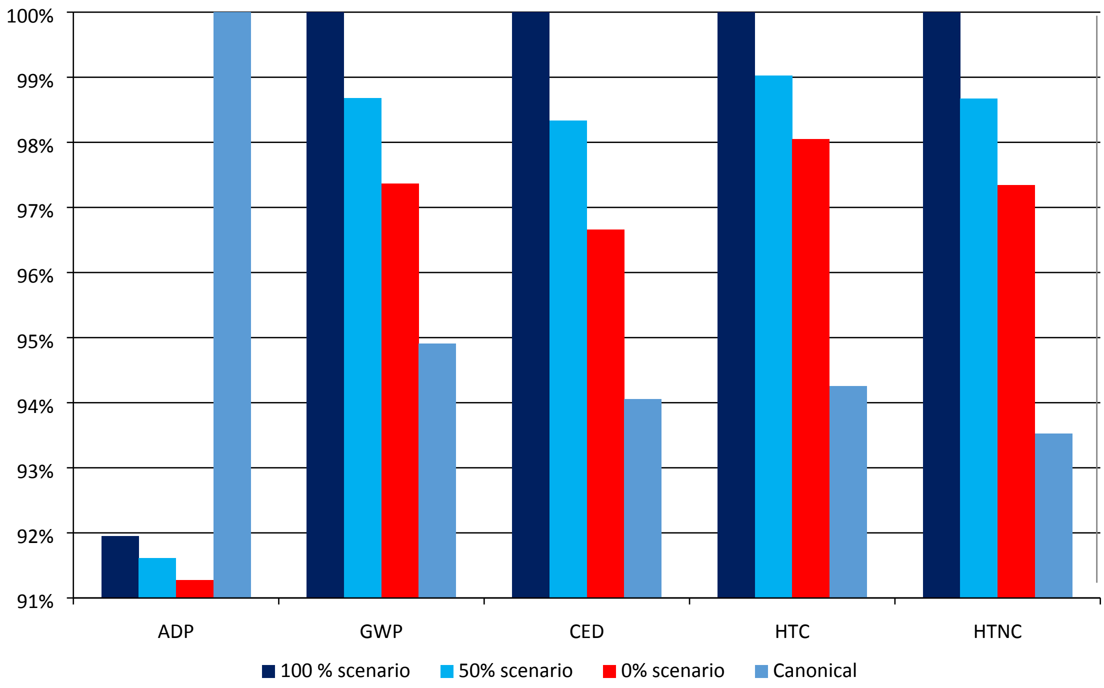

Figure S2. Sensitivity analysis of chlorobenzene with three scenarios for the most determining impact categories for PSCs with anti-solvent method.

From the outcomes of the sensitivity analysis in Figure S2, a general reduction of the impacts is clearly observed. However, the most favourable scenario, which is always the 0% scenario, never declines below 96%. Except for ADP category, impacts for 0% scenario never lessen below the impacts of the canonical perovskite. Hence, this reduction does not suppose a relevant achievement. For ADP category multiple cation/anion perovskite impact is already below canonical perovskite impact. However, the reduction in impact respect to the canonical perovskite impact is trivial.

2.2.2 Anti-solvent method analysis

Herein, a modest anti-solvent method analysis is developed in order to disclose the most environmental damaging compounds used to improve crystallisation, out of the most used to date. This analysis also allows perceiving if a significant change in the overall impact could be achieved with if another compound were dropped during perovskite deposition. According to the procedure described in section 2.4 of the manuscript, the same volume (1 µl) of compound is assumed. Multiplying this volume by the density of each compound their mass is obtained.

Therefore, relative impacts estimated for chlorobenzene (CB), diethyl ether (DE), xylene (XYL), toluene (TL), ethyl acetate (EA) and dichloromethane (DCM) are displayed in percentages in Figure S3. In this analysis all the categories considered in the study are included.

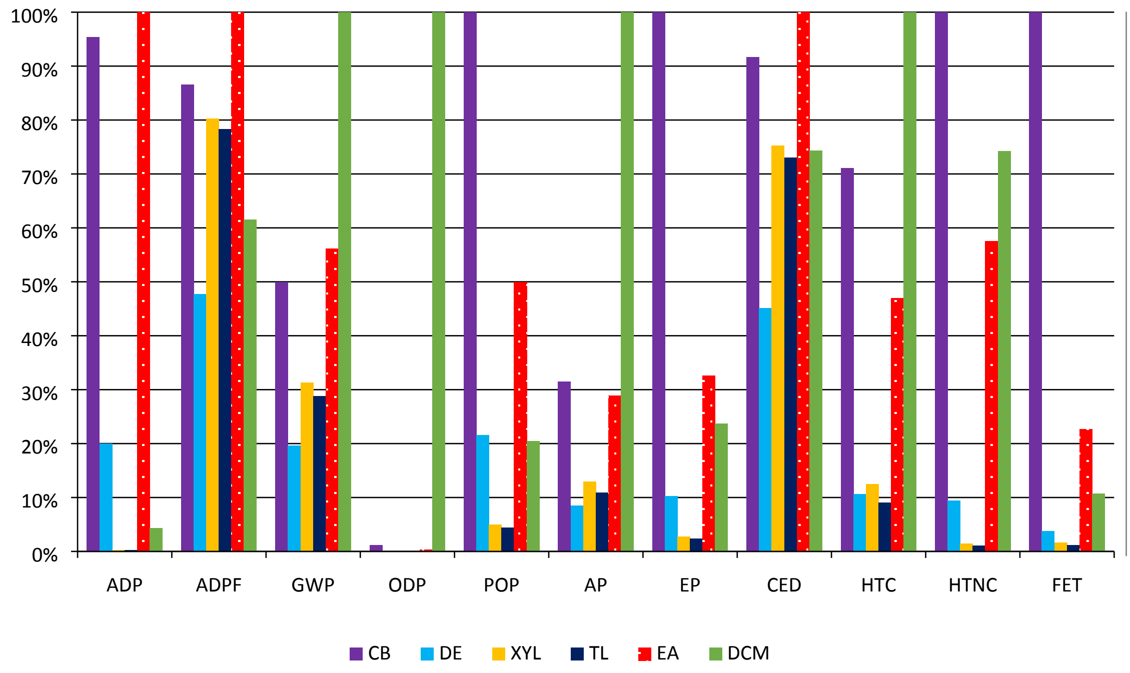

Figure S3. Relative impacts of the compounds used for the anti-solvent method considering the same volume deposited. Chemicals analysed are chlorobenzene (CB), diethyl ether (DE), xylene (XYL), toluene (TL), ethyl acetate (EA) and dichloromethane (DCM).

From the outcomes represented in Figure S3 EA, DCM and CB emerge as the most detrimental compounds. EA is the most harmful compound in ADP, ADPF and CED. DCM arises as the most adverse compound for GWP, ODP, AP and HTC. Meanwhile, CB results the most detrimental compound for POP, EP HTC and FET. Furthermore, CB is the second most harmful compound in ADP, ADPF and CED by roughly the 90% of the most harmful. In a nutshell, the fact that DL, TL and XYL are the most environmentally-safe compounds can be extracted from this analysis.

From Figure S2, it can be deduced that results cannot change for GWP, CED, HTC and HTNC by removing the usage of the compound used for the anti-solvent method. Therefore, it cannot be changed either by choosing another compound regardless of the reduction in the impact the new compound produces. However, for ADP category EA impact is slightly higher than that of CB. Hence, usage of EA instead of CB could pose that the impact of the former exceeds that of the latter. Impact of multiple cation/anion perovskite with x = 0 is analysed to solve this question, as it is the multication perovskite with the highest ADP impact. ADP impact result of this perovskite if EA were used instead of CB is depicted together with that of canonical perovskite in Table S14.

Table S14 Impact results of multication perovskite using EA in the anti-solvent method instead of CB together with impact result of canonical perovskite.

	ADP (kg Sb eq)
x = 0 (with EA)	$4.10 \cdot 10^{-9}$
Canonical	$4.17 \cdot 10^{-9}$

Results in Table S14 reveal that impact of the multiple cation/anion perovskite without caesium and using EA in the anti-solvent method does not surpass that of canonical perovskite. However, it gets so close that if the volume of the drop utilised were increased to 1.2 µl then its impact would be higher than that of canonical perovskite.

References

1 M. Saliba, T. Matsui, J.-Y. Seo, K. Domanski, J.-P. Correa-Baena, M. K. Nazeeruddin, S. M. Zakeeruddin, W. Tress, A. Abate, A. Hagfeldt and M. Grätzel, *Energy Environ. Sci.*, 2016, **9**, 1989–1997.

2 J. H. Noh, N. J. Jeon, Y. C. Choi, M. K. Nazeeruddin, M. Grätzel and S. Il Seok, *J. Mater. Chem. A*, 2013, **1**, 11842.

3 R. Frischknecht, N. Jungbluth, H. J. Althaus, G. Doka, R. Dones, T. Heck, S. Hellweg, R. Hischier, T. Nemecek, G. Rebitzer and M. Spielmann, *Int. J. Life Cycle Assess.*, 2005, 10, 3–9.

4 US6436879B1, 2002, 1–12.

5 F. Eloy, R. Lenaers and C. Moussebois, *Helv. Chim. Acta*, 1962, **45**, 437–441.

6 W. Lossen and P. Schifferdecker, *Justus Liebig's Ann. der Chemie und Pharm.*, 1873, **166**, 295–320.

7 W. S. Yang, J. H. Noh, N. J. Jeon, Y. C. Kim, S. Ryu, J. Seo and S. I. Seok, *Science*, 2015, **348**, 1234–1237.

8 H. Cho, S.-H. Jeong, M.-H. Park, Y.-H. Kim, C. Wolf, C.-L. Lee, J. H. Heo, A. Sadhanala, N. Myoung, S. Yoo, S. H. Im, R. H. Friend and T.-W. Lee, *Science*, 2015, **350**, 1222–1225.

9 S. Ahmad and G. Vijaya Prakash, in *International Conference on Fibre Optics and Photonics*, OSA, Washington, D.C., 2012, p. MPo.40.