Electronic Supplementary Information

A life cycle approach to solvent design: Challenges and opportunities for ionic liquids – application to CO_2 capture

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S1. Consumption and degradation calculations for ionic liquid [Bmim][OAc]

Degradation of [Bmim][OAc] at 71 °C, based on the operating conditions

Assumptions:

- 46,900 tonnes of CO_2 capture per year
- 18.9 m^3 of [Bmim][OAc] as initial charge in the absorber
- IL operating temperature = 71 °C
- Plant service life of 25 years

Based on the decomposition rate reported in,[1] [Bmim][OAc] has a service lifetime of 7500 hours, when operating at 71 °C:

$$\frac{75\ hr}{1\ \%} \times 100\% = 7500\ hr\ service\ lifetime$$

$$\frac{8000\ hr/y}{7500\ hr} = 1.06\ replacements\ per\ year$$

$$25\ year\ service\ life \times 1.06\ replacements\ per\ year = 26.7\ replacements$$

According to Shiflett et al.[2], around 18.9 m^3 or 19,992 kg of [Bmim][OAc] are required for the initial charge of the absorption column, assuming a density of 1057.8 kg m^{-3}.[3]

$$18.9\ m^3\ IL \times 1{,}057.8\ \frac{kg}{m^3} = 19{,}992\ kg\ IL$$

$$\frac{19{,}992\ kg}{46{,}900\ \frac{t\ CO2}{y} \times 25\ y} = 0.017\ \frac{kg\ IL}{t\ CO2}$$

$$0.017\ \frac{kg\ IL}{t\ CO2} \times 26.7\ replacements = 0.455\ \frac{kg\ IL}{t\ CO2}$$

$$0.455\ \frac{kg\ IL}{t\ CO2} \times 0.782\ t\ CO2\ per\ MWh = 0.356\ kg\ IL$$

S2. Estimation of raw material requirements for the production of [Bmim][OAc]

This section details the estimations of the quantities of the raw materials used for the synthesis [Bmim][OAc] shown in Table 8 and Table 9 in the paper. The raw materials and their molecular weights are listed in Table S7.

1-butyl-3-methylimidazolium acetate

The calculations for the production of 1000 g of [Bmim][OAc] are shown below:

$1000\ \text{g}\ C_{10}H_{18}N_2O_2 \div 198.26\ \frac{g}{mol} = 5\ \text{moles}\ C_{10}H_{18}N_2O_2$

$$5\ C_8H_{16}N_2O + 5\ C_2H_4O_2 \rightarrow 5\ \mathbf{C_{10}H_{18}N_2O_2} + 5\ H_2O$$

therefore:

$5\ \text{moles}\ C_8H_{16}N_2O\ \text{x}\ 156.23\ \frac{g}{mol} = 788\ \text{g}\ C_8H_{16}N_2O$

$5\ \text{moles}\ C_2H_4O_2\ \text{x}\ 60.05\ \frac{g}{mol} = 303\ \text{g}\ C_2H_4O_2$

$5\ \text{moles}\ H_2O\ \text{x}\ 18.02\ \frac{g}{mol} = 91\ \text{g}\ H_2O$

1-butyl-3-methylimidazolium hydroxide

The calculations for the production of 788 g of $[Bmim]OH^-$ are as follows:

$$5\ C_8H_{16}N_2O_4S + 5\ Ca(OH)_2 \rightarrow 5\ \mathbf{C_8H_{16}N_2O} + 5\ CaSO_4 + 5\ H_2O$$

where:

$5\ \text{moles}\ C_8H_{16}N_2O_4S\ \text{x}\ 236.3\ \frac{g}{mol} = 1{,}192\ \text{g}\ C_8H_{16}N_2O_4S$

$5\ \text{moles}\ Ca(OH)_2\ \text{x}\ 74\ \frac{g}{mol} = 373\ \text{g}\ Ca(OH)_2$

$5\ \text{moles}\ C_8H_{16}N_2O\ \text{x}\ 156.23\ \frac{g}{mol} = 788\ \text{g}\ C_8H_{16}N_2O$

$5\ \text{moles}\ CaSO_4\ \text{x}\ 136\ \frac{g}{mol} = 686\ \text{g}\ CaSO_4$

$5\ \text{moles}\ H_2O\ \text{x}\ 18.02\ \frac{g}{mol} = 91\ \text{g}\ H_2O$

1-butyl-3-methylimidazolium hydrogen sulfate

The calculations for the production of 1,192 g of $[Bmim][HSO_4]$ are as follows:

$$5\ C_9H_{18}N_2O_4S + 5\ H_2O \rightarrow 5\ \mathbf{C_8H_{16}N_2O_4S} + 5\ CH_3OH$$

hence:

$5\ \text{moles}\ C_9H_{18}N_2O_4S\ \text{x}\ 250.32\ \frac{g}{mol} = 1{,}263\ \text{g}\ C_9H_{18}N_2O_4S$

$5\ \text{moles}\ H_2O\ \text{x}\ 18.02\ \frac{g}{mol} = 91\ \text{g}\ H_2O$

$5\ \text{moles}\ C_8H_{16}N_2O_4S\ \text{x}\ 236.3\ \frac{g}{mol} = 1{,}192\ \text{g}\ C_8H_{16}N_2O_4S$

$5\ \text{moles}\ CH_3OH\ \text{x}\ 32\ \frac{g}{mol} = 162\ \text{g}\ CH_3OH$

1-buty-3-methylimidazolium methyl sulfate

1,263 g (5 moles) of [Bmim][$MeSO_4$] is produced according to the following reaction:

$$5\ C_7H_{12}N_2 + 5\ C_2H_6O_4S \rightarrow 5\ \mathbf{C_9H_{18}N_2O_4S}$$

thus:

$5\ \text{moles}\ C_7H_{12}N_2 \times 124.18\ \frac{g}{mol} = 626\ \text{g}\ C_7H_{12}N_2$

$5\ \text{moles}\ C_2H_6O_4S \times 126.13\ \frac{g}{mol} = 636\ \text{g}\ C_2H_6O_4S$

$5\ \text{moles}\ C_9H_{18}N_2O_4S \times 250.32\ \frac{g}{mol} = 1{,}263\ \text{g}\ C_9H_{18}N_2O_4S$

1-butylimidazole

The calculations for the production of 626 g (5 moles) of 1-butylimidazole are shown below:

$$5\ C_3H_4N_2 + 5\ C_4H_{10}O \rightarrow 5\ \mathbf{C_7H_{12}N_2} + 5\ H_2O$$

therefore:

$5\ \text{moles}\ C_3H_4N_2 \times 68.08\frac{g}{mol} = 343\ \text{g}\ C_3H_4N_2$

$5\ \text{moles}\ C_4H_{10}O \times 74.12\frac{g}{mol} = 374\ \text{g}\ C_4H_{10}O$

$5\ \text{moles}\ C_7H_{12}N_2 \times 124.18\ \frac{g}{mol} = 626\ \text{g}\ C_7H_{12}N_2$

$5\ \text{moles}\ H_2O \times 18.02\ \frac{g}{mol} = 91\ \text{g}\ H_2O$

1-imidazole

The production of 343 g (5 moles) of 1-imidazole proceeds according to the following Debus-Radziszewski reaction for imidazole synthesis:[4, 5]

$$5\ C_2H_2O_2 + 10\ NH_3 + 5\ CH_2O \rightarrow 5\ \mathbf{C_3H_4N_2} + 15\ H_2O$$

where:

$5\ \text{moles}\ C_2H_2O_2 \times 58.04\ \frac{g}{mol} = 293\ \text{g}\ C_2H_2O_2$

$10\ \text{moles}\ NH_3 \times 17.03\ \frac{g}{mol} = 171\ \text{g}\ NH_3$

$5\ \text{moles}\ CH_2O \times 30.03\ \frac{g}{mol} = 151\ \text{g}\ CH_2O_2$

$5\ \text{moles}\ C_3H_4N_2 \times 68.08\frac{g}{mol} = 343\ \text{g}\ C_3H_4N_2$

$15\ \text{moles}\ H_2O \times 18.02\ \frac{g}{mol} = 273\ \text{g}\ H_2O$

S3. Estimation of energy requirements

The energy requirements for the production of [Bmim][OAc] and all the precursors were estimated using the heat of formation of the reactants and products, according to the following equations:

$Q - W = \Delta H + \Delta Ek + \Delta Ep$ (S1)

where:
Q = reactor heat consumption
W = work
ΔH = enthalpy of reaction
ΔE_k = kinetic energy
ΔE_p = potential energy

$$\Delta H = \sum(n \times \hat{H})_{\text{output streams}} - \sum(n \times \hat{H})_{\text{input streams}} \quad \text{(S2)}$$

where:
n = molecular weight of reactants
$\hat{H}$ = specific enthalpy of reactants

$$\hat{H} = \Delta\hat{H}f° + \int_{T1}^{T2} Cp\, dT \quad \text{(S3)}$$

where:
$\Delta\hat{H}f$ °= heat of formation of reactants
Cp = calorific value of reactants
T1 = reference temperature (25 ºC)
T2 = temperature of the reactants

The estimated theoretical energy consumption was scaled-up to industrial scale by multiplying it by the correction factors reported in literature.[6] For heat requirements for endothermic reactions, the estimated theoretical value was converted to the actual heat consumption (assumed to be supplied by natural gas) using a correction factor of 4.2. Similarly, the theoretical heat generated by exothermic reactions was converted to the actual cooling electricity requirements using a correction factor of 3.2.

The physico-chemical properties of the chemicals involved in the synthesis of [Bmim][OAc] and its precursors are summarised in Table S1.

1-butyl-3-methylimidazolium acetate

The energy inventory for the production of 1000 g of [Bmim]oac has been estimated based on the heat requirements of the reactor using eqns. (S1)–(S3). Table S1 shows the input and output streams involved in the synthesis of [Bmim]oac based on:

$$5\ C_8H_{16}N_2O + 5\ C_2H_4O_2 \rightarrow 5\ \mathbf{C_{10}H_{18}N_2O_2} + 5\ H_2O$$

The specific enthalpies $\hat{H}$ for each chemical compound in the reaction were estimated according to eqn. (3) as follows:

$$\hat{H}1 = \Delta\hat{H}f°\ C_8H_{16}N_2O\ (25°C) + \int_{25\,°C}^{25\,°C} Cp\, dT = -180\,\frac{kJ}{mol} + 0 = -180\,\frac{kJ}{mol}$$

$$\hat{H}2 = \Delta\hat{H}f°\ C_2H_4O_2\ (25\,°C) + \int_{25\,°C}^{25\,°C} Cp\, dT = -484.04\,\frac{kJ}{mol} + 0 = -484.04\,\frac{kJ}{mol}$$

$$\hat{H}3 = \Delta\hat{H}f°\ C_{10}H_{18}N_2O_2\ (25\,°C) + \int_{25\,°C}^{25\,°C} Cp\, dT = -180\,\frac{kJ}{mol} + 0 = -180\,\frac{kJ}{mol}$$

$$\hat{H}4 = \Delta\hat{H}f°\ H_2O\ (25\,°C) + \int_{25\,°C}^{25\,°C} Cp\, dT = -285.83\,\frac{kJ}{mol} + 0 = -285.83\,\frac{kJ}{mol}$$

The enthalpy of the reaction was calculated using eqn. (S2) based on the specific enthalpy values $\hat{H}$ estimated for each chemical compound involved in the reaction of the synthesis of [Bmim]oac as shown in **Error! Reference source not found.**. Hence:

$$\Delta H = \left[\left(5 \text{ mol x } -180 \frac{kJ}{mol}\right) + \left(5 \text{ mol x } -285.83 \frac{kJ}{mol}\right)\right] - \left[\left(5 \text{ mol x } -180 \frac{kJ}{mol}\right) + \left(5 \text{ mol x } -484.04 \frac{kJ}{mol}\right)\right]$$
$$= 1733 \text{ kJ}$$

According to eqn. (S1), the heat required by the reactor is equal to the enthalpy of reaction assuming that no work is carried and that the kinetic and potential energy are zero. A negative ΔH value indicates that an exothermic reaction is taking place and therefore heat needs to be removed through cooling. Therefore, the theoretical heat needed to be removed from the reactor is equal to:

$Q \approx \Delta H = 1733 \text{ kJ} = 1.7 \text{ MJ} \approx 2 \text{ MJ}$
Using the correction factor 4.2, this heat is equivalent to 7.3 MJ of actual heat needed to heat the reactor using natural gas.

Table S1 Input and output streams for the production of [Bmim][OAc]

Chemical compound	n input (mol)	$\hat{H}$ input (kJ/mol)	n output (mol)	$\hat{H}$ output (kJ/mol)
$C_8H_{16}N_2O$	5	$\hat{H}1$	--	--
$C_2H_4O_2$	5	$\hat{H}2$	--	--
$C_{10}H_{18}N_2O_2$	--	--	5	$\hat{H}3$
H_2O	--	--	5	$\hat{H}4$

1-butyl-3-methylimidazolium hydroxide

Table S2 shows the input and output streams involved in the synthesis of 788 g of $[Bmim]OH^-$ based on the reaction below:

$$5\ C_8H_{16}N_2O_4S + 5\ Ca(OH)_2 \rightarrow 5\ \mathbf{C_8H_{16}N_2O} + 5\ CaSO_4 + 5\ H_2O$$

The specific enthalpies $\hat{H}$ for the compounds involved in the synthesis of $[Bmim]OH^-$ were estimated according to eqn. (S3):

$$\hat{H}1 = \Delta\hat{H}f^\circ\ C_8H_{16}N_2O_4S\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -584.3 \frac{kJ}{mol} + 0 = -584.3 \frac{kJ}{mol}$$

$$\hat{H}2 = \Delta\hat{H}f^\circ\ Ca(OH)_2\ (25^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -610.76 \frac{kJ}{mol} + 0 = -610.76 \frac{kJ}{mol}$$

$$\hat{H}3 = \Delta\hat{H}f^\circ\ C_8H_{16}N_2O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -180 \frac{kJ}{mol} + 0 = -180 \frac{kJ}{mol}$$

$$\hat{H}4 = \Delta\hat{H}f^\circ\ CaSO_4\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -1435.1 + 0 = -1435.1 \frac{kJ}{mol}$$

$$\hat{H}5 = \Delta\hat{H}f^\circ\ H_2O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -285.83 + 0 = -285.83 \frac{kJ}{mol}$$

The enthalpy of the reaction, calculated using eqn. (S2) is thus equal to:

$$\Delta H = \left[\left(5 \text{ mol x } -180 \frac{kJ}{mol}\right) + \left(5 \text{ mol x } -1435.1 \frac{kJ}{mol}\right) + \left(5 \text{ mol x } -285.83 \frac{kJ}{mol}\right)\right] - \left[\left(5 \text{ mol x } -584 \frac{kJ}{mol}\right) + \left(5 \text{ mol x } -610.76 \frac{kJ}{mol}\right)\right]$$
$$= 2248 \text{ kJ}$$

Therefore, making the same assumptions in eqn. (S1) as for the previous precursors, the theoretical heat needed to be added to the reactor is equal to 2.2 MJ. Applying the correction factor of 4.2, this heat is equivalent to 9.4 MJ of heat produced from natural gas to heat the reactor.

Table S2 Input and output streams for the production of $[Bmim]OH^-$

Chemical compound	n input (mol)	$\hat{H}$ input (kJ/mol)	n output (mol)	$\hat{H}$ output (kJ/mol)
$C_8H_{16}N_2O_4S$	5	$\hat{H}1$	--	--
$Ca(OH)_2$	5	$\hat{H}2$	--	--
$C_8H_{16}N_2O$	--	--	5	$\hat{H}3$
$CaSO_4$			5	$\hat{H}4$
H_2O	--	--	5	$\hat{H}5$

1-butyl-3-methylimidazolium hydrogen sulfate

Table S3 shows the input and output streams involved in the synthesis of 1,192 g of $[Bmim][HSO_4]$ based on the following reaction:

$$5\ C_9H_{18}N_2O_4S + 5\ H_2O \rightarrow 5\ \mathbf{C_8H_{16}N_2O_4S} + 5\ CH_4O$$

The specific enthalpies $\hat{H}$ for the compounds involved in the synthesis of $[Bmim][HSO_4]$ were estimated according to eqn. (S3):

$$\hat{H}1 = \Delta\hat{H}f^\circ\ C_9H_{18}N_2O_4S\ (25\ °C) + \int_{25\ °C}^{25\ °C} Cp\ dT = -584.3\ \frac{kJ}{mol} + 0 = -584.3\ \frac{kJ}{mol}$$

$$\hat{H}2 = \Delta\hat{H}f^\circ\ H_2O\ (25\ °C) + \int_{25\ °C}^{25\ °C} Cp\ dT = -285.83\ \frac{kJ}{mol} + 0 = -285.83\ \frac{kJ}{mol}$$

$$\hat{H}3 = \Delta\hat{H}f^\circ\ C_8H_{16}N_2O_4S\ (25\ °C) + \int_{25\ °C}^{25\ °C} Cp\ dT = -584.3\ \frac{kJ}{mol} + 0 = -584.3\ \frac{kJ}{mol}$$

$$\hat{H}4 = \Delta\hat{H}f^\circ\ CH_4O\ (25\ °C) + \int_{25\ °C}^{25\ °C} Cp\ dT = -238.93\ \frac{kJ}{mol} + 0 = -238.93\ \frac{kJ}{mol}$$

The enthalpy of the reaction, calculated using eqn. (S2) is thus equal to:

$$\Delta H = \left[\left(5\ mol\ x\ -584.3\ \frac{kJ}{mol}\right) + \left(5\ mol\ x\ -238.93\ \frac{kJ}{mol}\right)\right] - \left[\left(5\ mol\ x\ -584.3\ \frac{kJ}{mol}\right) + \left(5\ mol\ x\ -285.83\ \frac{kJ}{mol}\right)\right]$$
$$= 2604\ kJ$$

Therefore, making the same assumptions in eqn. (S1) as for the previous precursors, the theoretical heat needed in the reactor is equal to 2.6 MJ. Applying the correction factor of 4.2, this heat is equivalent to 10.9 MJ of heat from natural gas.

Table S3 Input and output streams for the production of $[Bmim][HSO_4]$

Chemical compound	n input (mol)	$\hat{H}$ input (kJ/mol)	n output (mol)	$\hat{H}$ output (kJ/mol)
$C_9H_{18}N_2O_4S$	5	$\hat{H}1$	--	--
H_2O	5	$\hat{H}2$	--	--
$C_8H_{16}N_2O_4S$	--	--	5	$\hat{H}3$
CH_4O	--	--	5	$\hat{H}4$

1-butyl-3-methylimidazolium methyl sulfate

The input and output streams involved in the synthesis of 1,263 g of $[Bmim][MeSO_4]$ are summarised in Table S4. The synthesis is carried out according to:

$$5\ C_7H_{12}N_2 + 5\ C_2H_6O_4S \rightarrow 5\ \mathbf{C_9H_{18}N_2O_4S}$$

The specific enthalpies were estimated as follows:

$$\hat{H}1 = \Delta\hat{H}f^\circ\ C_7H_{12}N_2\ (25\ °C) + \int_{25\ °C}^{25\ °C} Cp\ dT = 58.9\ \frac{kJ}{mol} + 0 = 58.9\ \frac{kJ}{mol}$$

$\hat{H}2 = \Delta\hat{H}f^\circ\ C_2H_6O_4S\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -735.25\ \frac{kJ}{mol} + 0 = -735.25\ \frac{kJ}{mol}$

$\hat{H}3 = \Delta\hat{H}f^\circ\ C_9H_{18}N_2O_4S\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -584.3\ \frac{kJ}{mol} + 0 = -584.3\ \frac{kJ}{mol}$

The total enthalpy is equal to:

$$\Delta H = \left[\left(5\ mol\ x\ -584.3\ \frac{kJ}{mol}\right)\right] - \left[\left(5\ mol\ x\ 58.9\ \frac{kJ}{mol}\right) + \left(10\ mol\ x\ -735.25\ \frac{kJ}{mol}\right)\right]$$

= 464 kJ

Making the same assumptions as for the other precursors, the total theoretical heating energy is equal to 0.5 MJ, so that the actual heat demand supplied by natural gas is estimated at 2 MJ.

Table S4 Input and output streams for the production of [Bmim][$MeSO_4$]

Chemical compound	n input (mol)	$\hat{H}$ input (kJ/mol)	n output (mol)	$\hat{H}$ output (kJ/mol)
$C_7H_{12}N_2$	5	$\hat{H}1$	--	--
$C_2H_6O_4S$	10	$\hat{H}2$	--	--
$C_9H_{18}N_2O_4S$	--	--	5	$\hat{H}3$

1-buthylimidazole

Table S5 shows the input and output streams involved in the synthesis of 626 g of 1-buthylimidazole based on the following reaction:

$$5\ C_3H_4N_2 + 5\ C_4H_{10}O \rightarrow 5\ \mathbf{C_7H_{12}N_2} + 5\ H_2O$$

The specific enthalpies were estimated as follows:

$\hat{H}1 = \Delta\hat{H}f^\circ\ C_3H_4N_2\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = 54.3\ \frac{kJ}{mol} + 0 = 54.3\ \frac{kJ}{mol}$

$\hat{H}2 = \Delta\hat{H}f^\circ\ C_4H_{10}O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = 328.3\ \frac{kJ}{mol} + 0 = 328.3\ \frac{kJ}{mol}$

$\hat{H}3 = \Delta\hat{H}f^\circ\ C_7H_{12}N_2\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = 58.9\ \frac{kJ}{mol} + 0 = 58.9\ \frac{kJ}{mol}$

$\hat{H}4 = \Delta\hat{H}f^\circ\ H_2O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -285.83\ \frac{kJ}{mol} + 0 = -285.83\ \frac{kJ}{mol}$

The total enthalpy is equal to:

$$\Delta H = \left[\left(5\ mol\ x\ 58.9\ \frac{kJ}{mol}\right) + \left(5\ mol\ x\ -285.83\ \frac{kJ}{mol}\right)\right] - \left[\left(5\ mol\ x\ 54.3\ \frac{kJ}{mol}\right) + \left(5\ mol\ x\ -328.3\ \frac{kJ}{mol}\right)\right]$$

= -3075 kJ

Therefore, making the same assumptions in eqn. (S1) as for the previous precursors, the total theoretical electricity needed to cool the reactor is equal to 3.1 MJ. Applying the correction factor of 3.2, the actual electricity needed to remove heat from the reactor is estimated at 9.8 MJ.

Table S5 Input and output streams for the production of 1-buthylimidazole

Chemical compound	n input (mol)	$\hat{H}$ input (kJ/mol)	n output (mol)	$\hat{H}$ output (kJ/mol)
$C_3H_4N_2$	5	$\hat{H}1$	--	--
$C_4H_{10}O$	5	$\hat{H}2$	--	--

$C_7H_{12}N_2$	--	--	5	$\hat{H}3$
H_2O	--	--	5	$\hat{H}4$

1-imidazole

The energy inventory for the production of 343 g of 1-imidazole has been estimated based on the heat requirements of the reactor using eqns. (S1)–(S3). Table S6 lists the input and output streams involved in the reaction shown below:

$$5\ C_2H_2O_2 + 10\ NH_3 + 5\ CH_2O \rightarrow 5\ \mathbf{C_3H_4N_2} + 15\ H_2O$$

The specific enthalpies were estimated as follows:

$$\hat{H}1 = \Delta\hat{H}f^\circ\ C_2H_2O_2\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -212\ \frac{kJ}{mol} + 0 = -212\ \frac{kJ}{mol}$$

$$\hat{H}2 = \Delta\hat{H}f^\circ\ NH_3\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -45.92\ \frac{kJ}{mol} + 0 = -45.92\ \frac{kJ}{mol}$$

$$\hat{H}3 = \Delta\hat{H}f^\circ\ CH_2O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -112.25\ \frac{kJ}{mol} + 0 = -112.25\ \frac{kJ}{mol}$$

$$\hat{H}4 = \Delta\hat{H}f^\circ\ C_3H_4N_2\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = 54.3\ \frac{kJ}{mol} + 0 = 54.3\ \frac{kJ}{mol}$$

$$\hat{H}5 = \Delta\hat{H}f^\circ\ H_2O\ (25\ ^\circ C) + \int_{25\ ^\circ C}^{25\ ^\circ C} Cp\ dT = -285.83\ \frac{kJ}{mol} + 0 = -285.83\ \frac{kJ}{mol}$$

Therefore, the overall enthalpy for the reactions is equal to:

$$\Delta H = \left[\left(5\ mol\ x\ 54.3\ \frac{kJ}{mol}\right) + \left(15\ mol\ x\ -285.83\ \frac{kJ}{mol}\right)\right] - \left[\left(5\ mol\ x\ -212\ \frac{kJ}{mol}\right) + \left(10\ mol\ x\ -45.92\ \frac{kJ}{mol}\right) + \left(5\ mol\ x\ -112.25\ \frac{kJ}{mol}\right)\right]$$

$$= -1{,}952\ kJ$$

Making the same assumptions as for the other precursors, the total theoretical cooling energy is equal to 2 MJ so that the actual electricity demand for cooling is estimated at 6.2 MJ.

Table S6 Input and output streams for the production of 1-imidazole

Chemical compound	**n input (mol)**	**$\hat{H}$ input (kJ/mol)**	**n output (mol)**	**$\hat{H}$ output (kJ/mol)**
$C_2H_2O_2$	5	$\hat{H}1$	--	--
NH_3	10	$\hat{H}2$	--	--
CH_2O	5	$\hat{H}3$	--	--
$C_3H_4N_2$	--	--	5	$\hat{H}4$
H_2O	--	--	15	$\hat{H}5$

Table S7 Physico-chemical properties of chemical compounds used in the synthesis of [Bmim][OAc] and its precursors

Chemical compound	Chemical formula	Molecular weight (g/mol)	Heat of formation, ΔH^{o}_{f} 25ºC (kJ/mol)	Assumptions	Source
1-butyl-3-methylimidazolium acetate	$C_{10}H_{18}N_2O_2$	198.3	-180	ΔHfº of [Bmim][Br] used as proxy.	[7]
1-butyl-3-methylimidazolium hydroxide	$C_8H_{16}N_2O$	156.23	-180	ΔHfº of [Bmim][Br] used as proxy.	[7]
1-butyl-3-methylimidazolium hydrogen sulfate	$C_8H_{16}N_2O_4S$	236.30	-584.3	ΔHfº of ionic liquid [Emim][ethyl sulfate] used as proxy.	[8]
1-butyl-3-methylimidazolium methyl sulfate	$C_9H_{18}N_2O_4S$	250.32	-584.3	ΔHfº of ionic liquid [Emim][ethyl sulfate] used as proxy.	[4]
1-buthylimidazole	$C_7H_{12}N_2$	124.18	58.9		[9]
Acetic acid	$C_2H_4O_2$	60.05	-484.04		-||-
Ammonia	NH_3	17.03	-45.92		-||-
Calcium hydroxide	$Ca(OH)_2$	74.00	-610.76		-||-
Calcium sulfate	$CaSO_4$	136.00	-1435.1		-||-
Dimethyl sulfate	$C_2H_6O_4S$	126.13	-735.25		-||-
Formaldehyde	CH_2O	30.03	-112.25		-||-
Glyoxal	$C_2H_2O_2$	58.04	-212		-||-
Imidazole	$C_3H_4N_2$	68.08	54.3		-||-
Methanol	CH_4O	32.04	-238.93		-||-
Water	H_2O	18.02	-285.83		-||-

S4. Environmental impacts of the UK and average EU-27 electricity mix, used in the study.

Table S7 UK and EU-27 average electricity mix.[10-12]

UK Electricity mix fuel source[10]	2017 average (%)	EU-27 Electricity mix fuel source[11, 12]	2018 average (%)
Coal	6.5	Coal	22.2
Natural gas	39.5	Natural gas	21.2
Oil	0.5	Oil	2.1
Nuclear	19.3	Nuclear	25.8
Hydro	1.8	Hydro	13
Wind and solar	18.4	Wind	11.3
Bioenergy	8.3	Solar	4.1
Pumped storage	-0.4	Geothermal	0.2
Waste & industrial gas	1.6	Other	0.1
Net imports	4.5	--	--

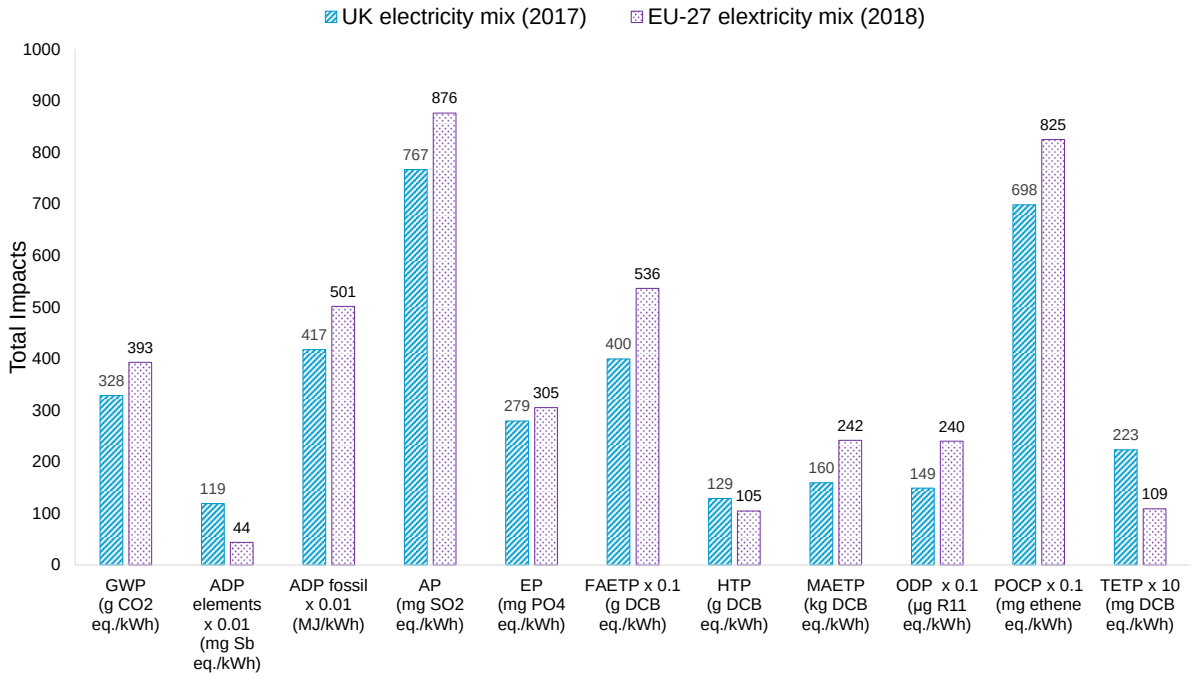

Figure S1 Life cycle environmental impacts of the UK and average EU-27 electricity mix based on average fuel sources reported in the literature. [10-12]

[The values for some impacts have been scaled to fit. The original values can be obtained by multiplying the value shown on top of the bars by the scaling factor given on the x-axis. GWP: global warming potential; ADP elements: abiotic depletion potential of elements; ADP fossil: abiotic depletion potential of fossil resources; AP: acidification potential; EP: eutrophication potential; FAETP: fresh water aquatic ecotoxicity potential; HTP: human toxicity potential; MAETP: marine aquatic ecotoxicity potential; ODP: ozone depletion potential; POCP; photochemical oxidants creation potential; TETP: terrestrial ecotoxicity potential. DCB: dichlorobenzene.]

S5. Life cycle stages contribution to other environmental impacts

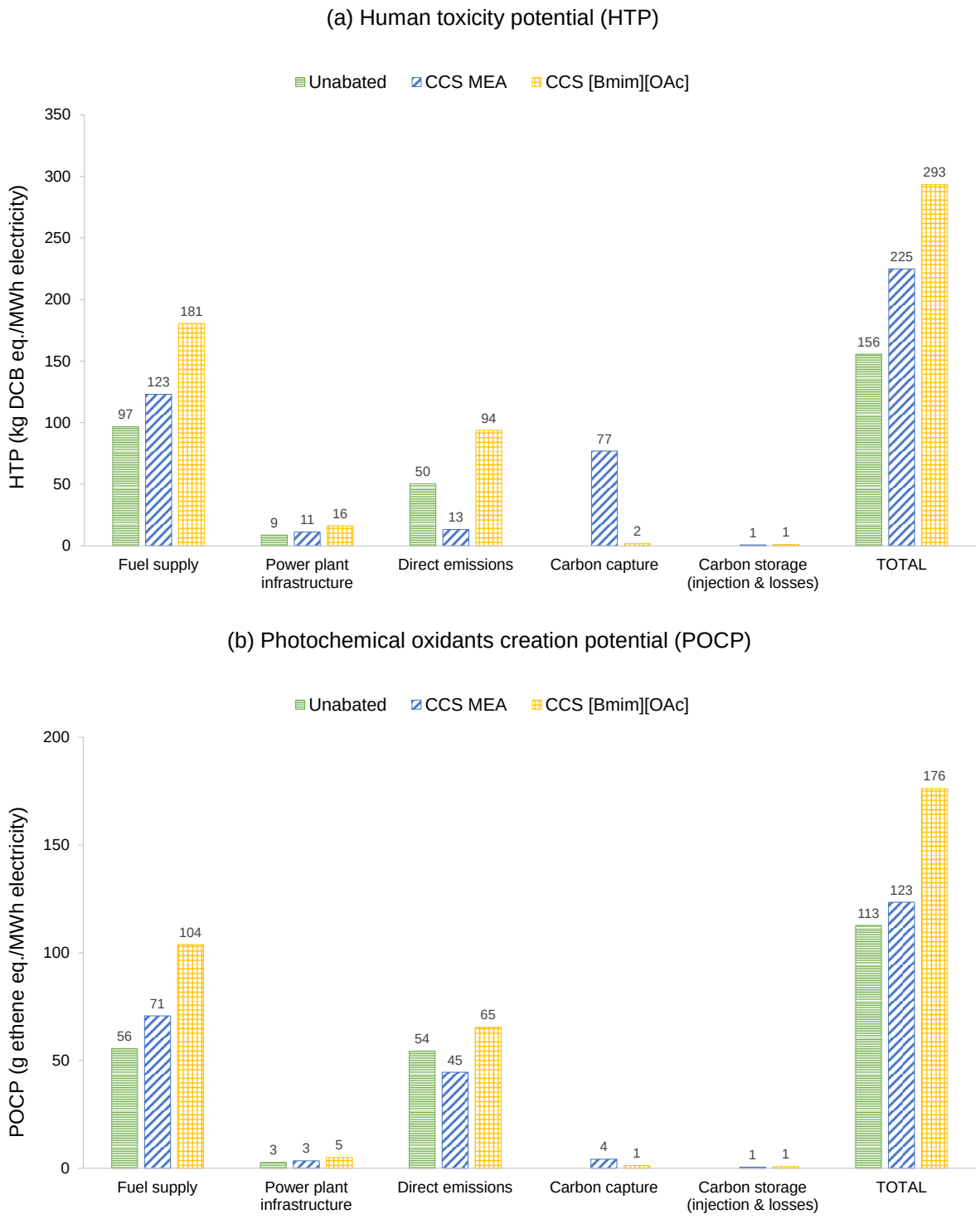

Figure S2 (a) Human toxicity potential, and (b) Photochemical oxidants creation potential of the three power plant systems with contribution from different life cycle stages.

[Coal power plant with carbon capture and storage using monoethanolamine for CO_2 capture (CCS MEA); coal power plant with carbon capture and storage using ionic liquid [Bmim][AOc] for CO_2 capture (CCS [Bmim][AOc]). Carbon storage includes CO_2 injection and CO_2 losses].

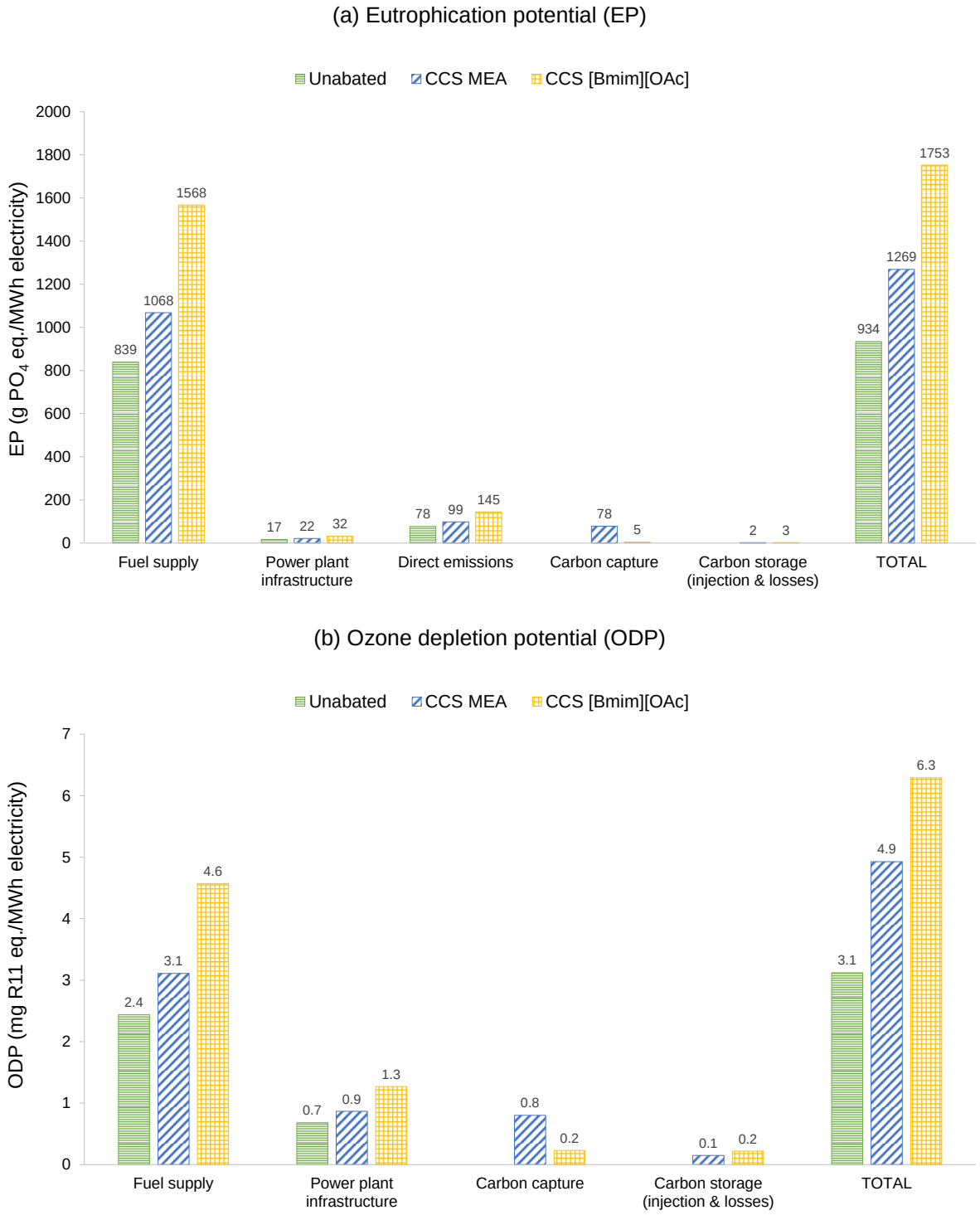

Figure S3 (a) Eutrophication potential, and (b) Ozone depletion potential of the three power plant systems with contribution from different life cycle stages.

[Coal power plant with carbon capture and storage using monoethanolamine for CO_2 capture (CCS MEA); coal power plant with carbon capture and storage using ionic liquid [Bmim][AOc] for CO_2 capture (CCS [Bmim][AOc]). Carbon storage includes CO_2 injection and CO_2 losses].

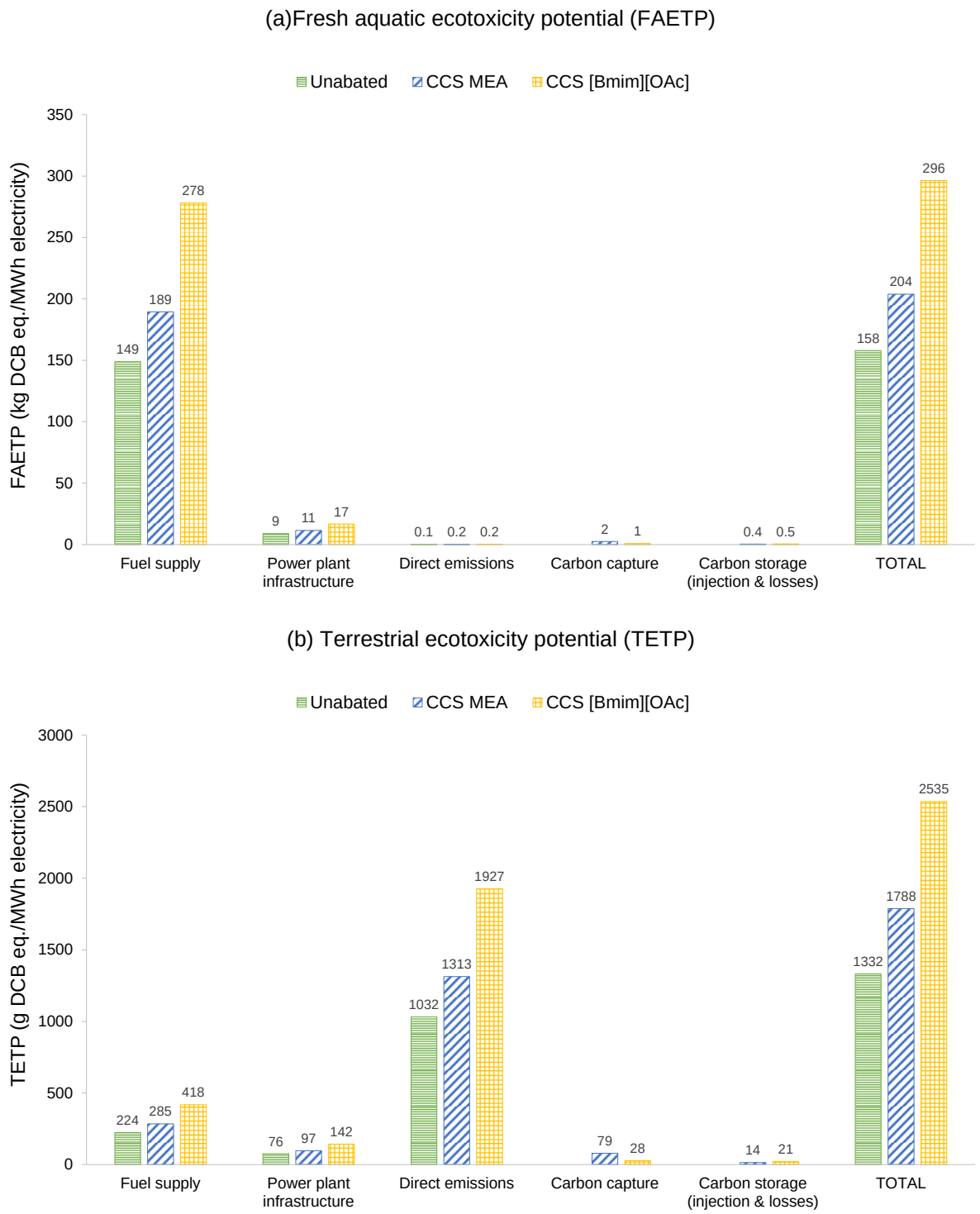

Figure S4 (a) Fresh aquatic ecotoxicity potential, and (b) Terrestrial ecotoxicity potential of the three power plant systems with contribution from different life cycle stages.

[Coal power plant with carbon capture and storage using monoethanolamine for CO_2 capture (CCS MEA); coal power plant with carbon capture and storage using ionic liquid [Bmim][AOc] for CO_2 capture (CCS [Bmim][AOc]). Carbon storage includes CO_2 injection and CO_2 losses].

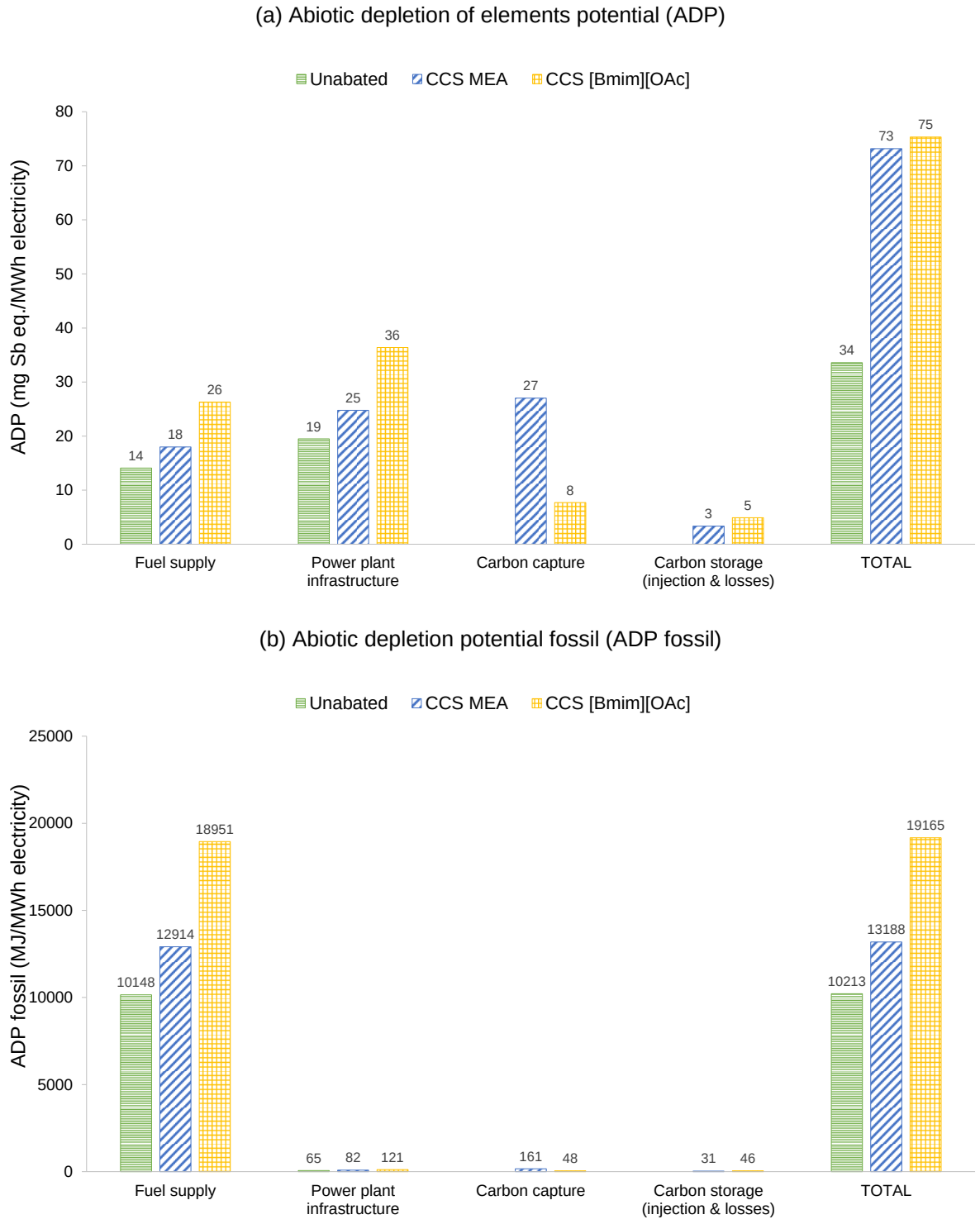

Figure S5 (a) Abiotic depletion of elements potential, and (b) Abiotic depletion potential fossil of the three power plant systems with contribution from different life cycle stages.

[Coal power plant with carbon capture and storage using monoethanolamine for CO_2 capture (CCS MEA); coal power plant with carbon capture and storage using ionic liquid [Bmim][AOc] for CO_2 capture (CCS [Bmim][AOc]). Carbon storage includes CO_2 injection and CO_2 losses].

S6. Contribution analysis of main precursors in the production of [Bmim][OAc]

Table S8 Environmental impacts and contribution analysis for the production of 1.26 kg of [Bmim][$MeSO_4$][a]

Environmental impacts	GWP (kg CO_2 eq.)	ADP elements (mg Sb eq.)	ADP fossil (MJ)	AP (g SO_2 eq.)	EP (g PO_4 eq.)	FAETP (kg DCB eq.)	HTP (kg DCB eq.)	MAETP (t DCB eq.)	ODP (mg R11 eq.)	POCP (g Ethene eq.)	TETP (g DCB eq.)
	4.6	25	94	23	8	1.0	2.6	3	0.39	3	42
Life cycle stages[b]	% Contributions										
1-butylimidazole	77	56	77	41	51	57	60	63	66	67	62
Dimethyl sulfate	18	26	20	56	45	32	26	28	27	30	24
Process energy	2.9	0.0	2.2	0.4	0.2	0.1	0.6	0.1	5.0	0.8	0.2
Chemical plant & transport	1.7	18.1	1.0	2	4	11	13.2	8.5	1.8	1.8	13.7

[a] Required for the production of 1 kg of [Bmim][OAc]. [b] Cradle to gate.

Table S9 Environmental impacts and contribution analysis for the production of 1.19 kg of [Bmim][HSO_4][a]

Environmental impacts	GWP (kg CO_2 eq.)	ADP elements (mg Sb eq.)	ADP fossil (MJ)	AP (g SO_2 eq.)	EP (g PO_4 eq.)	FAETP (kg DCB eq.)	HTP (kg DCB eq.)	MAETP (t DCB eq.)	ODP (mg R11 eq.)	POCP (g Ethene eq.)	TETP (g DCB eq.)
	5.3	29	101	24	9	1	3	3.2	0.48	2.87	47
Life cycle stages[b]	% Contributions										
[Bmim][$MeSO_4$]	85	85	88	96	95	90	86	92	77	94	88
Water	0	0	0	0	0	0	0	0	0	0	0
Process energy	14	0	11	2	1	1	3	1	21	4	1
Chemical plant & transport	1	15	1	2	4	9	11	7	1	2	11

[a] Required for the production of 1 kg of [Bmim][OAc]. [b] Cradle to gate.

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