Selective calcium removal at near-ambient temperature in a multimineral recovery process from seawater reverse osmosis brine and ex-ante life cycle assessment

Supplementary Information

Raffaele Molinari*[1], Ahmet Halil Avci[1], Efrem Curcio[1], David Sanchez Domene[2], Carolina Villa Gonzalez[2], Jose Jorge Espi Gallart[2], , Pietro Argurio[1]

[1] Department of Environmental Engineering (DIAm), University of Calabria, via P. Bucci, cubo 44/A, Rende (CS), Italy.

[2] Eurecat, Centre Tecnològic de Catalunya, Waste, Energy and Environmental Impact, Manresa, Spain

(*) Corresponding author

Raffaele Molinari, Professor.

Mail address: Department of Environmental Engineering (DIAm), University of Calabria, via P. Bucci, cubo 44/A, I-87036 Arcavacata di Rende (CS), Italy.

Tel: +39-0984-496699

Fax: +39-0984-496655

E-mail address: raffaele.molinari@unical.it

Table S1. Impact category indicators use in the life cycle assessment and their acronyms

Environmental indicator	unit	Acronym	Description
Climate Change	kg CO2 eq	CC	The indicator quantifies the radiative forcing as Global Warming Potential (GWP100). Baseline model of the IPCC 2013 + factors adapted from EF guidance.
Ozone depletion	kg CFC11 eq	OD	The indicator quantifies the Ozone Depletion Potential (ODP), which estimate the destructive effects on the stratospheric ozone layer over a time horizon of 100 years.
Ionising radiation	kBq U235 eq	IR	The indicator quantifies the impact of ionizing radiation on the population, in comparison to Uranium 235.
Photochemical ozone formation	kg NMVOC eq	POF	The indicator quantifies the Photochemical ozone creation potential (POCP), which is an expression of the potential contribution to photochemical ozone formation
Particulate matter	disease incidence	PM	The indicator quantifies the disease incidence due to kg of PM2.5 emitted. It is calculated applying the average slope between the Emission Response Function (ERF) working point and the theoretical minimum-risk level. Exposure model based on archetypes that include urban environments, rural environments, and indoor environments within urban and rural areas.
Human toxicity, non-cancer	CTUh (Comparative Toxic Unit for humans)	HTnc	USEtox consensus multimedia model. It spans two spatial scales: continental scale consisting of six compartments (urban air, rural air, agricultural soil natural soil, freshwater, costal marine water), and the global scale with the same structure but without the urban air.
Human toxicity, cancer	CTUh (Comparative Toxic Unit for humans)	HTc	USEtox consensus multimedia model. It spans two spatial scales: continental scale consisting of six compartments (urban air, rural air, agricultural soil natural soil, freshwater, costal marine water), and the global scale with the same structure but without the urban air.
Acidification	mol H+ eq	AP	The indicator quantifies accumulated exceedance characterizing the change in critical load exceedance of the sensitive area in terrestrial and main freshwater ecosystems, to which acidifying substances deposit.
Eutrophication freshwater	kg P eq	EPf	This indicator is an expression of the degree to which the emitted nutrients reaches the freshwater end compartment (phosphorus considered as limiting factor in freshwater).

(continued)

Environmental indicator	unit	Acronym	Description
Eutrophication marine	kg N eq	EPm	This indicator is an expression of the degree to which the emitted nutrients reaches the marine end compartment (nitrogen considered as limiting factor in marine water).
Eutrophication terrestrial	mol N eq	EPt	The indicator quantifies accumulated exceedance characterizing the change in critical load exceedance of the sensitive area, to which eutrophying substances deposit.
Ecotoxicity freshwater	CTUe (Comparative Toxic Unit for ecosystems)	ETf	USEtox consensus multimedia model. It spans two spatial scales: continental scale consisting of six compartments (urban air, rural air, agricultural soil natural soil, freshwater, costal marine water), and the global scale with the same structure but without the urban air.
Land Use	Pt (Dimensionless)	LU	Soil quality index, this indicator was re-Calculated by Joint Research Centre (JRC) starting from LANCA® v2.2 as baseline model for land use.
Water use	m3 world eq	WU	The indicator quantifies the Relative Available WAter REmaining (AWARE) per area in a watershed, after the demand of humans and aquatic ecosystems has been met.
Resource use - fossils	MJ	RUf	The indicator quantifies abiotic resource depletion fossil fuels based on lower heating value. Depletion model based on use-to-availability ratio. Full substitution among fossil energy carriers is assumed.
Resource use - mineral and metals	kg Sb eq	RUe	The indicator quantifies abiotic resource depletion for mineral and metal resources. Depletion model based on use-to-availability ratio.

For further information about the impact category indicators see
https://simapro.com/wp-content/uploads/2022/07/DatabaseManualMethods.pdf
pag 15-19

https://lirias.kuleuven.be/retrieve/355474

Table S2. Life Cycle Inventory (LCI) assumptions and limitations for the near-ambient temperature calcium precipitation system scenario. wh: working hours.

LCI component	Assumptions and limitations	Ecoinvent process / proxy
Thermostat	Model: MAGIO-MX-BC-12 Weight: 4,6Kg Lifespan: 10y*225d*24h=54000wh 0,5h of use considered	Electric kettle {GLO}| market for electric kettle | Cut-off, U
Stirrer	Model: OHS 200 Advance Weight: 14,6Kg Lifespan: 10y*225d*24h=54000wh 3h and 10min of use considered	Electric motor, for electric scooter {GLO}| market for | Cut-off, U
Peristaltic pump	Weight: 5,5Kg Lifespan: 10y*225d*24h=54000wh 10min of use considered	Water pump, 22kW {GLO}| market for water pump, 22kW | Cut-off, U
Tanks	Weight: 3,0Kg Lifespan: 15y*225d*24h=81000wh 4h and 5min of use considered	Polyethylene, high density, granulate {GLO}| market for | Cut-off, U + Blow moulding {GLO}| market for | Cut-off, U
Thermostat	Nominal power: 3,0 kW Power consumption: 3,0*0,8(power consumption factor)*0,5h = 1,3 kWh	Electricity, low voltage {Europe without Switzerland}| market group for | Cut-off, U
$Stirrer_{LOW}$	Power consumption: 0,3 kWh (direct measure, working for 3 hours)	Electricity, low voltage {Europe without Switzerland}| market group for | Cut-off, U
$Stirrer_{HIGH}$	Power consumption: 0,083 kWh (direct measure, working for 10 minutes)	Electricity, low voltage {Europe without Switzerland}| market group for | Cut-off, U
Peristaltic pump	Nominal power: 0,373 kW Power consumption: 0,373*0,8(power consumption factor)*0,17h (10min) = 0,05 kWh	Electricity, low voltage {Europe without Switzerland}| market group for | Cut-off, U
RO brine	Reverse Osmosis treatment waste stream, considered as burden free	NA
Na_2CO_3 salt	Direct data from study (250g of Na_2CO_3 salt + 2,5l of water)	Soda ash, light, crystalline, heptahydrate {GLO}| market for | Cut-off, U
Water for Na_2CO_3 solution	Direct data from study (250g of Na2CO3 salt + 2,5l of water)	Water, deionised {Europe without Switzerland}| market for water, deionised | Cut-off, U
Cleaning Water	10 litres being reused with assumpted loss of 1% (100ml) per cleaning process	Water, deionised {Europe without Switzerland}| market for water, deionised | Cut-off, U
Bag-filter	PP Bag-filter of 200g Service life: Filtration up to 1000m3	Textile, nonwoven polypropylene {GLO}| market for textile, nonwoven polypropylene | Cut-off, U
Cleaning Water	10 litres being reused with assumpted loss of 1% (100ml) per cleaning process	Emissions to air
Bag-filter	managed as non-recyclable plastic waste	Waste plastic, mixture {RER}| market group for waste plastic, mixture | Cut-off, U
Precipitated $CaCO_3$	By-product (credited considering avoided CaCO3 mineral production; “avoided burden” approach)	Calcium carbonate, precipitated {RER}| calcium carbonate production, precipitated | Cut-off, U
Decalcified brine	Product	NA

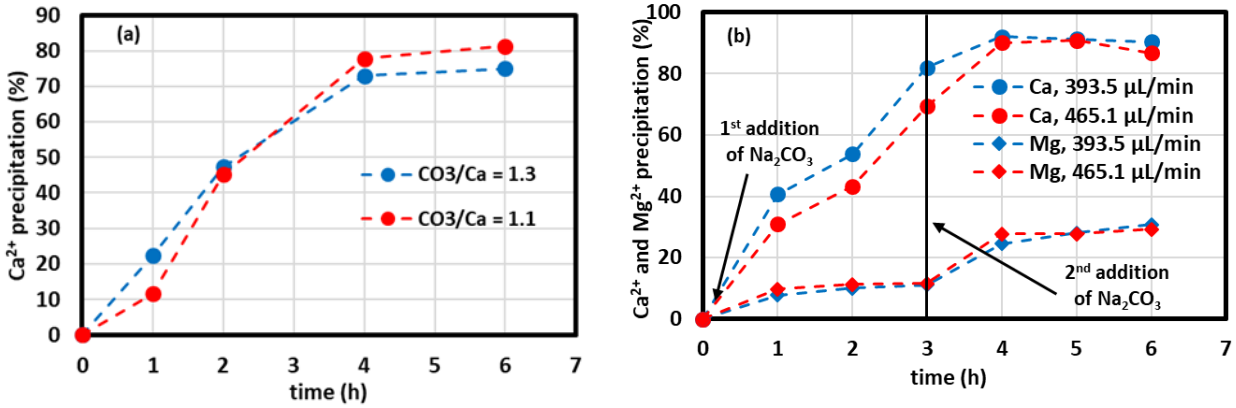

Figure S1. %Ca precipitation at T = 60 °C at different CO3/Ca molar ratio and different Na_2CO_3 addition flowrates vs. the time (V_{SB} = 250 mL, [Na_2CO_3] = 1 M). (a) Na_2CO_3 addition flowrate 393.5 and 465.1 µL/min for CO3/Ca molar ratio 1.1 ($V_{Na2CO3,\ added}$ = 5903.15 µL) and 1.3 ($V_{Na2CO3,\ added}$ = 6976.45 µL), respectively; (b) same of run (a) but successive Na_2CO_3 addition for a total $V_{Na2CO3,\ added}$ =5903.15x2 and 6976.45x2 µL.

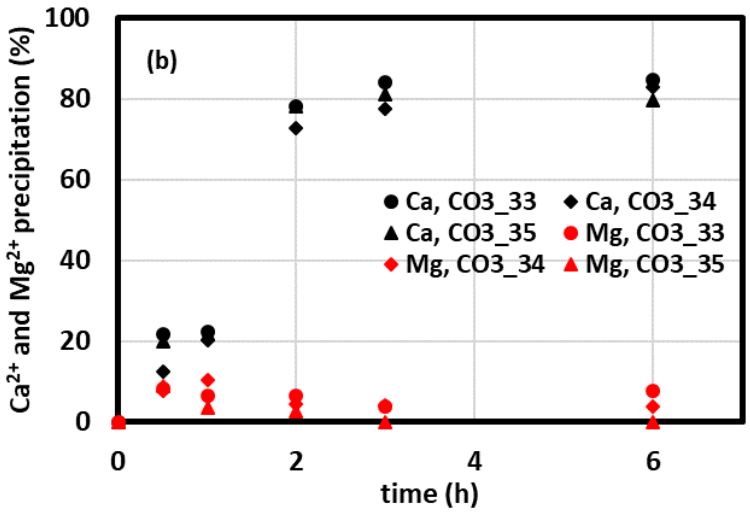

Figure S2. % Ca and %Mg precipitation in presence of antiscalant (2 mg L^{-1}) using different addition mode of the precipitant vs. time (V_{SB} = 500 mL; $V_{Na2CO3,added}$ = 18.2 mL, [Na_2CO_3] = 0.65 M, CO3/Ca ratio = 1.1; CO3_33: syringe pump (910 µL/min); CO3_34: addition to 100 mL of SB and mixing by vortex, then addition to remaining 400 mL and energic stirring for 5 minutes; CO3_35: addition to 100 mL of SB and mixing by vortex then addition to remaining 400 mL and sonication for 5 minutes; T = 35 °C).

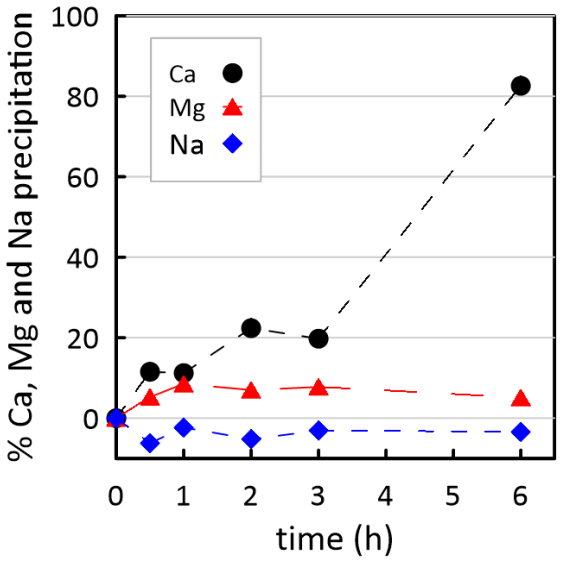

Figure S3: % Ca and %Mg precipitation in presence of minor ions and antiscalant at T = 15°C (V_{SB} = 500 mL, Na_2CO_3 addition: 18.2 mL, 0.65 M, 1.82 mL/min for 10 min; CO3/Ca ratio = 1.1)

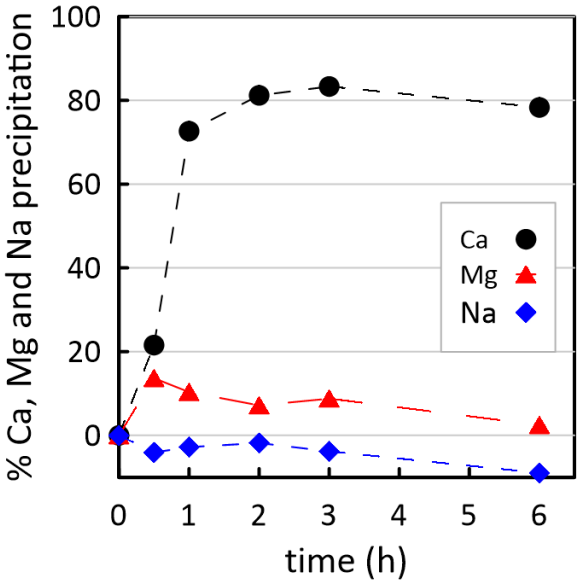

Figure S4. % Ca and %Mg precipitation in presence of minor ions and antiscalant at T = 35°C. (V_{SB} = 500 mL, Na_2CO_3 addition: 18.2 mL, 0.65 M, 1.82 mL/min for 10 min; CO3/Ca ratio = 1.1).

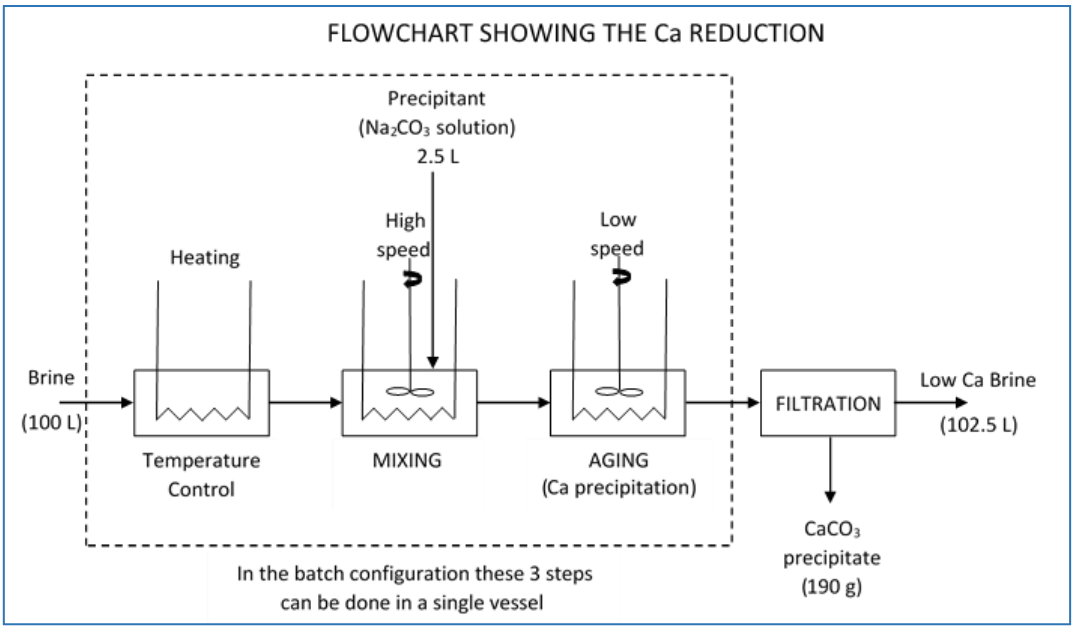

Figure S5. Flowchart showing the Ca reduction from RO-brine

Table S3. Life cycle impact assessment (LCIA) results of the near-ambient temperature calcium precipitation process. Calculated with Environmental Footprint 3.0 method (EF 3.0, adapted for SimaPro), using SimaPro LCA software.

Impact Category	Inputs												Outputs		Total (without AP-approach)	Total (with AP-approach)
	Capital Goods				Operations				Consumables				Waste	By-product		
	Thermostat	Stirrer	Pump	Tanks	Thermostat	Stirrer LOW	Stirrer HIGH	Pump	Na_2CO_3 salt	Water for Na_2CO_3 solution	Cleaning Water	Bag-filter	Bag-filter	Precipitated CaCO3		
CC	5,87E-04	2,51E-03	2,16E-05	5,60E-04	4,79E-01	1,20E-01	3,33E-02	1,99E-02	1,13E-01	1,12E-03	4,49E-05	5,91E-05	2,80E-05	-6,77E-02	7,70E-01	7,02E-01
OD	3,15E-11	1,46E-10	1,17E-12	1,62E-11	2,39E-08	5,99E-09	1,66E-09	9,92E-10	4,73E-09	6,14E-10	2,46E-11	2,28E-12	1,22E-13	-7,85E-09	3,82E-08	3,03E-08
IR	4,57E-05	4,26E-04	1,55E-06	4,76E-05	2,74E-01	6,84E-02	1,90E-02	1,13E-02	5,90E-03	1,23E-04	4,94E-06	4,19E-06	3,76E-08	-5,53E-03	3,79E-01	3,73E-01
POF	3,30E-06	1,19E-05	1,17E-07	1,81E-06	1,09E-03	2,73E-04	7,58E-05	4,52E-05	4,33E-04	3,27E-06	1,31E-07	1,98E-07	1,27E-08	-2,44E-04	1,94E-03	1,70E-03
PM	1,12E-10	2,09E-10	2,29E-12	2,26E-11	8,60E-09	2,15E-09	5,97E-10	3,56E-10	1,58E-08	8,56E-11	3,42E-12	2,36E-12	7,81E-13	-2,41E-09	2,80E-08	2,56E-08
HTc	3,26E-11	4,02E-10	4,97E-12	4,46E-12	6,26E-09	1,56E-09	4,35E-10	2,59E-10	3,97E-09	4,56E-11	1,82E-12	4,81E-13	1,33E-13	-2,30E-09	1,30E-08	1,07E-08
HTnc	4,02E-12	9,42E-12	3,43E-13	1,80E-13	1,96E-10	4,90E-11	1,36E-11	8,12E-12	1,32E-10	1,24E-12	4,97E-14	1,90E-14	1,33E-14	-6,25E-11	4,13E-10	3,51E-10
AP	1,62E-05	3,83E-05	3,81E-07	2,46E-06	2,70E-03	6,76E-04	1,88E-04	1,12E-04	1,51E-03	1,05E-05	4,19E-07	2,59E-07	9,69E-09	-3,65E-04	5,26E-03	4,89E-03
EPf	3,14E-07	2,90E-06	2,90E-08	1,59E-07	4,79E-04	1,20E-04	3,33E-05	1,98E-05	5,10E-05	4,02E-07	1,61E-08	1,44E-08	9,93E-11	-1,78E-05	7,07E-04	6,89E-04
EPm	7,17E-07	1,52E-05	3,10E-08	4,79E-07	4,51E-04	1,13E-04	3,13E-05	1,87E-05	1,59E-04	1,03E-06	4,10E-08	5,14E-08	2,33E-08	-6,31E-05	7,90E-04	7,27E-04
EPt	7,50E-06	4,05E-05	3,75E-07	4,96E-06	3,97E-03	9,93E-04	2,76E-04	1,65E-04	3,96E-03	1,02E-05	4,09E-07	5,36E-07	4,55E-08	-7,01E-04	9,43E-03	8,73E-03
ETf	3,38E-02	3,23E-01	2,81E-03	7,28E-03	6,38E+00	1,59E+00	4,43E-01	2,64E-01	1,37E+01	3,99E+00	1,60E-01	6,67E-04	9,03E-05	-4,20E+00	2,69E+01	2,27E+01
LU	3,03E-03	2,57E-02	1,73E-04	4,30E-03	1,93E+00	4,83E-01	1,34E-01	8,00E-02	1,30E+00	5,13E-03	2,05E-04	1,27E-04	8,46E-06	-2,19E-01	3,96E+00	3,74E+00
WU	6,39E-04	1,06E-03	9,61E-06	2,12E-04	1,18E-01	2,95E-02	8,19E-03	4,89E-03	1,66E-01	1,12E-01	4,50E-03	2,24E-05	1,26E-06	-1,98E-02	4,45E-01	4,25E-01
RUf	9,93E-03	2,87E-02	2,51E-04	1,41E-02	1,01E+01	2,52E+00	7,00E-01	4,18E-01	1,11E+00	1,43E-02	5,70E-04	1,69E-03	9,32E-06	-9,90E-01	1,49E+01	1,39E+01
RUe	5,28E-08	6,63E-07	7,30E-09	2,63E-09	4,41E-06	1,10E-06	3,06E-07	1,83E-07	4,37E-06	1,65E-08	6,61E-10	3,69E-10	2,68E-12	-2,38E-06	1,11E-05	8,73E-06