Supplementary Material

Life Cycle Sustainability Assessment of Microbially Induced Calcium Carbonate Precipitation (MICP) Soil Improvement Techniques

Estimating MICP By-products

To estimate changes in *in-situ* MICP by-product masses within all columns during the rinsing process, several ammonium (NH_4^+) by-product contributions were accounted for. First, aqueous NH_4^+ masses existing within columns at various times during rinsing were determined from direct aqueous NH_4^+ concentrations measured at four sampling port locations (labeled at P_1 through P_4 in **Figure 1**). Since aqueous NH_4^+ concentrations were not uniform across column lengths during the rinsing process, aqueous NH_4^+concentrations were integrated along column lengths, using known column pore volumes, to estimate cumulative aqueous NH_4^+ masses existing within columns.

Second, sorbed NH_4^+ by-product masses were also accounted for in these calculations. NH_4^+ ions sorb to soil particle surfaces, resulting from their attraction to negatively-charged soil minerals, and were measured at the end of all rinse injections using a potassium chloride extraction process. For all measurements, sorbed NH_4^+ masses were found to vary near-linearly with changes in aqueous NH_4^+ concentrations [1]. Although sorbed NH_4^+ masses were not measured directly at various points in time during the rinsing process, aqueous NH_4^+ concentrations existing within columns during rinsing were used to estimate sorbed NH_4^+ masses in time. In these calculations, a linear relationship was assumed between sorbed NH_4^+ masses and aqueous NH_4^+ concentrations. At higher aqueous NH_4^+ concentrations, estimated sorbed NH_4^+ masses were not allowed to exceed the cation exchange capacity (CEC) of the Concrete Sand material (CEC = 2.58 meq/100 grams of soil) and no further increases in sorbed NH_4^+ masses were allowed.

Third, NH_4^+ by-product masses resulting from unhydrolyzed urea were accounted for. While this urea was not hydrolyzed to form NH_4^+ during the experiment, it is likely that the urea would eventually be hydrolyzed to form NH_4^+ when left in subsurface soils. In order to estimate NH_4^+ by-product masses resulting from unhydrolyzed urea at the start of rinsing, the stoichiometry of the urea hydrolysis reaction (i.e., two moles of total NH_4^+ produced per mole of hydrolyzed urea) and mass balance were used accounting for sorbed and aqueous NH_4^+ masses. While the removal of unhydrolyzed urea was not measured directly during the rinsing process, it was assumed that this unhydrolyzed urea was distributed uniformly across column lengths and was removed from columns in a manner that was consistent with passive ions due to the non-polarity of urea molecules. Passive tracer testing removal curves were therefore used to estimate changes in

unhydrolyzed urea concentrations in time during rinsing. Before rinsing, concentrations of unhydrolyzed urea were generally small in both stimulated columns (~23 to 57 mM), however, concentrations of unhydrolyzed urea in the augmented column were much larger (~158 mM) due to poor ureolytic activity.

Following the above approach, changes in aqueous NH_4^+ masses, sorbed NH_4^+ masses, and NH_4^+ masses resulting from unhydrolyzed urea were estimated in time during rinsing. (shown in **Figure 3**) All three forms of NH_4^+ were accounted for when estimating changes in total NH_4^+ by-product masses during rinsing. **Figure 3a** presents aqueous NH_4^+ masses, sorbed NH_4^+ masses, and NH_4^+ masses from unhydrolyzed urea as a function of increasing rinsing time and solution volume for a single column (Column 1). **Figure 3b** presents estimated changes in total NH_4^+ masses for all three columns during rinsing. All NH_4^+ masses in time are tabulated in **Supplementary Table S3**.

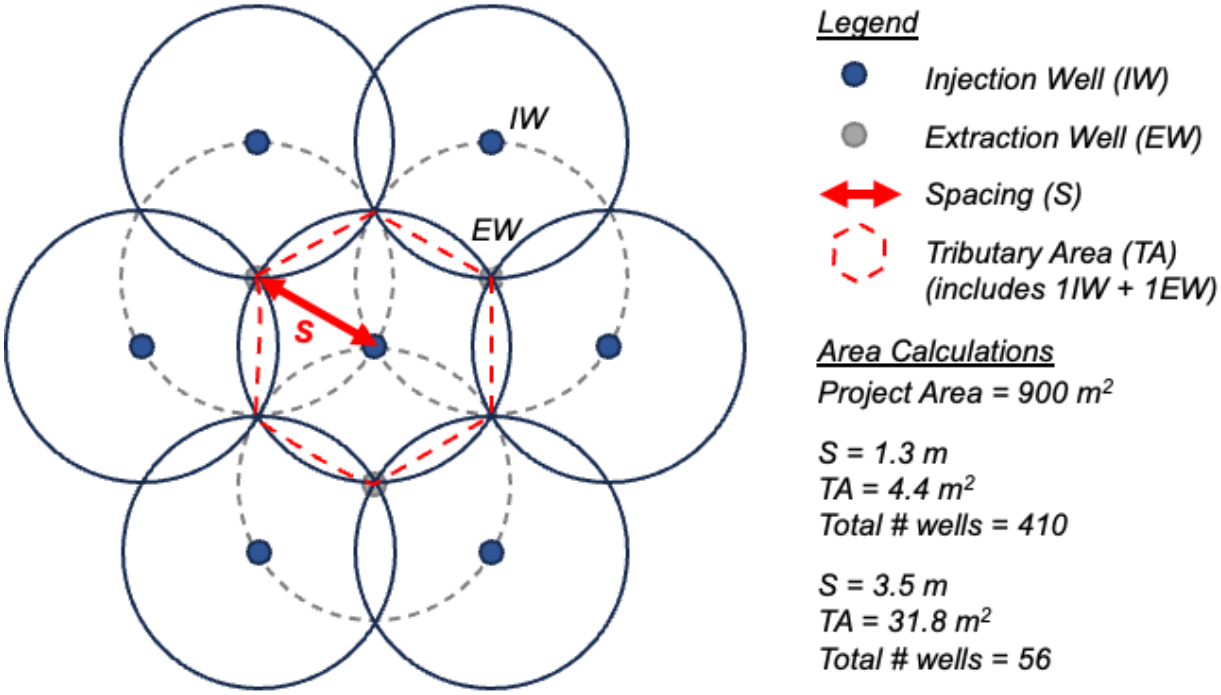
Methods for Upscaling LCSA Results

The life cycle sustainability assessment (LCSA) results, which model the impacts of meter-scale column testing performed in the laboratory, were upscaled to estimate the environmental impacts of MICP for two field-scale implementation scenarios. The methods for upscaling the LCSA results are summarized below. It is noted that these methods contain some simplifying assumptions, and a more comprehensive LCSA of MICP in field applications, including assessment of impacts from materials transportation, equipment mobilization, well construction, and effluent management, is needed.

For Scenario 1, the LCSA results for each MICP treatment approach (i.e., Column 1 through 3) for the case when V_{s4} = 150 m/s and optimal rinsing (i.e., 1.8 PV = 90 L) was employed were upscaled using a scale factor of ~69,000 (9,900 m^3 ÷ 0.14 m^3; the improvement volume at the hypothetical project site divided by the treated soil volume in the experimental column tests). The field-scale results consider impacts from materials use and process emissions but exclude impacts associated with energy consumption from pumping due to a lack of data that cannot be addressed by scaling up experimental processes.

In Scenario 2, only high and low ureolytic rate stimulation (Column 1 and 2) were considered viable treatment options because the augmented column (Column 3) did not meet defined performance criterion (V_s = 400 m/s) during the column experiment. For this scenario, the number of cementation treatments (N_{cement}) required for each column to meet the V_s = 400 m/s criterion were calculated by linear interpolation. Column 1 required 2.23 cementation treatments (using V_s data from Location 2) and Column 2 required 8.88 cementation treatments (using V_s data from Location 4). The LCSA results were reevaluated for the cases when V_{s2} = 400 m/s and N_{cement} = 2.23 for Column 1 and when V_{s4} = 400 m/s and N_{cement} = 8.88 for Column 2. The LCSA results were then upscaled using scale factors of ~178,000 (9,900 m^3 ÷ 0.06 m^3) and ~69,000 (same as in Scenario 1) for Column 1 and 2, respectively. The larger scale factor for Column 1 was used to account for the fact that Column 1 did not achieve V_s = 400 m/s across the entire column length (only to Location 2, positioned 1.33 m from the injection source) and thus would require smaller well spacing when implemented in the field. During field application, the high ureolytic

rate stimulation approach (Column 1) would require injection-to-outlet well spacings of approximately 1.3 m, while the low ureolytic rate stimulation approach (Column 2) could use larger injection-to-outlet well spacings of approximately 3.5 m, assuming a tow-dimensional triangular well pattern (shown in **Supplementary Figure S1**).



Supplementary Figure S1. Schematic of two-dimensional triangular well pattern with injection-to-outlet well distances of 1.3 m and 3.5 m between the injection source, consistent with the distance to V_s locations 2 and 4 in the column experiments.

Supplementary Table S1. Summary of Treatment Schemes and Solution Constituents (adapted from Lee et al. [1] and San Pablo et al. [2]).

Solution Constituent	Column 1: Stimulation (high ureolytic rate)			Column 2: Stimulation (low ureolytic rate)			Column 3: Augmentation		All Columns
	Enrichment	Flush	Cementation	Enrichment	Flush	Cementation	Augmentation	Cementation	NH_4^+ Rinsing
Calcium chloride (mM)	--	--	250	--	--	250	--	250	200
Urea (mM)	50	--	250	50	--	250	--	250	--
Ammonium chloride (mM)	100	12.5	12.5	100	12.5	12.5	--	12.5	--
Sodium acetate (mM)	42.5	42.5	42.5	42.5	42.5	42.5	--	42.5	--
Sodium chloride (g/L)	--	--	--	--	--	--	9	--	--
Sodium hydroxide (g/L)	1.28	--	--	1.28	--	--	--	--	--
Yeast extract (g/L)	0.2	--	0.2	0.04	--	0.02	--	--	--
S. pasteurii (cells/mL)	--	--	--	--	--	--	9.36×10^7* 1.40×10^6**	--	--
Number of Injections	6	1	9	6	1	9	2	9	1
Injection Volume (L)	76	76	76	76	76	76	76* 456**	76	525
Injection Duration (min)	186	186	186	186	186	186	186* 1,140**	186	700

*First augmentation treatment.

**Second augmentation treatment.

Supplementary Table S2. Reference LCI Datasets.

Process or Flow	Reference	Notes or LCI Title
Materials		
Calcium chloride	ecoinvent Database (2015)	soda production, solvay process [RER]
Urea	GaBi Professional Database (2017)	Urea (stami carbon process) [US]
Ammonium chloride	ecoinvent Database (2015)	ammonium chloride production [GLO]
Ammonium sulfate	ecoinvent Database (2015)	market for ammonium sulfate, as N [GLO]
Sodium acetate trihydrate	GaBi Professional Database (2017)	Acetic acid from methanol (low pressure carbonylation) (Monsanto process) [US], Sodium hydroxide (caustic soda) mix (100%) [US], Water deionized [US]; mass-based allocation
Sodium chloride	GaBi Professional Database (2017)	Sodium chloride (rock salt) [US]
Sodium hydroxide	GaBi Professional Database (2017)	Sodium hydroxide (caustic soda) mix (100%) [US]
Water	GaBi Professional Database (2017)	Tap water from surface water [US]
Energy		
Electricity	GaBi Professional Database (2017)	Electricity grid mix [US]

Note: RER = Europe, US = United States, and GLO = Global. These shortcuts are used by ecoinvent [3] and thinkstep [4] to describe the geographical location of a reference LCI dataset. For example, GLO LCI data represent global average activities and processes.

Supplementary Table S3. Mass of *In-situ* MICP By-products with Increasing Rinsing Time and Volume.

	Cumulative rinsing time (min)	0	60	120	240	360	540	700
	Cumulative injected rinse volume (L)	0	45	90	180	270	405	525
Column 1: Stimulation (high ureolytic rate)	Aqueous NH_4^+ (as N) (g)	273.18	146.47	52.40	17.93	9.08	4.80	2.51
	Unhydrolyzed $(NH_2)_2CO$ (as N) (g)	81.71	20.99	0.61	0.00	0.00	0.00	0.00
	Sorbed NH_4^+ (as N) (g)	105.74	105.74	62.45	21.36	10.82	6.85	6.85
	Total (as N) (g)	460.63	273.20	115.46	39.29	19.90	11.64	9.36
Column 2: Stimulation (low ureolytic rate)	Aqueous NH_4^+ (as N) (g)	308.37	94.57	33.82	9.75	4.45	1.74	0.99
	Unhydrolyzed $(NH_2)_2CO$ (as N) (g)	31.13	8.00	0.23	0.00	0.00	0.00	0.00
	Sorbed NH_4^+ (as N) (g)	102.70	102.70	33.96	9.79	5.19	5.19	5.19
	Total (as N) (g)	442.19	205.27	68.01	19.54	9.65	6.93	6.18
Column 3: Augmentation	Aqueous NH_4^+ (as N) (g)	130.77	80.99	72.33	21.37	9.25	3.67	2.70
	Unhydrolyzed $(NH_2)_2CO$ (as N) (g)	223.42	73.22	15.30	0.66	0.03	0.00	0.00
	Sorbed NH_4^+ (as N) (g)	105.11	96.52	86.20	25.47	11.02	7.55	7.55
	Total (as N) (g)	459.31	250.74	173.82	47.50	20.30	11.22	10.25

Supplementary Table S4. Unit Costs for LCCA.

Process or Flow	**CAS No.**	**Unit Cost ($/unit)**	**Reference**
Materials			
Calcium chloride (kg)	10043-52-4	5.90	Avantor
Urea (kg)	57-13-6	106.08	Thermo Fisher Scientific
Ammonium chloride (kg)	12125-02-9	125.67 or 226.00*	Thermo Fisher Scientific
Ammonium sulfate (kg)	7783-20-2	200.00	Thermo Fisher Scientific
Sodium acetate (kg)	6131-90-4	83.80	Thermo Fisher Scientific
Sodium chloride (kg)	7647-14-5	32.60	Thermo Fisher Scientific
Sodium hydroxide (kg)	1310-73-2	197.00	Thermo Fisher Scientific
Tris base (kg)	77-86-1	240.00	Thermo Fisher Scientific
Yeast extract (kg)	--	980.00 or 314.00**	Thermo Fisher Scientific
Water (L)	--	0.002	Circle of Blue 2019
Energy			
Electricity (MJ)	--	0.03	EIA 2020

*Unit cost for Column 3 only.

**Unit cost for Column 1 only.

References

1. Lee, M. *et al.* Investigating Ammonium By-product Removal for Ureolytic Bio-cementation Using Meter-scale Experiments. *Sci. Rep.* **9**, 1–15 (2019).
2. San Pablo, A. C. *et al.* Meter-scale Bio-cementation Experiments to Advance Process Control and Reduce Impacts: Examining Spatial Control, Ammonium By-product Removal, and Chemical Reductions. *J. Geotech. Geoenviron. Eng.* **146**, 04020125 (2020).
3. ecoinvent. ecoinvent 3.5. (2018). at <https://www.ecoinvent.org/database/ecoinvent-35/ecoinvent-35.html>
4. thinkstep. GaBi ts 8: System software and databases for life cycle engineering. (2018).