

**Supplemental Information: Alkaline Mineral Residues from Pulp Mills as a Sustainable  
and Economical Alternative to Lime Fertilizers**

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## Methods

### Soil Collection and Preparation

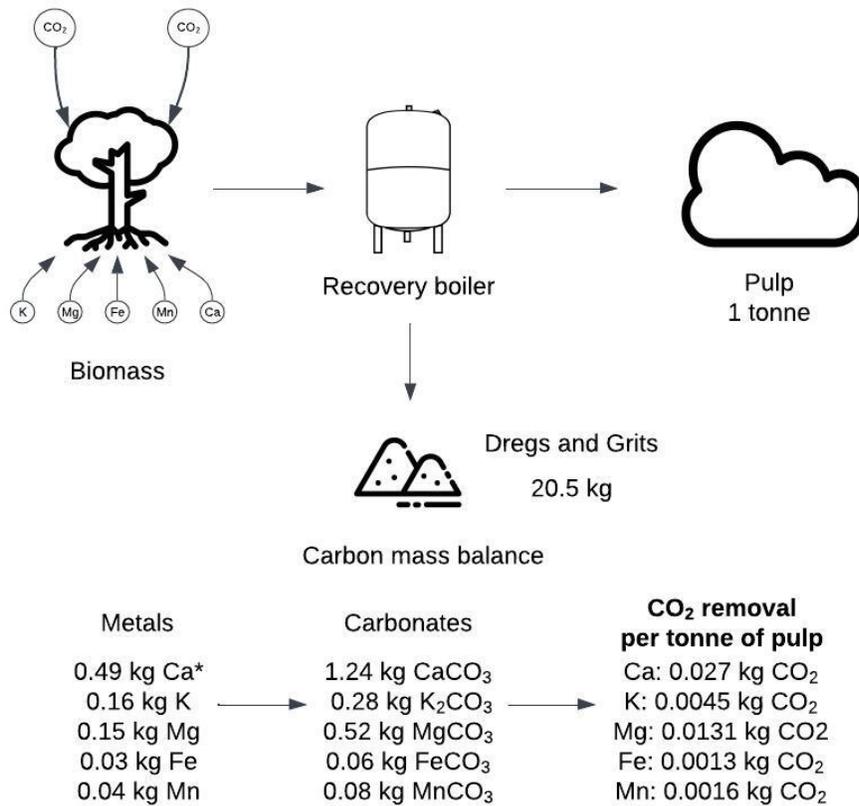
To identify these soil types, we combined mapped soil classification data (Cook et al., 2024) with recent satellite-derived maps of existing cotton fields (USDA, 2020) and managed pine stands (Thomas et al., 2021) across the southeastern US. We ranked each soil type by the total acreage of cotton or loblolly each hosted and selected the top 100 soil types for each crop (out of approximately 42,000 unique soil types present in the southeastern US). After excluding soil types with an average pH > 6 (Soil survey staff, 2020), we searched for qualifying top-100 soil types within up to approximately 100 miles of any of the identified pulp mills, then contacted landowners managing either cotton or plantation pine covering any of these soil types to arrange for site access and sample acquisition.

### Life Cycle and Techno-Economic Analysis

$$\frac{g \text{ of carbonate}}{g \text{ of metal}} = \text{molar mass of carbonate} \left( \frac{g}{mol} \right) * \frac{1 \text{ mole of carbonate}}{\# \text{ moles of metal in carbonate}} * \frac{1 \text{ mol of metal}}{\text{mass of 1 mol of metal}} \quad (S1)$$

$$\frac{\text{mass of carbonate (g)}}{\text{mass of DG (g)}} = \text{Mass fraction of metal in DG (\%)} * \frac{\text{mass of carbonate (g)}}{\text{mass of metal (g)}} \quad (S2)$$

$$\frac{\text{mass of CO}_2 \text{ (g)}}{\text{mass of DG (g)}} = \frac{\text{mass of carbonate (g)}}{\text{mass of DG (g)}} * \frac{\text{molar mass of CO}_2 \left( \frac{g}{mol} \right)}{\text{molar mass of carbonate} \left( \frac{g}{mol} \right)} \quad (S3)$$



**Figure S1. Process flow diagram and mass balance of carbon within dregs and grits**

For every 1 tonne of pulp produced, 20.5 kg of DGs are produced as byproduct. Within the DGs, a certain percentage of the mass is made up of metals that are capable of forming carbonates in the recovery boiler, namely calcium, potassium, magnesium, iron, and manganese. Using the mass of metals in the DGs, the mass of their respective carbonates, and the ratio of the molar mass of CO<sub>2</sub> to the molar mass of carbon, we were able to calculate the mass of CO<sub>2</sub> that is drawn down for every tonne of pulp or DGs produced (Figure S1). For calcium, several additional steps (shown in equations S4-S5) were taken to calculate the calcium that originates in tree biomass as opposed to the mineral CaCO<sub>3</sub> used in the pulping process.

$$\frac{CaO \text{ in pine biomass}}{K_2O \text{ in pine biomass}} = \text{Ash content of pine chips (\%)} * \frac{\text{Mass fraction of CaO (\%)}}{\text{Mass fraction of K}_2\text{O (\%)}} \quad (S4)$$

$$\frac{\text{Mass of Ca from biomass in DG (g)}}{1 \text{ g of K in DG}} = \frac{\text{CaO in pine biomass}}{\text{K}_2\text{O in pine biomass}} * \frac{\text{Molar mass of Ca} \left( \frac{\text{g}}{\text{mol}} \right)}{\text{Molar mass of CaO} \left( \frac{\text{g}}{\text{mol}} \right)} * \frac{\text{Molar mass of K}_2\text{O} \left( \frac{\text{g}}{\text{mol}} \right)}{\text{Molar mass of K} \left( \frac{\text{g}}{\text{mol}} \right)} \quad (\text{S5})$$

The mass fraction of the carbonates within the ash content of trees was used in order to determine the amount of calcium relative to the other previously mentioned metals (Vassilev et al., 2010). Using this ratio, the mass fraction of Ca<sup>2+</sup> within DGs that originated in the tree biomass can be calculated which enabled us to repeat equations S4-S5 to finish off the carbonate mass balance and resulting levelized CO<sub>2</sub> removal. Following the calculation of the CO<sub>2</sub> drawdown attributed to DGs, the emissions released by preparing DGs for soil application were then modeled.

We assumed a transport distance of 50 miles to the agricultural fields, and the emissions of the trucks returning to the pulp mill are accounted for. The cost of the industrial crusher was calculated based on the amount of DGs processed per year (Breunig et al., 2024). The cost of the dryer was based on an industry quote (FEECO International, 2025). The plant utilization, indirect capital costs, capital cost contingency, return on equity, project life, and capital recovery factor were chosen based on standards found in industry and literature (Humbird et al., 2011; Keith et al., 2018; Pett-Ridge et al., 2023). The overall costs of the proposed system were calculated using equations S6 – S12 listed below. The inputs required are capital recovery factor, facility utilization, return on equity, capital costs, and operating costs.

$$\text{Levelized cost of DG production} = \text{net operating cost} + \text{levelized capital cost} \quad (\text{S6})$$

$$\text{Levelized capital cost} = \text{capital intensity} \times \frac{\text{capital recovery factor}}{\text{biorefinery utilization}} \quad (\text{S7})$$

$$\text{Capital intensity} = \frac{\text{total capital cost}}{\text{CO}_2 \text{ capacity}} \quad (\text{S8})$$

*Total capital cost = direct capital cost + indirect capital cost + contingency cost (S9)*

*Direct capital cost*

$$= \sum_i \text{reference cost of equipment or material}_i \times \left( \frac{\text{cost year index}_i}{\text{reference index}_i} \right) \times \times \text{instal}$$

$$\text{Capital recovery factor} = \frac{\text{weighted average cost of capital } (i) \times (i + 1)^{\text{project life}}}{(1 + i)^{\text{project life}} - 1} \quad (S11)$$

*Weighted average cost of capital (i)*

$$= (\text{return on equity} \times \text{equity financing } (\%)) + (\text{interest on debt} \times \text{debt financing}) \quad (S12)$$

## **Material Characterization**

X-Ray diffraction (XRD) was used to identify the diffraction patterns of the various materials, specifically the DGs to compare to calcite. A PANalytical Empyrean X-ray diffractometer was used for data collection in the  $2\theta$  range of  $10^\circ$  -  $80^\circ$ . Diffraction was scanned at 0.1313-degree intervals. Scanning electron microscopy (SEM) coupled with energy dispersive spectroscopy (EDS) was used for elemental analysis and backscattered electron images. A variable pressure Hitachi SU3900 at a voltage of 20 kV and a resolution of 4.0 nm was used for SEM imaging. The North Carolina Department of Agriculture (NCDA) analyzed the 22 soils using a Mehlich-3 soil extraction test which provided information on common nutrients such as phosphorus and calcium, heavy metals such as arsenic and nickel, and chemical properties including cation exchange capacity and base saturation. The NCDA also provided nutrient measurements, chemical properties, and heavy metals content of the DGs.

Plot	State Name	Recent Crop	Number of Buckets	W/V g/cc	HM %	CEC meq/100cc	p H	BS %	Bp H	Ac meq/100cc
AL_1	Alabama	Either winter wheat or rye, previous crop was cotton	4	1.36	0.41	5.3	5.9	76	6.29	1.2
AL_2	Alabama	Grass and cotton both	2	1.42	0.36	3.7	5.7	72	6.34	1
GA_1	Georgia	Cotton	2	1.54	0.36	3.6	6.3	83	6.45	0.6
GA_2	Georgia	Cotton	4	1.54	0.46	4.2	6.1	83	6.42	0.7
LA_1	Louisiana	clover (cover), but recently had cotton in cotton-corn-pea nut rotation	2	1.29	0.71	7.6	6.6	93	6.46	0.6
MF_1	Georgia	Loblolly	2	1.09	2.92	6.8	4.4	23	5.29	5.2
PC_10	Alabama	Loblolly	2	0.98	0.81	4.4	4.5	18	5.71	3.6
PC_3	Louisiana	Loblolly	2	1.08	0.51	12.7	6.4	90	6.28	1.3
PC_5	Arkansas	Loblolly probably (based on aerial) - clearcut right now	2	1.18	0.22	5.2	4.8	52	5.98	2.5
PC_9	Alabama	Loblolly	2	1.24	0.09	4	5.1	57	6.18	1.7
RW_19	Louisiana	Loblolly	4	1.41	0.86	4.1	4.6	31	5.89	2.8
RY_1	Georgia	Loblolly	4	1.46	1.14	5.9	3.4	12	5.3	5.2
RY_2	Georgia	Loblolly	2	1.27	1.61	6.6	3.3	14	5.17	5.7
TR_1	Georgia	Loblolly	2	1.36	0.46	4.2	4.8	52	6.1	2
TR_10	Arkansas	Loblolly	2	1.25	0.32	5.9	4.8	66	6.09	2
TR_11	Texas	Loblolly	4	1.19	0.32	9.4	4.9	74	6	2.4
TR_2	Georgia	Loblolly	2	1.27	0.32	4	5.4	67	6.27	1.3
TR_3	Alabama	Loblolly	4	1.47	0.41	2.7	4.9	52	6.27	1.3
TR_4b	Alabama	Loblolly	2	1.42	0.46	2.4	5.1	44	6.26	1.4
TR_4g	Alabama	Loblolly	2	1.39	0.51	3.5	5.3	59	6.24	1.4
TR_5	Alabama	Loblolly	2	1.4	0.51	2.8	4.5	24	6.07	2.1
TR_7	Mississippi	Sweetgum	2	1.05	1.25	3.8	4.6	27	5.91	2.8

Plot	State Name	Recent Crop	Ca	Mg	P	K	S	Zn	Cu	Mn	Na
			mg/dm <sup>3</sup>								
AL_1	Alabama	Either winter wheat or rye, previous crop was cotton	580	109	48	111	21	2.52	1.36	156	7
AL_2	Alabama	Grass and cotton both	360	61	91	145	12	2.4	2.68	95	10
GA_1	Georgia	Cotton	420	49	82	180	7	3.88	0.46	31	8
GA_2	Georgia	Cotton	500	61	83	170	11	4.6	0.76	33	10
LA_1	Louisiana	clover (cover), but recently had cotton in cotton-corn-pea nut rotation	1180	85	84	184	8	2.84	1.06	63	10
MF_1	Georgia	Loblolly	220	49	14	57	22	4.64	0.22	4	19
PC_10	Alabama	Loblolly	100	24	5	39	15	0.64	0.36	2	9
PC_3	Louisiana	Loblolly	1780	243	13	207	10	5	1.2	501	19
PC_5	Arkansas	Loblolly probably (based on aerial) - clearcut right now	380	85	12	70	11	1.96	1.06	32	10
PC_9	Alabama	Loblolly	300	73	2	41	22	0.44	0.4	60	9
RW_19	Louisiana	Loblolly	180	24	5	31	8	0.92	0.34	41	9
RY_1	Georgia	Loblolly	80	24	19	37	7	0.68	0.08	2	14
RY_2	Georgia	Loblolly	80	49	6	35	5	1.64	0.08	1	20
TR_1	Georgia	Loblolly	340	49	17	35	9	0.6	0.22	20	7
TR_10	Arkansas	Loblolly	600	85	6	61	8	1.56	0.42	25	9
TR_11	Texas	Loblolly	1080	158	6	82	16	1.08	1.22	265	21
TR_2	Georgia	Loblolly	380	73	5	68	9	0.44	0.4	194	8
TR_3	Alabama	Loblolly	220	36	32	39	7	1.32	0.26	34	9
TR_4b	Alabama	Loblolly	180	12	5	29	9	0.4	0.36	106	8
TR_4g	Alabama	Loblolly	340	36	7	29	7	1.8	0.48	143	7
TR_5	Alabama	Loblolly	100	12	7	20	10	0.84	0.34	17	9
TR_7	Mississippi	Sweetgum	120	36	8	47	30	0.92	1.1	13	19

Plot	State Name	Recent Crop	As ppm	Cd ppm	Cr ppm	Ni ppm	Pb ppm	Se ppm
AL_1	Alabama	Either winter wheat or rye, previous crop was cotton	0.6	BDL	0.2	0.13	5.8	0.1
AL_2	Alabama	Grass and cotton both	0.2	BDL	0.2	0.12	3.5	BDL
GA_1	Georgia	Cotton	0.8	0.1	0.2	0.13	2.8	BDL
GA_2	Georgia	Cotton	0.6	BDL	0.2	0.13	4.1	0.1
LA_1	Louisiana	clover (cover), but recently had cotton in cotton-corn-pea nut rotation	0.9	0.1	0.2	0.51	2.2	0.1
MF_1	Georgia	Loblolly	0.1	0.1	0.1	0.31	5.0	0.1
PC_10	Alabama	Loblolly	0.1	BDL	0.3	0.15	3.4	0.1
PC_3	Louisiana	Loblolly	0.2	0.2	0.1	2.16	3.7	0.3
PC_5	Arkansas	Loblolly probably (based on aerial) - clearcut right now	0.1	BDL	0.2	0.42	4.8	BDL
PC_9	Alabama	Loblolly	0.1	BDL	0.2	0.09	3.0	0.2
RW_19	Louisiana	Loblolly	0.1	BDL	0.1	0.19	4.0	0.1
RY_1	Georgia	Loblolly	BDL	BDL	BDL	0.14	1.2	BDL
RY_2	Georgia	Loblolly	BDL	BDL	BDL	0.13	1.0	BDL
TR_1	Georgia	Loblolly	0.1	BDL	0.1	0.22	3.1	BDL
TR_10	Arkansas	Loblolly	0.2	0.1	0.2	0.34	4.8	0.1
TR_11	Texas	Loblolly	0.1	BDL	0.2	0.63	4.5	0.3
TR_2	Georgia	Loblolly	0.2	BDL	0.2	0.12	3.7	0.1
TR_3	Alabama	Loblolly	0.1	BDL	0.2	0.13	1.6	0.2
TR_4b	Alabama	Loblolly	0.1	BDL	0.2	0.15	2.7	0.1
TR_4g	Alabama	Loblolly	0.1	BDL	0.1	0.17	2.5	0.1
TR_5	Alabama	Loblolly	0.1	BDL	0.2	0.15	1.9	BDL
TR_7	Mississippi	Sweetgum	0.2	BDL	0.1	0.17	6.6	0.2

**Figure S2. North Carolina Department of Agriculture soil report data**

## Report Abbreviations

Ac	exchangeable acidity
B	boron
BS%	% CEC occupied by basic cations
Ca%	% CEC occupied by calcium
CEC	cation exchange capacity
Cu-I	copper index
ESP	exchangeable sodium percent
HM%	percent humic matter
K-I	potassium index
K <sub>2</sub> O	potash
Mg%	% CEC occupied by magnesium
MIN	mineral soil class
Mn	manganese
Mn-AI1	Mn-availability index for crop 1
Mn-AI2	Mn-availability index for crop 2
Mn-I	manganese index
M-O	mineral-organic soil class
N	nitrogen
Na	sodium
NO <sub>3</sub> -N	nitrate nitrogen
ORG	organic soil class
pH	current soil pH
P-I	phosphorus index
P <sub>2</sub> O <sub>5</sub>	phosphate
S-I	sulfur index
SS-I	soluble salt index
W/V	weight per volume
Zn-AI	zinc availability index
Zn-I	zinc index

Figure S3. North Carolina Department of Agriculture soil report abbreviations

Material	Nutrient Measurements (ppm)													
	N	P	K	Ca	Mg	S	Fe	Mn	Zn	Cu	B	C	Al	Na
Dregs	443	1230	7910	299000	8840	10200	1430	2850	266	60.7	16.5	103000	2010	37200
Dregs	1070	1200	7190	315000	9210	12300	1400	3030	274	63.8	15.2	105000	2100	34400
Grits	992	2030	911	373000	3310	1320	1870	26.6	48.1	4.64	10.7	74200	5130	6630
Grits	833	1820	863	362000	3430	1280	2150	26.5	46.6	4.49	10.1	76500	4980	6500

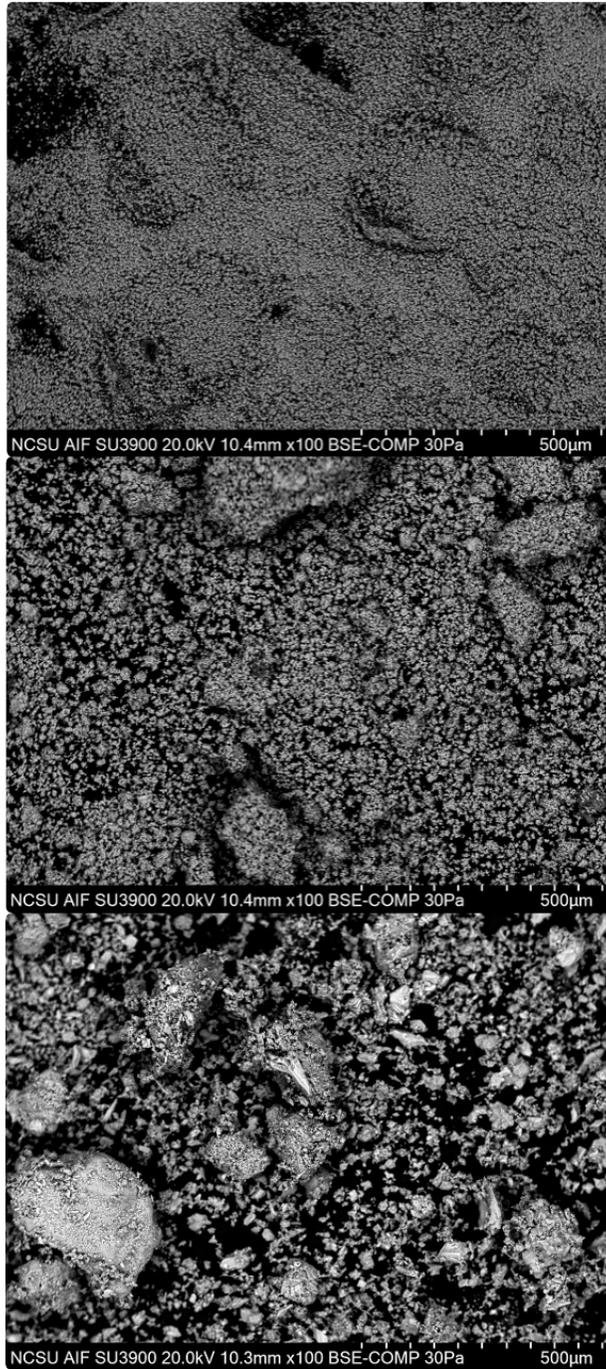
Material	SS (10 <sup>-5</sup> S/cm)	EC (mS/cm)	pH	CCE (%)	ALE (tons)	C:N	DM (%)
Dregs		3450		12.63	93.9	0.96	232:1
Dregs		3550		12.54	94	0.96	97.4:1
Grits		1290		12.57	94.8	0.95	74.9:1
Grits		998		12.57	93.8	0.96	91.9:1

Material	Heavy Metals (ppm)					
	As	Cd	Cr	Ni	Pb	Se
Dregs	0.61	1.88	13	30	129	1.64
Dregs	0.67	1.98	13.1	31.2	9.07	1.7
Grits	0.87	0.01	5.31	6.51	0.47	0.18
Grits	0.79	0.01	5.3	6.83	0.27	0.13

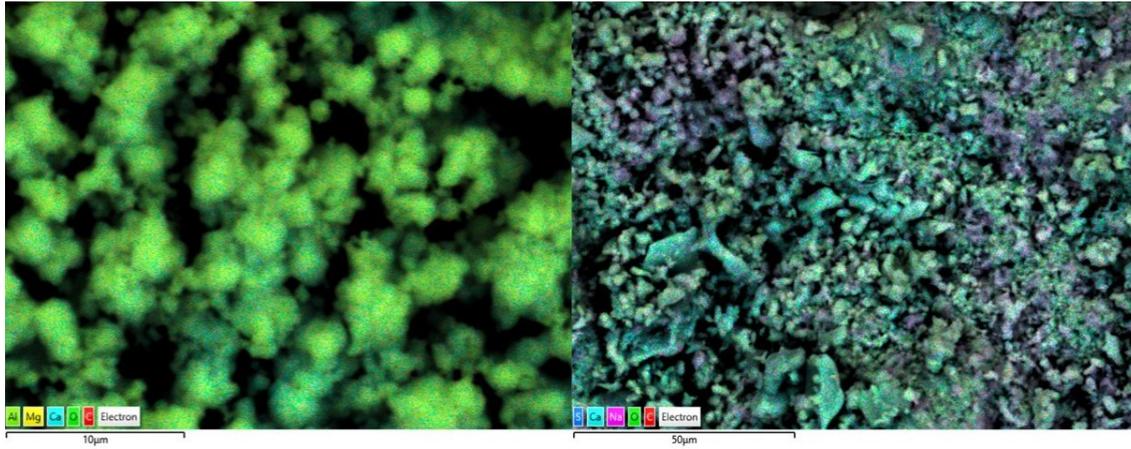
**Figure S4. North Carolina Department of Agriculture waste report data for dregs and grits**

ALE is Agricultural Lime Equivalence. The ALE indicates the amount of the waste material that provides a limiting effect equivalent to one ton of agricultural grade limestone. BD is Bulk Density in lb/yd <sup>3</sup> . CCE is Calcium Carbonate Equivalence and is used to determine ALE. C:N ratio is the Carbon:Nitrogen ratio.	DM% is percent Dry Matter [for semi-solid and solid waste, this value facilitates conversion of dry-basis concentrations (ppm) back to wet-basis of original sample]. EC (Electrical Conductivity) measures salinity, or soluble salts (SS). pH measures basicity/acidity.	Al = Aluminum As = Arsenic B = Boron Ca = Calcium Cd = Cadmium Cl = Chloride Cr = Chromium	Cu = Copper Fe = Iron K = Potassium Mg = Magnesium Mn = Manganese Mo = Molybdenum N = Nitrogen Na = Sodium	NH <sub>4</sub> -N = Ammonium -N Ni = Nickel NO <sub>3</sub> -N = Nitrate -N P = Phosphorus Pb = Lead S = Sulfur Se = Selenium
meq/L = milliequivalent per liter;	mS = millisiemens;	ppm = parts per million or mg/L;	S = siemens;	T = trace (<0.005 lb/unit)

**Figure S5. North Carolina Department of Agriculture waste report abbreviations**



**Figure S6. Scanning electron microscopy images of calcite (top), dregs (middle), grits (bottom)**



**Figure S7. SEM Results with elemental mapping for calcite (left) and grits (right)**

**Table S1. Heavy metal concentration of DGs and limits established by EPA for soil amendments**

	Maximum allowable concentration of industrial solid waste applied to soil (ppm) (USDA, 2000)	Dregs (ppm)	Grits (ppm)
Arsenic	75	0.64	0.83
Cadmium	85	1.93	0.01
Chromium	3000	13.1	5.31
Nickel	75	30.6	6.7
Lead	420	69	0.4
Selenium	100	1.69	0.16

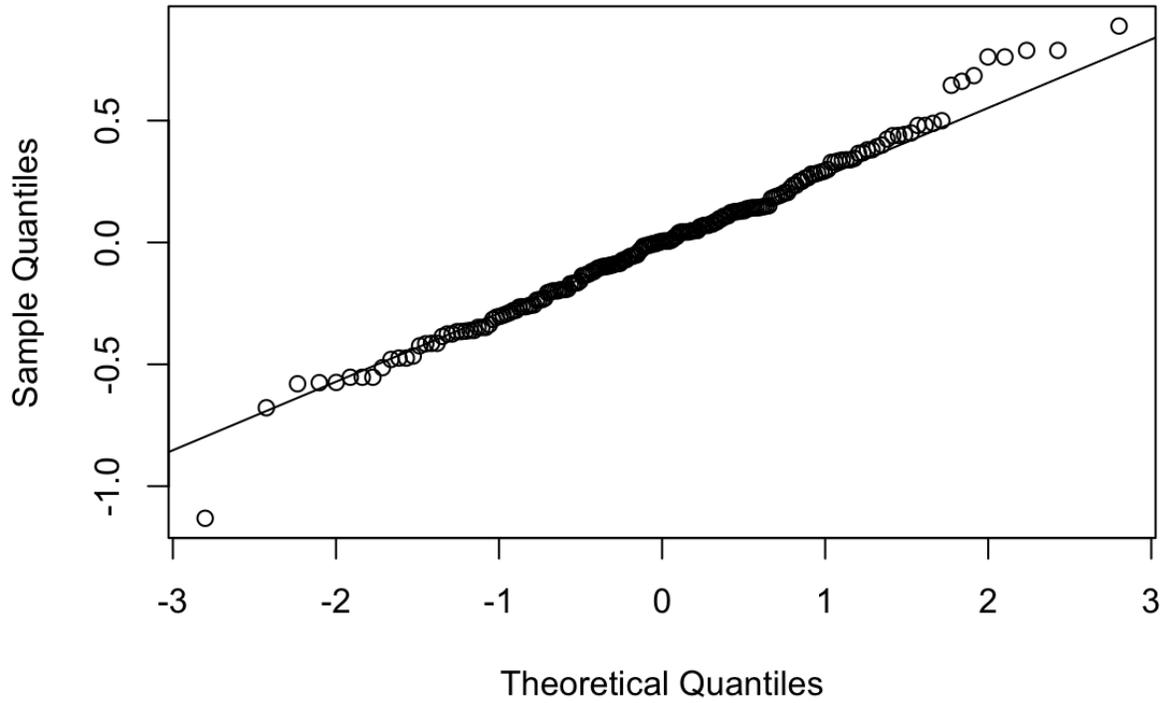


Figure S8. Testing of Normality for soil pH change data (Q-Q Plot)

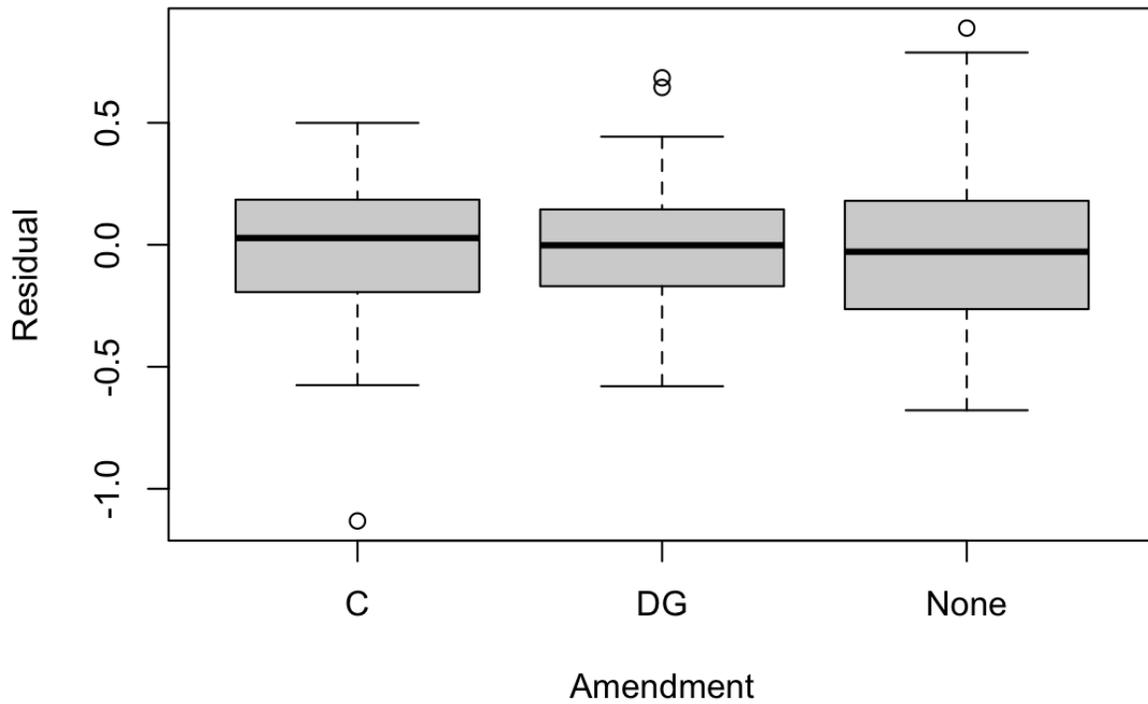


Figure S9. Testing of Homogeneous Variance for soil pH change data

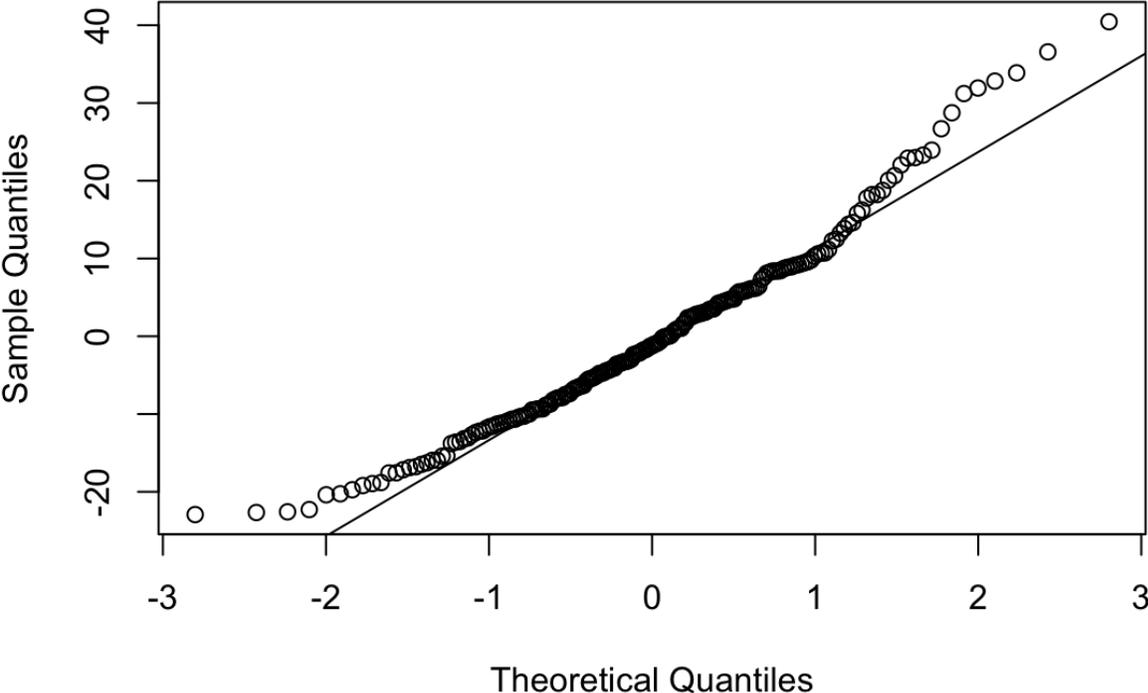
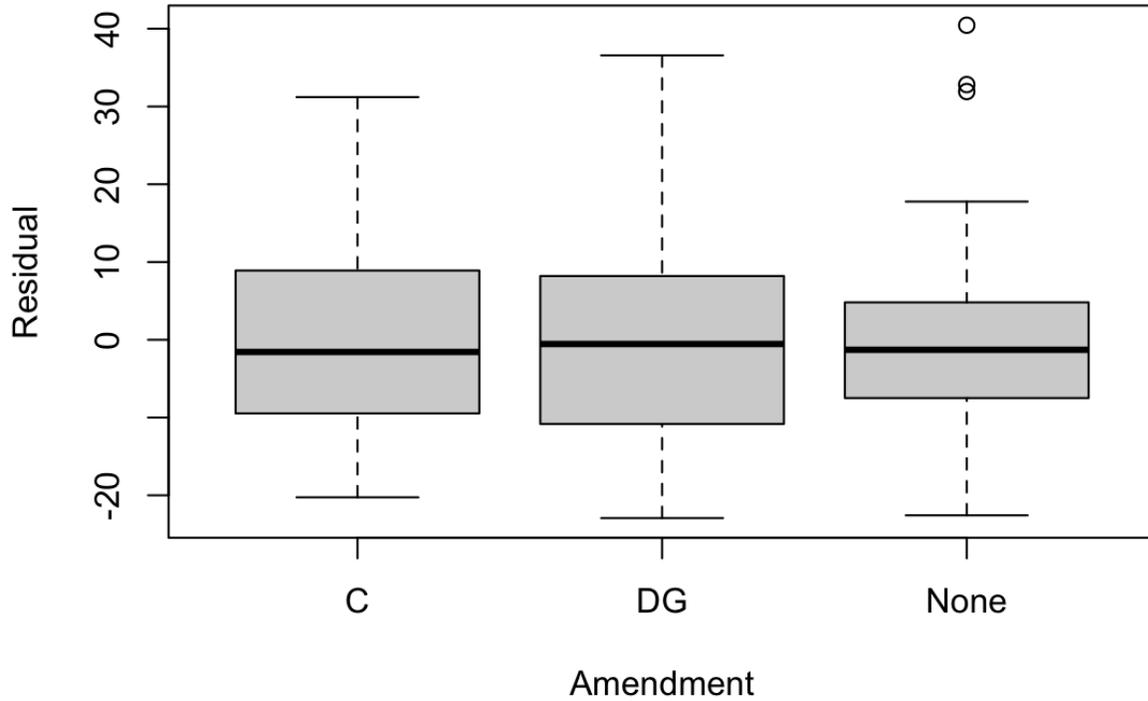


Figure S10. Testing of Normality for soil carbon change data

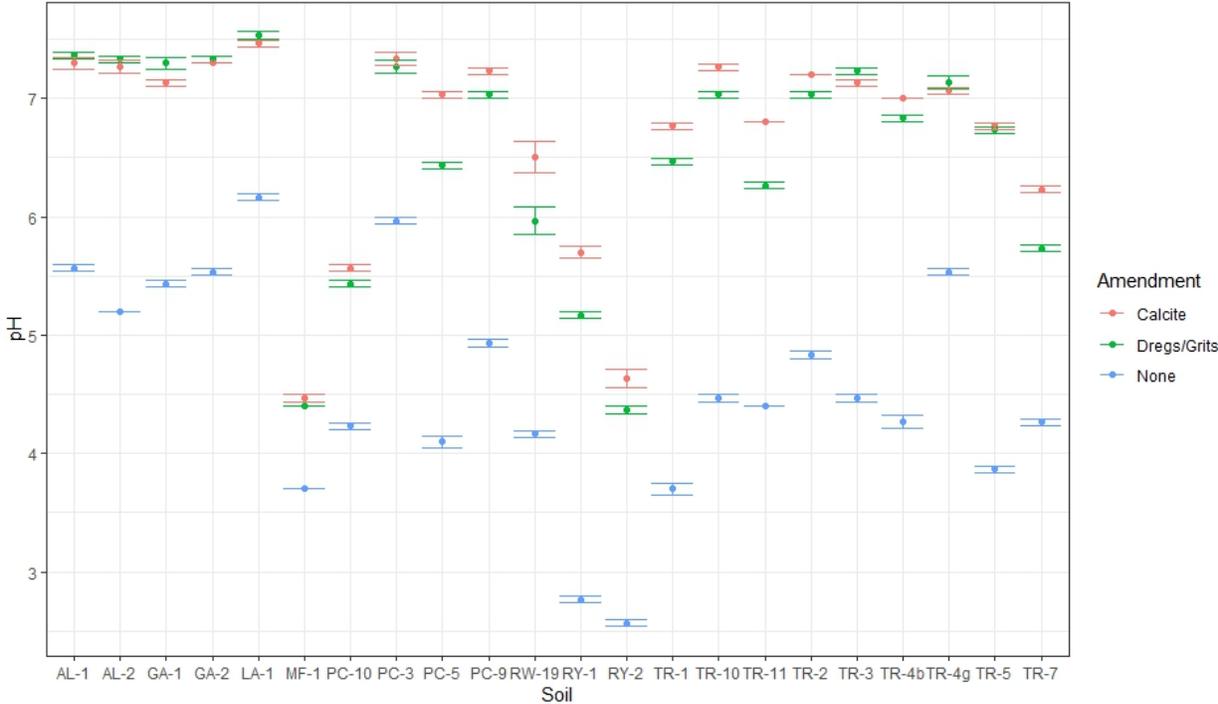


**Figure S11. Testing of Homogeneous Variance for soil carbon change data**

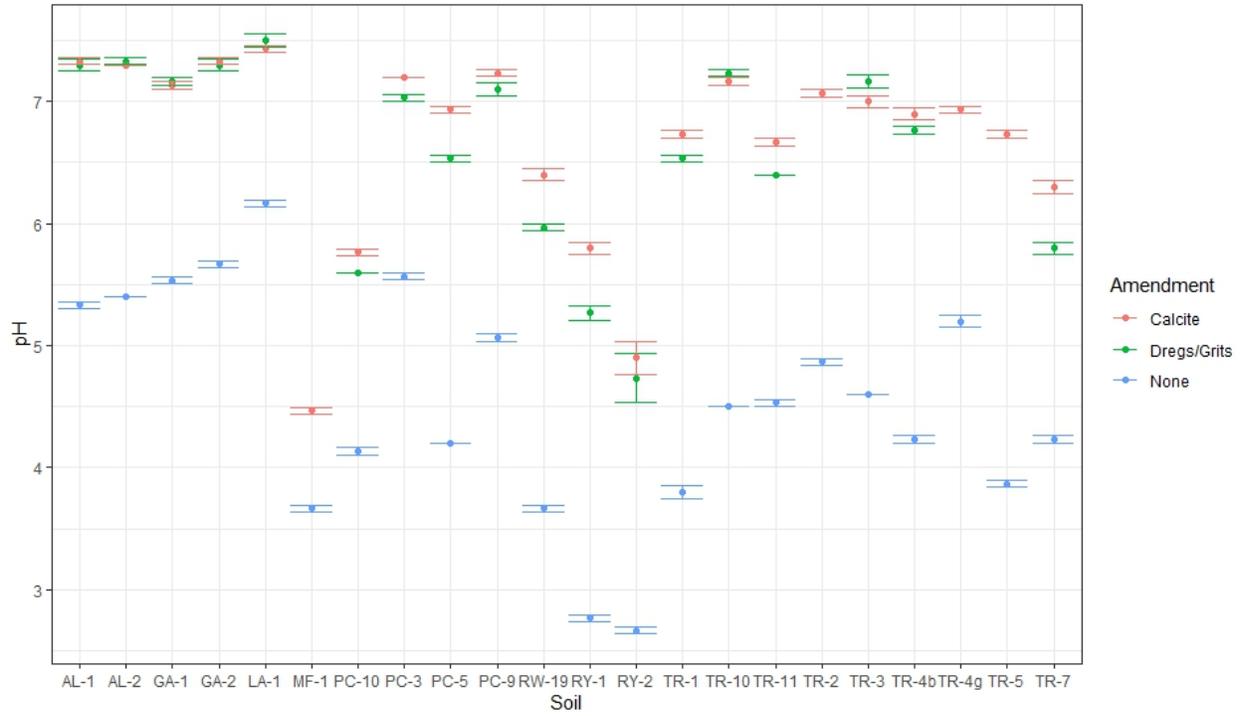
**Table S2. Emissions associated with the post-production drying, crushing, and transport of dregs and grits following generation at a pulp mill without drying**

Process step	Emissions (tCO <sub>2</sub> /year)	Levelized emissions (tCO <sub>2</sub> /tDG)
Atmospheric fixation of CO <sub>2</sub>	-1818	-0.17
Crushing	4	0.00
Embodied equipment emissions	293	0.04
Front end loading DGs	476	0.07

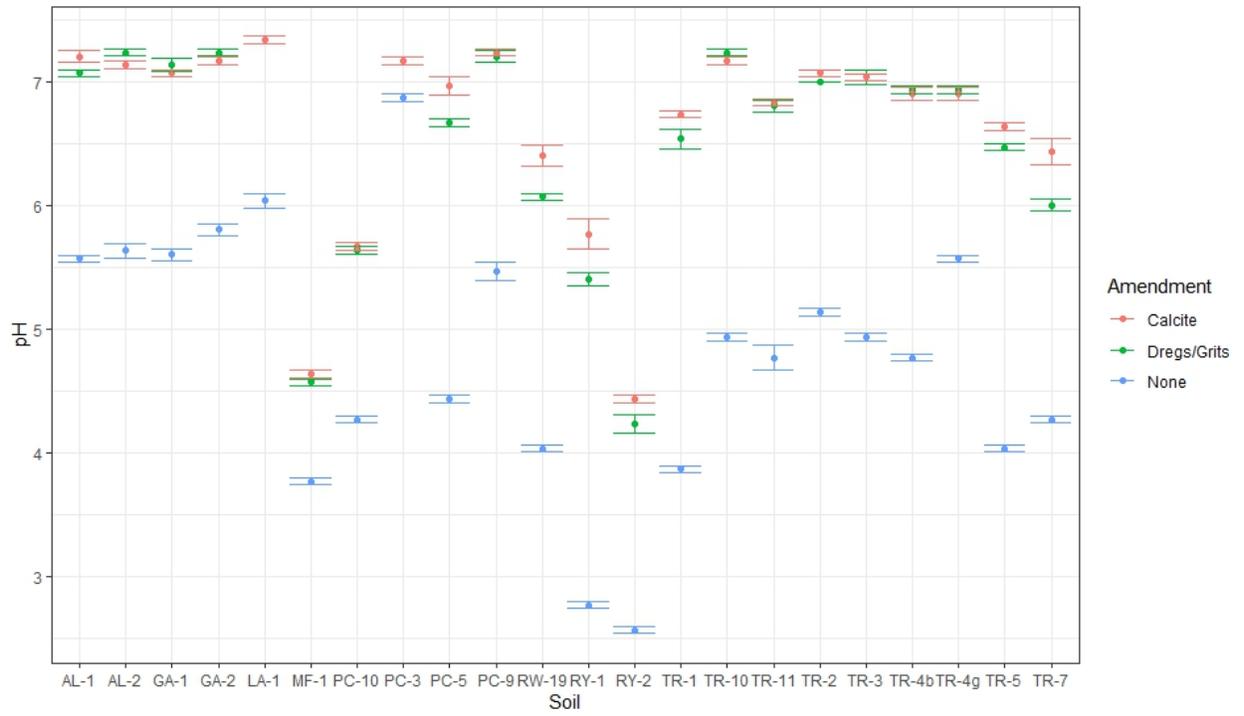
into trucks and transport to fields		
Field weathering of DGs	-1	0.00
Total	-498	-0.07



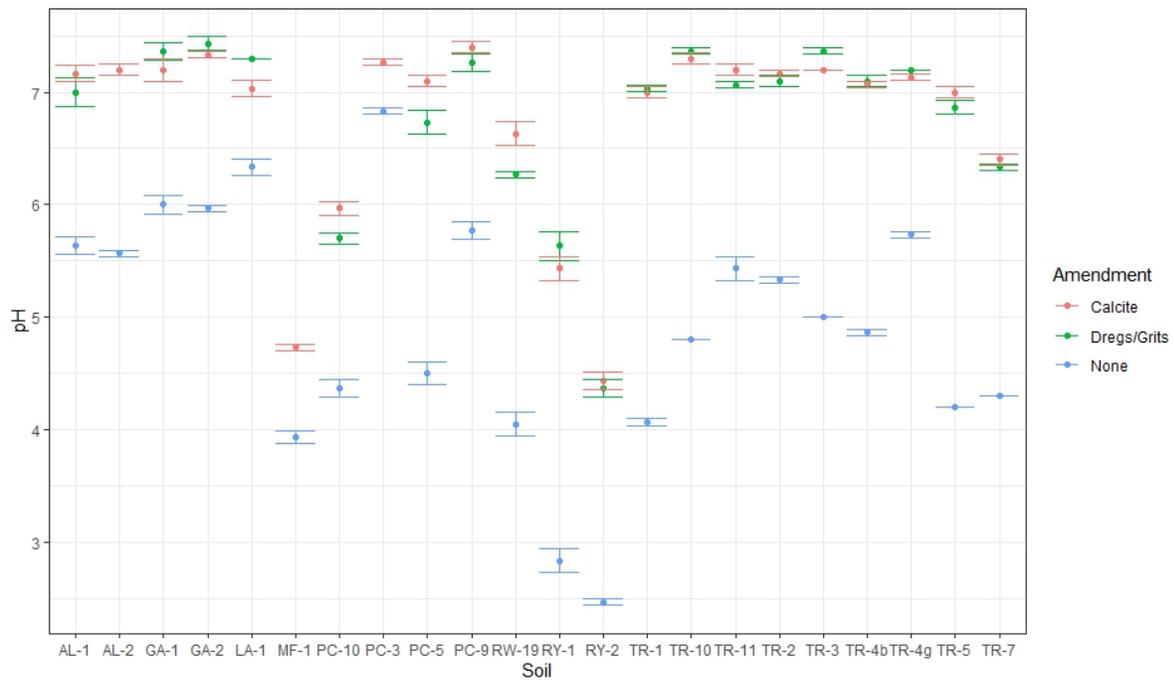
**Figure S13. 0-day soil incubation results**



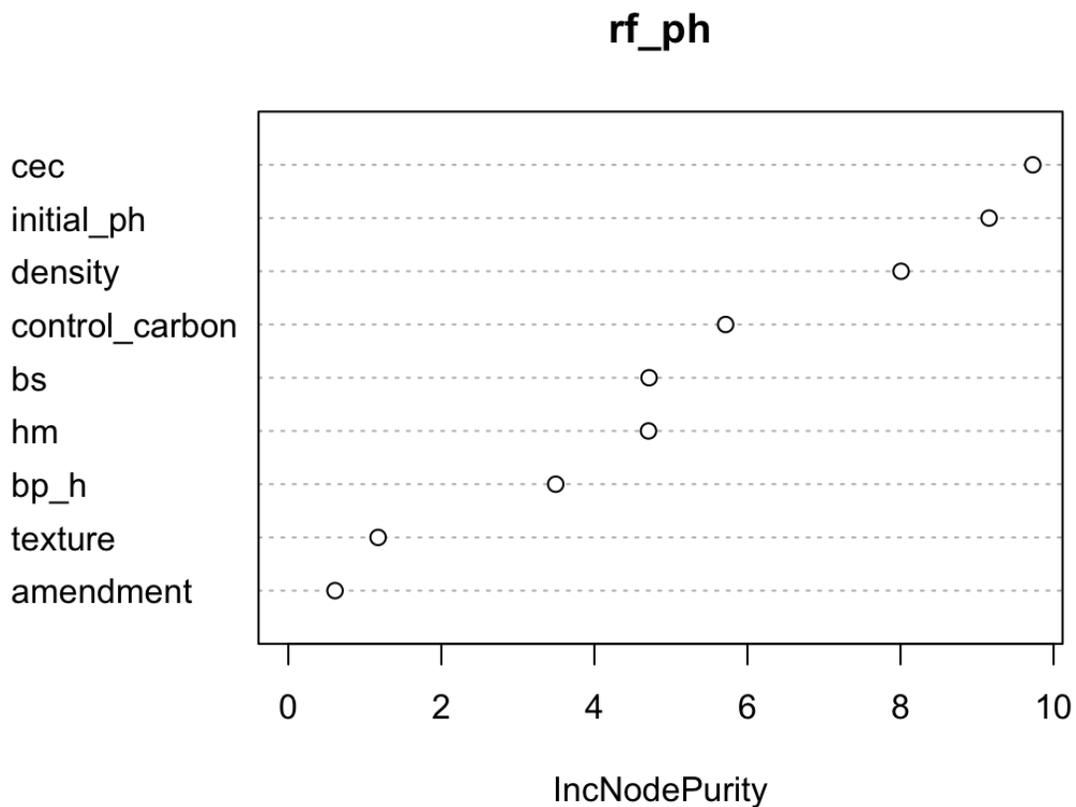
**Figure S14. 1-day soil incubation results**



**Figure S15. 7-day soil incubation results**



**Figure S16. 30-day soil incubation results**



**Figure S17. Variable Importance Plot – Soil pH Random Forest Model**

Type of random forest: regression

Number of trees: 500

No. of variables tried at each split: 3

Mean of squared residuals: 0.04403721

% Var explained: 88.56

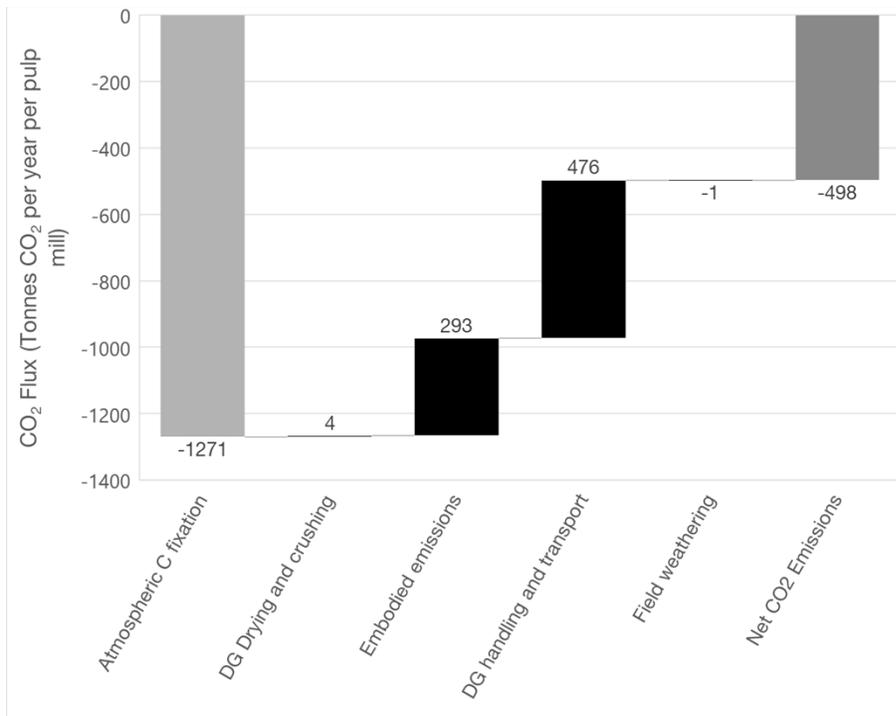
**Figure S18. Random Forest Model Out-Of-Bag Estimates**



**Figure S19. Random Forest Model Out-Of-Bag Error Plot**

<b>Soil</b>	<b>Initial Carbon (%)</b>	<b>Initial Absolute Carbon (g-C/kg-soil)</b>
GA1	0.537	5.37
GA2	0.618	6.18
TR3	0.654	6.54
RY1	2.519	25.19
AL2	0.525	5.25
TR4B	1.194	11.94
RW19	1.733	17.33
TR5	1.478	14.78
TR4G	1.117	11.17
AL1	0.592	5.92
TR1	1.441	14.41
LA1	0.672	6.72
RY2	3.104	31.04
TR2	1.394	13.94
TR10	1.092	10.92
PC9	0.922	9.22
TR11	1.359	13.59
PC5	1.323	13.23
MF1	6.822	68.22
PC3	2.003	20.03
TR7	2.082	20.82
PC10	3.026	30.26

**Figure S20. Pre-incubation carbon contents of all 22 soils**



**Figure S21. Waterfall plot of annual life cycle emissions associated with the production of dregs and grits as a soil amendment without drying. Gray bars represent carbon drawdown, black bars represent carbon emissions to the atmosphere.**

## References

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