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Alkaline mineral residues from pulp mills as a sustainable and economical alternative to lime fertilizers

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Lime is typically comprised of calcium carbonate ($CaCO_3$) or calcium magnesium carbonate ($CaMg(CO_3)_2$) and is needed to maintain a proper pH in agriculture and forestry soils, but it represents a major cost to growers and results in significant greenhouse gas emissions due to the mining, crushing, and transport required. There is a need to identify existing sources of alkaline mineral wastes and assess their potential as sustainable and cost-effective alternatives to lime. Dregs and grits (DGs) are highly alkaline mineral residues from biomass pulp mills that form during the recovery of pulping chemicals and are mostly comprised of $CaCO_3$. For the first time, we assess the efficacy of industrially sourced DGs on 22 acidic soils across the Southeastern United States that are currently used in either agriculture or forestry operations. The application of DGs was found on average to result in the same soil pH response as calcite over multiple soil incubations (0 day to 120 days). An analysis of the carbon content of the soils after the incubation revealed that soils incubated with DGs experienced a statistically significant increase in soil carbon relative to control soils and those incubated with $CaCO_3$. The generation and application of DGs emits 0.35 tonnes of CO_2 for every tonne of DGs, which compares favorably to the 0.86 tonnes of CO_2 for every tonne of agricultural lime generated and $85 per tonne lime reported in the literature. DGs prove to be a sustainable and economical substitute for agricultural lime in the acidic soils of the Southeastern United States.

Sustainability spotlight

This study advances sustainable resource use by demonstrating that alkaline residues from pulp mills, known as dregs and grits, can replace conventional agricultural lime without sacrificing soil performance. By diverting an industrial waste stream from landfills and reducing emissions associated with lime mining and production, this approach promotes circularity across the agriculture and pulp and paper sectors. Life-cycle analysis shows up to a 60% reduction in CO_2 emissions, with potential for net carbon removal when air-drying is used. The work directly supports the UN Sustainable Development Goals on Responsible Consumption and Production (SDG 12), Climate Action (SDG 13), and Life on Land (SDG 15) by transforming industrial byproducts into low-carbon soil amendments that enhance agricultural and environmental resilience.

1. Introduction

1.1. Emission reduction

In order to achieve the climate goal of limiting global warming to 1.5 °C, society must achieve net-zero greenhouse gas emissions by 2050 according to most projections established by the Intergovernmental Panel on Climate Change (IPCC).[1] While the rapid development of renewable energy, electric vehicles, and carbon removal technologies provides hope that these goals can be achieved, there still exist several industrial sectors that produce uniquely challenging emissions to abate. Hard-to-abate sectors traditionally include iron and steel, pulp and paper, cement, mining, transport, and agriculture.[2] These sectors are inherently difficult to decarbonize for various reasons such as the natural production of methane from livestock in agriculture, the high-energy requirements for processes like mining, or the use and eventual disposal of biomass in pulp and paper production. As a result of these challenges, efforts must be made to embrace circularity and limit emissions wherever possible. Herein, we identify a unique opportunity to reduce emissions in the agriculture, pulp and paper, and mining sectors through the use of alkaline waste materials from Kraft pulp mills as a soil amendment in substitution for traditional agricultural lime. Twenty-two acidic soils from across the

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United States were incubated with calcium carbonate ($CaCO_3$) and a combination of dregs and grits (DGs) from a Kraft pulp mill to compare the impact on soil pH over a period of 120 days. A techno-economic and a life cycle analysis are included in order to determine the economic feasibility and environmental impact of replacing a portion of the agricultural lime market with DGs.

1.2. Soil acidity

Plants are typically most productive in soils that have a pH in the range of 6 to 7.5.[3] Below this range, soils begin to experience the negative effects of soil acidity. Soil acidity is caused by an overabundance of hydrogen and aluminum ions, which can occupy the exchange sites in the soil, cause leaching of critical plant nutrients, such as calcium (Ca^{2+}) and (Mg^{2+}), and lead to toxic accumulation of certain nutrients such as manganese.[4] The soils in the Southeastern United States are particularly acidic (Fig. 1), caused primarily by high rainfall resulting in weathering of soils and leaching of alkaline cations.[5,6]

Acidic soils can have a negative effect on belowground and aboveground biomass, and agricultural lime is commonly used to offset these effects.[4,8] Cotton is especially sensitive to pH, as an increase in soil pH from 5.0 to 6.0 has been shown to increase the crop yield by 30–60%.[8] While agriculture has historically utilized lime and fertilizers far more than silviculture, there are still reported benefits from forestry fertilization including an increase in the above-ground biomass and a decrease in tree mortality.[9] Loblolly pine, the most commonly planted tree in the U.S., has shown increased tree volume and height when fertilized with a combination of nitrogen and phosphorus, with a few studies indicating the additional benefits of lime.[10–12] In 2022, the U.S. agricultural sector used 13.5 million tonnes of limestone, resulting in the release of 2.9 million tonnes of CO_2 based on the established emission factor of 0.059 tonnes of C released per tonne of limestone applied.[13,14] This emission factor only accounts for C released during the dissolution of $CaCO_3$ following field application and does not account for the carbon released during mining and production of agricultural lime. The USDA reports that the average cost of agricultural lime in North Carolina is $85 per metric ton as of December 2024 and serves as a significant cost to growers (USDA, 2024). The combination of low soil pH, relatively high lime costs, and a high number of pulp mills makes the Southeastern United States an ideal location for sustainable alkaline substitutes.

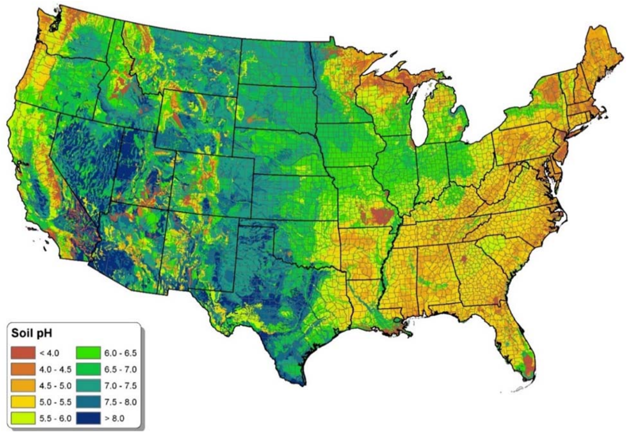


Fig. 1 Soil pH in the contiguous United States.[7]

1.3. Dregs and grits

In Kraft pulping of biomass for the production of pulp and paper, metal carbonates are purged from the recovery boiler and lime slaker resulting in the generation of DGs.[15] Pulping companies typically treat these highly alkaline materials as waste products, with 81% of mill waste ending up in landfills.[16] However, due to their high calcium carbonate equivalency (CCE), an indicator of a chemical's ability to neutralize acids, DGs present an ideal opportunity to employ circularity through application as a soil amendment. Not only could this reduce downstream emissions for the pulp and paper industry, a sector responsible for 1.3% of global industrial CO_2 emissions,[17] it can also reduce the carbon intensity of agricultural operations, a sector that is credited with over 20% of global greenhouse gas emissions.[18] There is growing evidence of down-stream carbon removal through the application of carbonates in agricultural soils.[19] Typically referred to as enhanced rock weathering (ERW), the process begins when CO_2 from the atmosphere binds to water to create carbonic acid. This acid then dissolves calcium and magnesium carbonates commonly found in agricultural lime (and DGs) to produce bicarbonate (HCO_3^-) ions. Depending on the chemistry and flow in local waterways, these ions can run off to the ocean where they remain for hundreds of years, resulting in atmospheric carbon removal.[20] Accounting for the carbon mineralized inside the recovery boiler and ERW of carbonates following dissolution, using DGs as soil amendment presents two potential pathways for atmospheric carbon removal (Fig. 2).[21,22]

The potential of DGs to serve as a sustainable and cost-effective alternative to commercial lime products in agriculture and silviculture is supported by the published literature, albeit in a limited manner. Cabral *et al.* (2008) conducted soil incubations using dregs, grits, and lime as amendments in one soil type with an initial pH of 5.5 and found that the pulp mill wastes provided a pH increase similar to the agricultural lime. The authors also determined that the metal content of DGs was not high enough to limit their usage as a soil amendment. Pöykiö and Nurmesniemi (2008), while not explicitly mentioning DGs, also proposed the use of calcium carbonate waste from pulp mills as a potential soil amendment. Pértile *et al.* (2017) applied dregs and lime to soybean and bean fields planted in acidic soil over a period of 6 years, and the soil pH and crop yields increased similarly for both amendment groups. Several other studies explored the use of DGs as a soil amendment and concluded that they are capable of neutralizing soil acidity and increasing the soil pH, and as a result are a viable alternative soil amendment.[23–25]

Previous studies have proven the efficacy of DGs to increase the soil pH in a similar fashion to commercial lime products

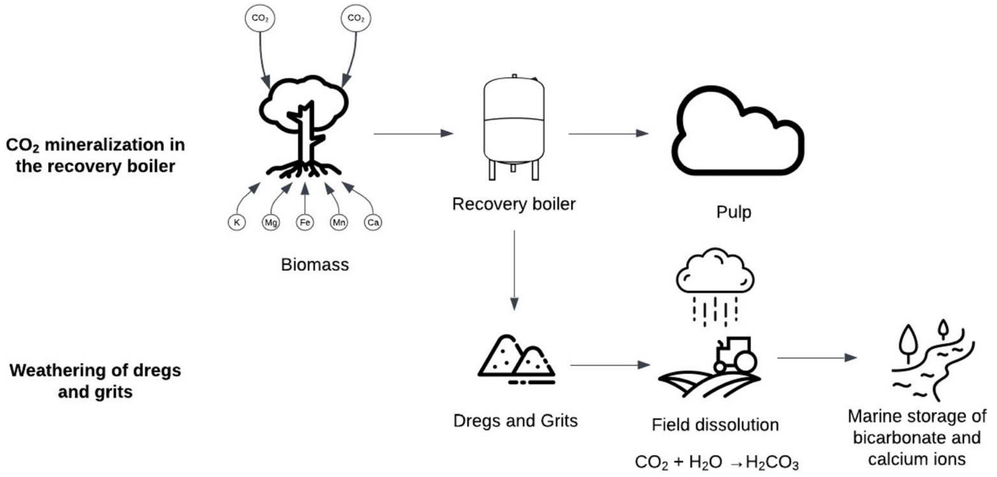


Fig. 2 Two pathways for DGs to remove atmospheric CO_2.

using a small subset of soils, and thus there is a need for a more comprehensive study that uses a larger variety of soil samples from a wide range of geographical locations. Furthermore, there is a need to understand the efficacy of DGs on agricultural and forestry soils in close proximity to pulp mills in the U.S., given that the U.S. is a major global producer of pulp and paper products with most of the production concentrated in the Southeastern states.[26] For the first time, we assess the efficacy of industrially sourced DGs on 22 acidic soils (average initial pH of 4.6) across the Southeastern U.S. that are currently used in either agriculture or forestry operations with a specific focus on soils used to grow Southeastern crop staples: cotton and loblolly pine. Each soil sampled was representative of common cotton or loblolly soils within 50 miles of an industrial pulp mill, thereby reducing transport costs and emissions, should the DGs be used in the future. Additional benefits of DG substitution include a reduction in energy intensive mining, less requirement of mineral treatment and crushing, and a lack of toxic heavy metals. We also assess the potential for carbon sequestration by tracking the carbon flux and performing life cycle assessment. Finally, we conduct a techno-economic assessment to understand the economic viability of using DGs as an alternative to commercial lime products.

2. Experimental

2.1. Soil collection and preparation

Twenty-two soils were collected based on GIS modeling analysis considering soil types, proximity to pulp mills, cotton cropland, and loblolly pine plantations. Soils to be used in incubation were first gathered from field sites that were selected to capture the most common low-pH soil types used in loblolly and cotton production in the area around each of the pulp mills. Methods for selecting and acquiring soils can be found in the SI.

A total of twenty-two sites were sampled in December 2022 and January 2023, representing five fields under recent cotton cultivation, sixteen current or former loblolly stands, and one sweetgum stand (Fig. 3).

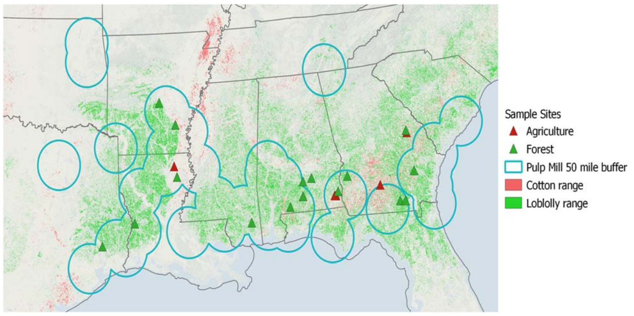


Fig. 3 GIS modeling used to identify soils. Sampling took place between November 2022 and January 2023.

Samples were taken from the top approximately 15 cm of soil from within a 20 m sampling zone within either the cotton field or the pine stand. Samples were dried at ambient temperature, sieved to 2 mm, and stored at 25 °C prior to use in incubations.

2.2. Material characterization

The DGs were provided by Westrock's Covington, Virginia pulp mill, which utilizes Kraft chemistry for wood pulping and chemical recovery. The calcite used was 99% extra pure calcium carbonate from Acros Organics with a particle size ≤ 50 microns. Commercial agricultural lime typically has a particle size ≤ 250 microns for at least 50% of the material with smaller sizes not usually being measured.[27,28] Since smaller particle sizes are correlated with higher reactivity, the $CaCO_3$ used in our trials is likely more reactive than traditional agricultural lime, thus resulting in a higher standard for the DGs to be compared to. We employed a range of analytical techniques and equipment for characterization of the amendments and soils including X-ray diffraction (XRD), scanning electron microscopy (SEM) coupled with energy dispersive spectroscopy (EDS), and nutrient and heavy metal measurement by the North Carolina Department of Agriculture (NCDA). Further information regarding the specific methods of characterization and additional data can be found in the SI.

Table 1 DG soil amendment calculations and assumptions

Input	Value
DGs generated during production of one ton of Kraft pulp (dry kg)	20.5 (ref. 15)
DG production rate in Southeast pulp mills (MMtDG/y)	0.67 (ref. 29 and 30)
Pulp mills analyzed for this study	44
DGs available per mill (wet tonnes per year)	15 154
Cotton acreage within a 50 mile radius of analyzed pulp mills (hectares)	644 859
Loblolly pine acreage within a 50 mile radius of pulp mills (acres)	7 116 114
Application frequency of cotton soil amendment (years between applications)	2 (ref. 31 and 32)
Application frequency of loblolly pine soil amendment (years between applications)	25 (ref. 33)
CCE of DGs (%)	94.1
CCE of $CaCO_3$ (%)	100
Application rate of DGs (t per acre)	0.49
Application rate of $CaCO_3$ (t per acre)	0.46

2.3. Amendment calculations

There were three amendment treatment groups for the soil incubations: a control group with no amendment added, a DGs group, and a calcite group. DGs were combined into one amendment group to simplify the proposed process of $CaCO_3$ substitution. The amount of amendment added to the soils was based on the amount of DGs available in the 44 Southeastern pulp mills analyzed by our group divided by the acreage of loblolly pine and cotton as well as theapplication frequency (Table 1). Thus, the dosage of DGs per unit area was equal to a hypothetical dosage wherein all available DGs from the 44 Southeastern pulp mills were used on existing cotton and loblolly pine soils within 50 miles of each mill (eqn (1)).

$$\text{Application rate (t per acre)} = \frac{\text{total DGs available (tonnes per year)}}{\left(\dfrac{\text{loblolly acreage within 50 miles of pulp mills}}{\text{years between application}}\right) + \left(\dfrac{\text{cotton acreage within 50 miles of pulp mills}}{\text{years between application}}\right)} \tag{1}$$

The calcite application rate was calculated and normalized based on the CCE of the DG amendment (eqn (2)).

$$\text{Application rate of calcite (t per acre)} = \text{application rate of DGs} \times \frac{\text{CCE of DGs}}{\text{CCE of calcite}} \tag{2}$$

2.4. Soil incubation methods

Soil incubation methods were selected from previous publications.[34,35] Each individual soil incubation consisted of 150 grams of soil added to a 16 oz. mason jar with a 1 cm hole drilled in the top to allow for gas exchange. Amendment was added to the soils and mixed with water to achieve a soil saturation of 80%. Once the soil, amendment, and water were added, they were mixed together with a spatula and left to sit on a lab bench top (Fig. 4).

2.5. Post-incubation testing

Following incubation, soils were dried at 50 °C. Once dry, the pH of the soil was tested in a 0.01 M calcium chloride ($CaCl_2$) solution. Following the 120 day incubation period, a LECO 828 carbon/nitrogen analyzer was used to determine the carbon content of the dried samples in an effort to quantify the increase or decrease in carbon content following amendment application and incubation.

2.6. Post-incubation calculations

The change in carbon content of soils incubated with amendments relative to the control group was calculated as a percent change (eqn (3)). The difference in carbon content as a result of DGs or calcite treatment was also calculated in terms of percent change (eqn (4)). The DGs contain roughly 9% carbon by mass, while commercial calcite contains roughly 12%.

$$\text{Percent change in carbon (\%)} = \frac{\text{carbon content} - \text{carbon content of soils with no amendment}}{\text{carbon content of soils with no amendment}} \times 100\% \tag{3}$$

$$\text{Percent change in carbon for DGs relative to calcite amendment groups (\%)} = \text{\% change in carbon for the DG group} - \text{\% change in carbon for the calcite group} \tag{4}$$

2.7. Statistical analysis

A linear mixed-effects model was used to quantify t(1) the change in the pH of soils relative to control soils and (2) the percent change in carbon relative to control soils after 120 days of incubation. We treated the soil type as a random effect and the amendment type as a fixed effect while using the lme4, lmerTest, and emmeans packages in R.[36,37] An alpha value of

Fig. 4 120 day soil incubation featuring 264 mason jars, 22 soils, and 4 amendment groups. The incubation durations tested were 0, 1, 7, 30, and 120 days. Soils were weighed every 7–10 days, and water was added to maintain the soil saturation throughout the incubation.

0.05 was used to compare the DGs and calcite results.[38] A random forest model was developed to determine the most important variables for predicting the change in the soil pH during incubation with DGs and calcite.[39] The following variables were selected for the random forest model: the amendment used on the soil, the initial density of the soil, initial carbon content of the soil, cation exchange capacity (CEC), the percentage of CEC occupied by basic cations, initial soil buffer pH, initial soil pH, humic matter, and texture of the soil.

2.8. Techno-economic and life cycle analyses

We developed a techno-economic analysis using Aspen Plus and Microsoft Excel software. The model simulates the preparation of one year's worth of DGs from one Southeastern pulp mill for field application and the transport of those DGs to agricultural fields. We also account for carbon drawn down from the atmosphere by tree biomass used for pulping (SI). The metals in the DGs are assumed to originate in soils that grew the tree biomass used for pulping (Fig. 5).

Components modeled in Aspen Plus include: a combustion reactor, a heat exchanger, a dryer, and a crusher, for drying and reducing the particle size of the DGs. Following the on-site preparation of the pulp mill DGs, a front-end loader moves the amendments onto trucks for transportation to nearby agricultural fields.

The physical and chemical characteristics input into the models are based on the DGs used in the soil incubations. The desired particle size is based on Rockydale Quarries' dolomitic agricultural limestone product.[40] Assumptions for the LCA are based on values found in the literature (Table 2).

We modelled the dryer in Aspen based on the heat duty required for drying the amount of DGs produced annually at a pulp mill. The crusher is designed to achieve a specified particle size distribution similar to that of commercial aglime. Natural gas was combusted, and the corresponding emissions were calculated through the use of a CO_2 emission factor for natural gas.[42] The electricity powering the crusher was purchased off-site with an emission factor reflecting that of waste wood combustion.[43] In order to calculate the costs of the system, the drying and crushing of DGs were treated and

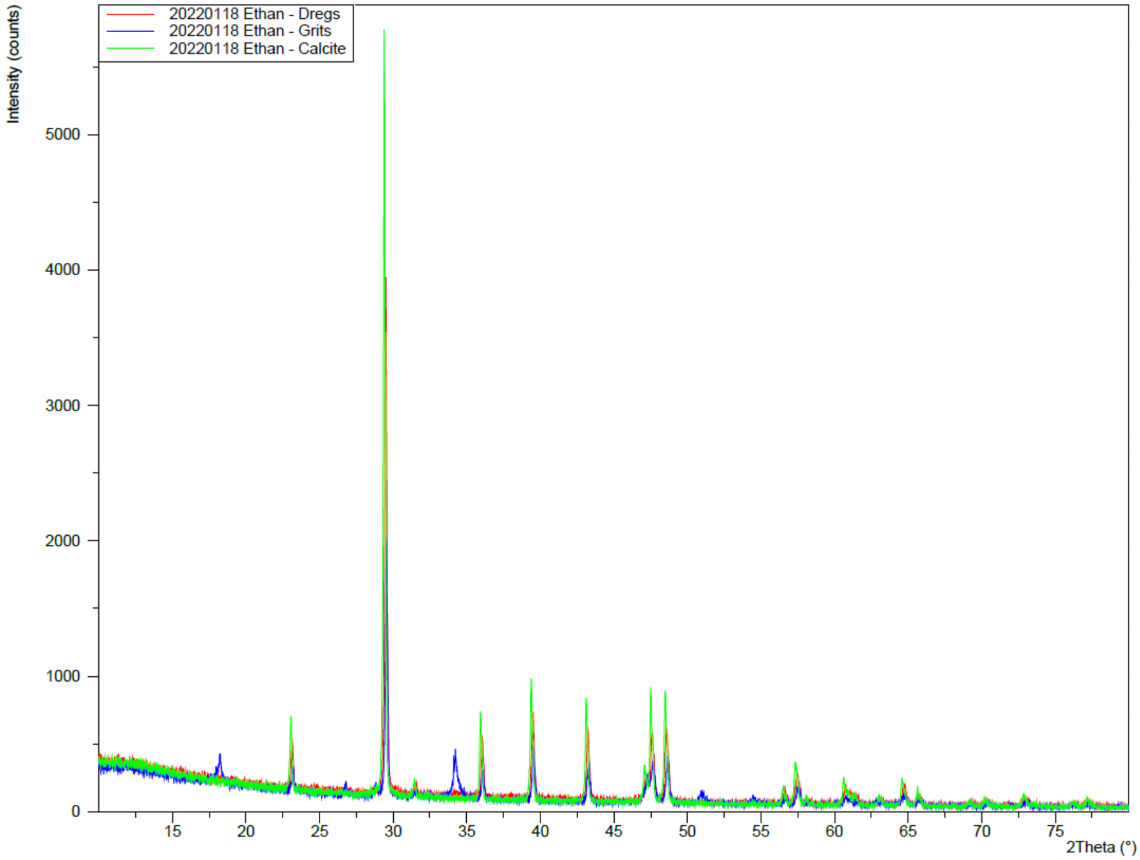


Fig. 5 Overlaid X-ray diffraction results demonstrating the similarity of dregs, grits, and calcite.

Table 2 Base-case LCA process model inputs and technical assumptions for drying, crushing, and transport of DGs

Input	Value
Dryer fuel	Natural gas[41]
Crusher energy source	Electricity
CO_2 emission factor of natural gas	52.91 kg per MMBtu (ref. 42)
CO_2 emission factor for electricity generated from waste wood combustion	93.88 kg per MMBtu (ref. 43)
Front-end loader emissions	0.696 $kgCO_2$ per m^3 (ref. 44)
Emissions from transport of DGs	88 gCO_2 per t-mile (ref. 45)
Embodied equipment emissions	0.32 $kgCO_2e$ per USD (ref. 46)

modeled as a new greenfield facility that would be added onto an existing pulp mill. The TEA was developed using information from the Aspen process model and assumptions from the literature (Table 3). Additional TEA methods and equations are provided in the SI.

3. Results and discussion

3.1. Amendment characterization

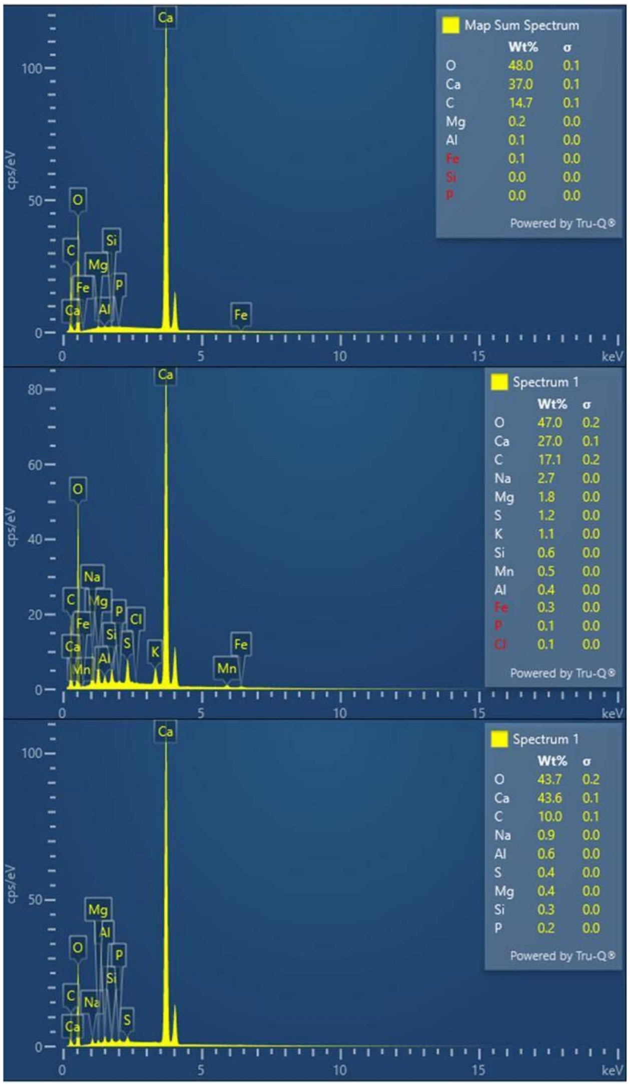
XRD results reveal that dregs, grits, and calcite have similar 2θ peaks, with the major peaks occurring at $2\theta = 23°$, 29°, 36°, 39°, 43°, and 48° (Fig. 5).

These peaks correspond to the typical strongest peaks for calcite and indicate that the materials have similar crystalline structures.[55] Meanwhile, SEM-EDS results highlight the similarity of the elemental makeup of dregs, grits, and calcite, with all three predominately (>90%) made up of oxygen, calcium, and carbon (Fig. 6).

The DGs have a higher amount of sodium due to the use of sodium hydroxide and sodium sulfate in the pulping process.[15] The presence of sodium in the DGs, however, does not exceed the ceiling sodium adsorption ratio (SAR) of 13, a measure of sodium relative to calcium and magnesium in a soil, as recommended by the USDA.[56] Since the SAR of the DGs themselves is below 13, repeated application over time will not cause soils to breach the recommended SAR ceiling of 13. The heavy metal content of the DGs is also below the concentration limits set by the EPA for the application of industrial waste materials, such as DGs from pulp mills, to soils (Table S1).[57] However, there are still risks associated with increasing the content of certain metals in soils through long-term reapplication. Accumulation of sodium in soil can lead to structural degradation and

Table 3 Base-case techno-economic assumptions for drying, crushing, and transport of DGs

Parameter	Base case value
Electricity cost	\$0.09 per kWh (ref. 47)
Natural gas cost	\$2.68 per MMBtu (ref. 48)
DG one-way transport	50 miles
Transport unit cost	\$0.159 per mile × t (ref. 45)
Rotary drum dryer purchase cost	\$400,000 (ref. 49)
Industrial crusher unit cost	\$20 per tonne per year capacity[50]
Plant utilization	90% (ref. 51)
Indirect capital costs	40% (ref. 52)
Annual maintenance costs	3% of total capital costs[53]
Capital cost contingency	7.5% of total capital costs[51]
Percent debt financing	0%
Percent equity financing	100%
Return on equity	10% (ref. 51)
Project life	20 years (ref. 51)
Capital recovery factor	11.7% (ref. 54)
Cost year	2025



Map Sum Spectrum

	Wt%	σ
O	48.0	0.1
Ca	37.0	0.1
C	14.7	0.1
Mg	0.2	0.0
Al	0.1	0.0
Fe	0.1	0.0
Si	0.0	0.0
P	0.0	0.0

Spectrum 1

	Wt%	σ
O	47.0	0.2
Ca	27.0	0.1
C	17.1	0.2
Na	2.7	0.0
Mg	1.8	0.0
S	1.2	0.0
K	1.1	0.0
Si	0.6	0.0
Mn	0.5	0.0
Al	0.4	0.0
Fe	0.3	0.0
P	0.1	0.0
Cl	0.1	0.0

Spectrum 1

	Wt%	σ
O	43.7	0.2
Ca	43.6	0.1
C	10.0	0.1
Na	0.9	0.0
Al	0.6	0.0
S	0.4	0.0
Mg	0.4	0.0
Si	0.3	0.0
P	0.2	0.0

Fig. 6 SEM-EDS results for calcite (top), dregs (middle), and grits (bottom).

a reduced infiltration rate, aluminum buildup in acidic soils can occupy exchange sites and harm root development, and iron toxicity can disrupt plant cellular function.[58–60] Previous studies have indicated that the metal content within DGs should not cause issues or exceed established limits, but long-term application of DGs could still lead to accumulation of metals.[61,62]

3.2. Soil pH

On average, the 22 soils experienced an increase in pH of 1.94 for both the DGs and calcite amendment groups (Fig. 7). We used a linear mixed-effects model and calculated a p-value of 0.997 when comparing the impact on pH for the DGs and calcite amendment groups. The high p-value indicates minimal or no evidence that there is a statistical difference between the two

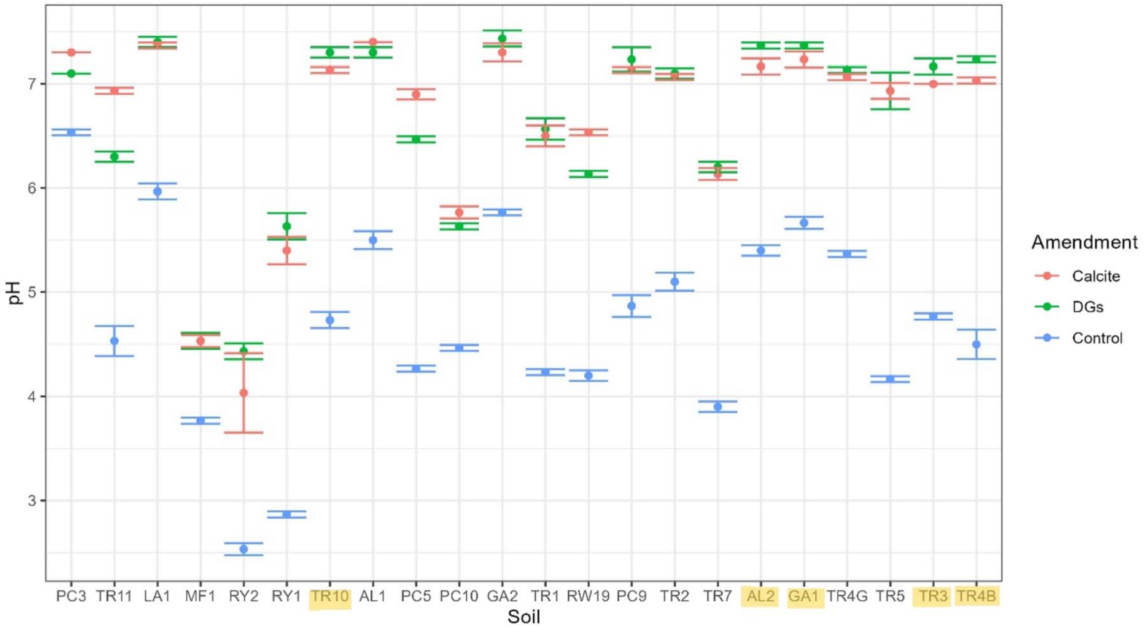


Fig. 7 Soil pH for 22 soils following 120 day soil incubation with each of the calcite, dregs/grits, and control amendment groups. Soils are ordered based on descending CEC (PC-3 has the highest CEC, and TR-4B has the lowest CEC).

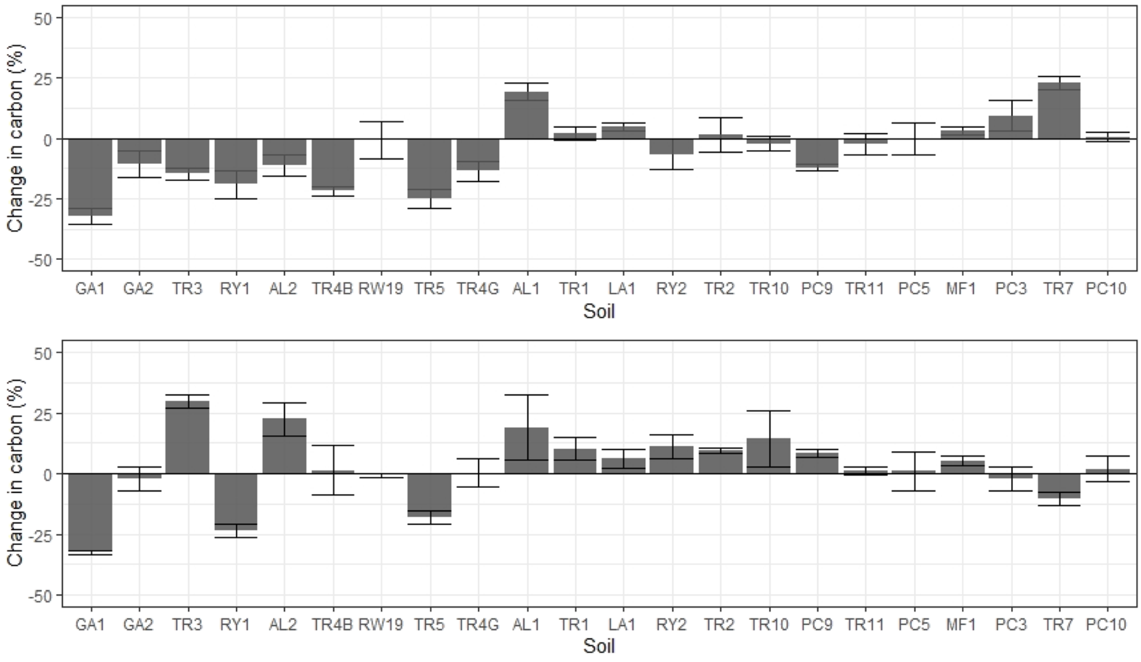


Fig. 8 Change in carbon content and standard deviation for soils incubated with calcite (top) and soils incubated with DGs (bottom) relative to the control. Soils are ordered based on the descending density (GA-1 is the most dense soil, and PC10 is the least dense soil).

groups in terms or their impact on soil pH following a 120 day soil incubation.[63,64]

Of the remaining 11 soils, 5 experienced a slightly higher pH increase when incubated with DGs than when incubated with calcite (highlighted in Fig. 7). Of these 5 soils, four of them have relatively low CECs compared to the rest of the soils. With lower CECs, these soils are less capable of adsorbing cations, thus resulting in a higher presence of positively charged ions in solution and should result in a lower pH. Both calcite and DGs contain calcium, but DGs contain additional cations such as magnesium and sodium. It is an unexpected outcome for the four low-CEC soils to experience a higher pH increase when incubated with DGs rather than calcite. However, the differences in pH change were minor (<0.10) and are likely not indicative of a noteworthy trend. The DGs and calcite amendments on average increased the soil pH by 1.94 units, demonstrating a similar efficacy to raise the pH. It should also be noted that real-world conditions experienced by crops and soils in the field differ from those that were tested in these incubations. Temperature fluctuations, nutrient leaching, runoff, crop nutrient uptake, and microbial dynamics can alter the impact that these amendments have on soils outside of a controlled lab setting. This study proves that in a controlled soil incubation, DGs are capable of achieving similar levels of soil pH increase to $CaCO_3$. The results for the other four soil incubation time periods and their corresponding pH plots can be found in the SI.

We developed a random forest model to predict the change in the pH of a soil when incubated with DGs or calcite. The model explained 88% of the variance in pH change following incubation, with the most important variables for predicting the change in pH being the CEC of the soil, the initial pH of the soil, and density of the soil. The variable importance plot as well as out-of-bag error estimates can be found in the SI. CEC is the capacity of a soil to adsorb and retain positively charged ions such as calcium, magnesium, or zinc.[65] A higher CEC represents a greater potential for a soil to retain cations, including hydrogen ions on exchange sites.[65] Limiting the number of free hydrogen ions increases the pH buffer capacity of soils, thus explaining the reduced pH change in soils with a relatively high CEC. The initial pH impacts the amount of pH change that can realistically be expected, as soils with a lower pH may experience a higher pH increase than those with a higher initial pH (Fig. 7). Lastly, soil bulk density impacts the infiltration of water and nutrients into pore spaces, thus potentially limiting the transport and adsorption of ions from dissolved soil amendments.[66]

3.3. Soil carbon

We measured the carbon content before and after the 120 day soil incubations to identify any differences in carbon retention for the DGs and calcite soils. Out of the 22 incubated soils, 6 experienced an increase in carbon content relative to the control soils when incubated with calcite, while 11 experienced an increase in carbon content when incubated with DGs (Fig. 8). The remaining 11 soils experienced no change in soil carbon or a decrease in soil carbon, potentially due to the increase in pH resulting in elevated microbial activity and increased decomposition of organic matter in the soils.[67]

On average, the DGs group experienced an increase in soil carbon content of 2.41%, while the calcite soils experienced a decrease in soil carbon content of 4.88%. For additional context, the pre-incubation carbon content of the soils originally ranged from 5.30 g-C per kg-soil (AL-2) to 68.2 g-C per kg^{-1}-soil (MF-1). Our analyzer did not differentiate between organic and inorganic carbon fractions; therefore, our reported values are the total carbon content of the soils. Pre-incubation carbon content of all soils can be found in the SI. We used a linear mixed-effects model and calculated a p-value of 0.004 when comparing the impact on soil carbon for the DGs and calcite amendment groups. The low p-value provides significant evidence that there is a statistical difference between the two groups in terms of their impact on soil carbon following a 120 day soil incubation. The difference in the impacts on soil carbon indicates that substituting DGs for calcite could result in

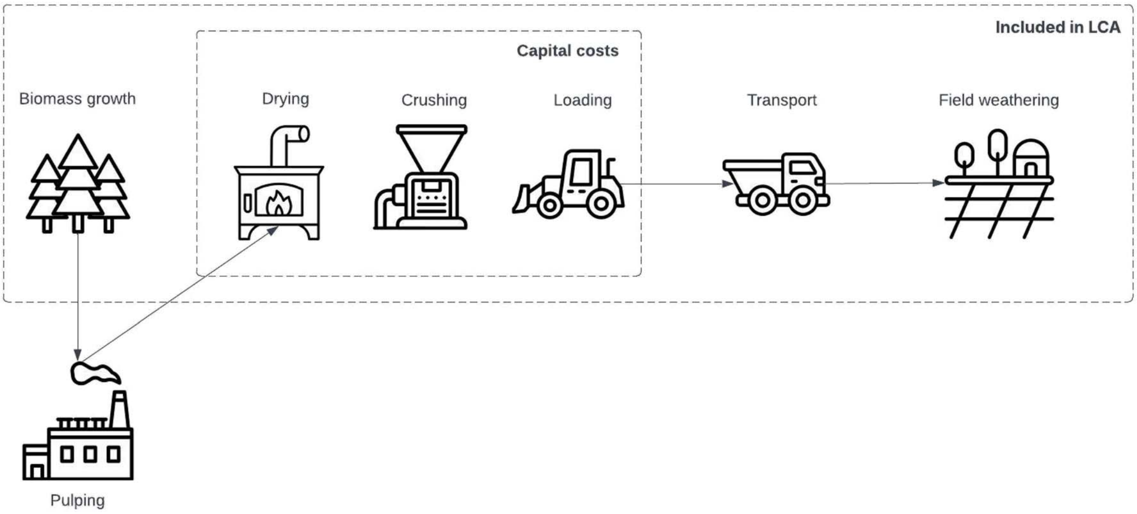


Fig. 9 Life cycle analysis boundaries.

a positive increase in the carbon content of agricultural soils. While this is a promising result, the controlled setup of the incubation experiments and lack of gas flux measurements must be considered, and field trials with gas measurements should be conducted before making conclusions about the short- and long-term carbon sequestration potential of DGs.

Similar soil incubation studies have yielded results that indicate liming can have a negative or neutral net impact on soil carbon.[68,69] However, there are multiple variables at play when analyzing the impact of liming on soil carbon mineralization, which has led to conflicting results in the past.[70,71] Liming adds inorganic carbon and increases the soil pH, which can increase the microbial efficiency and consumption of soil organic carbon (SOC), or liming can improve the soil structure, thus resulting in more secure stocks of SOC.[70] In the case of DGs and calcite, the primary difference between the two materials is the presence of metals such as aluminum and iron in DGs, which have been shown to decrease the respiration rates of soil microbes.[72,73] Substituting DGs for calcite provides an opportunity to limit microbial respiration and reduce the amount of carbon released into the atmosphere from fertilized soils. This theory should be tested further in future field trials with gas flux and respiration measurements.

DGs performed similarly to calcite as a soil amendment in a controlled laboratory incubation, raising the pH of the 22 acidic soils by 2 pH units on average and highlighting the potential of DGs to serve as an aglime substitute, should future field trials prove fruitful. Soils incubated with DGs experienced a statistically significant increase in soil carbon content relative to control soils. The presence of metals such as aluminum and iron within DGs potentially decreased microbial respiration within the soils and limited carbon losses to the atmosphere. Future work should include field trials, incorporate the use of rainwater containing carbonic acid (H_2CO_3), and the measuring of soil respiration rates to quantify the impact on the carbon flux and soil carbon sequestration potential.

3.4. Economic and life-cycle analyses

In order to understand the economic feasibility and GHG emissions of substituting DGs for commercial lime fertilizers, we developed a life cycle analysis coupled with a techno-economic model for the preparation and transport of DGs.

3.5. Life-cycle analysis

The emissions calculated for the DGs were those associated with the pre-harvesting atmospheric fixation of CO_2, post-production drying, crushing, transportation, and those released during weathering of the DGs in agricultural fields (Fig. 9). The pulp mill was excluded from the life cycle analysis boundaries, since the emissions of the pulping process are attributed to the primary pulp products, not the DG waste products. This ensures that emissions are not double-counted.

Throughout the life cycle of one pulp mill's annual generation of DGs and their use as a soil amendment, 2595 tonnes of CO_2 were released into the atmosphere (Fig. 10), which equated to an emission factor of 0.36 tCO_2 per tDGs.

Based on previously discussed pH results, the 7375 dry tonnes of DGs produced yearly at each Southeastern pulp mill could replace 6973 tonnes of agricultural lime. Preparation of DGs as a soil amendment would emit 2595 tonnes of CO_2 per year or 0.35 tonnes of CO_2 per tonne of DGs. The IPCC uses an emission factor of 0.86 tonnes of CO_2 per tonne of lime produced.[74] Therefore, the production of 6973 tonnes of lime results in the release of 5997 tCO_2 per year. Substituting DGs for agricultural lime could result in a reduction in emissions of 3402 tonnes of CO_2 per year per pulp mill. Applying it to the 44

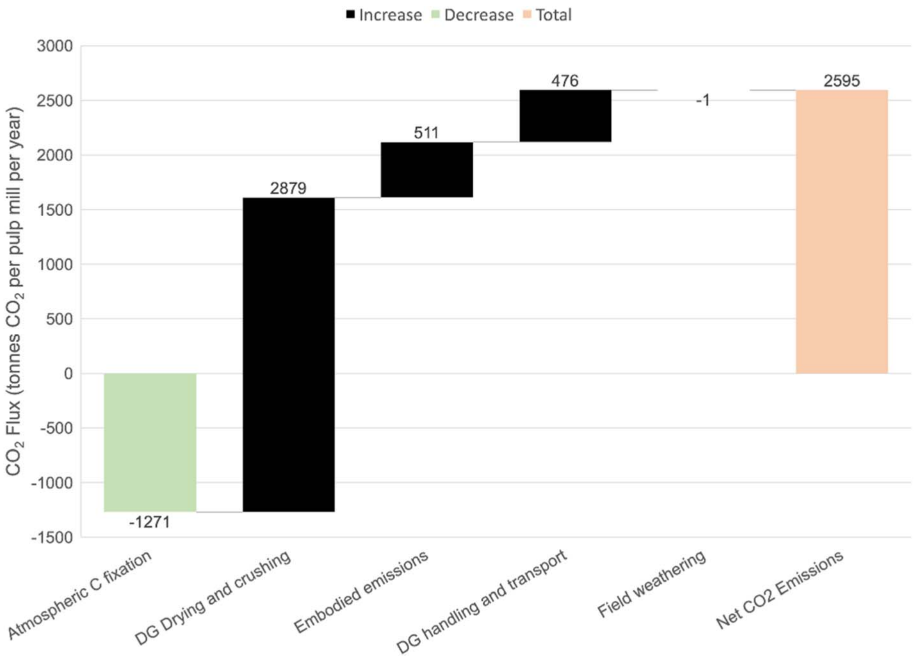


Fig. 10 Waterfall plot of annual life cycle emissions associated with the production of DGs as a soil amendment.

pulp mills included in the region of this study, this equates to 149 688 tCO_2 per year. While these results are most applicable to soils in the Southeastern United States within a 50 mile radius of pulp mills, DGs are still produced by pulp mills across the country. If DGs from all pulp mills in the U.S. were used as a replacement for agricultural lime, 329 994 tonnes of CO_2 per year could be avoided, but this value depends heavily on the incubation results translating to other climatic regions, soil types, and agricultural systems. However, it is also worth noting that if the drying portion of the DGs preparation process is removed and replaced with air-drying in a greenhouse, the emissions decrease significantly. Air-drying DGs instead of utilizing an industrial dryer results in the removal of 498 tonnes of CO_2 per pulp mill per year (Fig. S13), which equates to an emission factor of −0.07 tCO_2 per tDGs (Table S2). In the air-drying scenario relative to the industrial dryer scenario, a larger land area would be required, the drying process would take longer, particularly in the winter, labor costs and requirements would likely be higher, and there could be risks of contamination during the longer drying period. We did not attempt to quantify these impacts, thus the air-drying scenario results should be viewed cautiously and treated as a best-case scenario for low-cost preparation of DGs.

Table 4 Itemized costs associated with the post-production drying, crushing, and transport of DGs following generation at a pulp mill

Equipment costs	Installed capital cost ($)
Dryer	680 000
Crusher	250 750
Front-end loader	665 917
Contingency	167 650
Indirect costs	638 667
Total capital cost	2 402 983
Levelized capital cost ($ per tDG)	16.29
Operating costs	Annual cost ($ per year)
Energy	15 390
Transport	87 947
Labor	216 000
Maintenance	72 090
Net operating costs ($ per year)	391 425
Levelized operating costs ($ per tDGs)	53.08
Total levelized cost ($ per tCO_2 removed)	69.36

Since DGs are merely a byproduct of a larger manufacturing process, there are fewer emissions that are directly a result of their production. Therefore, when available, DGs from chemical pulp mills can serve as a sustainable substitute for agricultural lime, and potentially even as a net carbon sink when opting for slower but still reliable air-drying.

3.6. Techno-economic analysis

The techno-economic analysis included costs associated with the energy used during drying, crushing, and transporting the DGs, as well as the capital costs of additional equipment required to carry out these tasks (Table 4). The equipment used is a rotary drum dryer, an industrial crusher used for reducing the particle size, and a front-end loader used for moving the DGs into trucks for transport. When calculating the leveled cost, capital costs were divided by 20 years, which is a standard project lifetime in process modeling.[51]

Accounting for the equity and capital cost financing, the levelized cost of the DGs comes out to $96 per tonne. While this does not compare favorably to agricultural lime, which currently costs $85 per tonne, there does exist the potential to

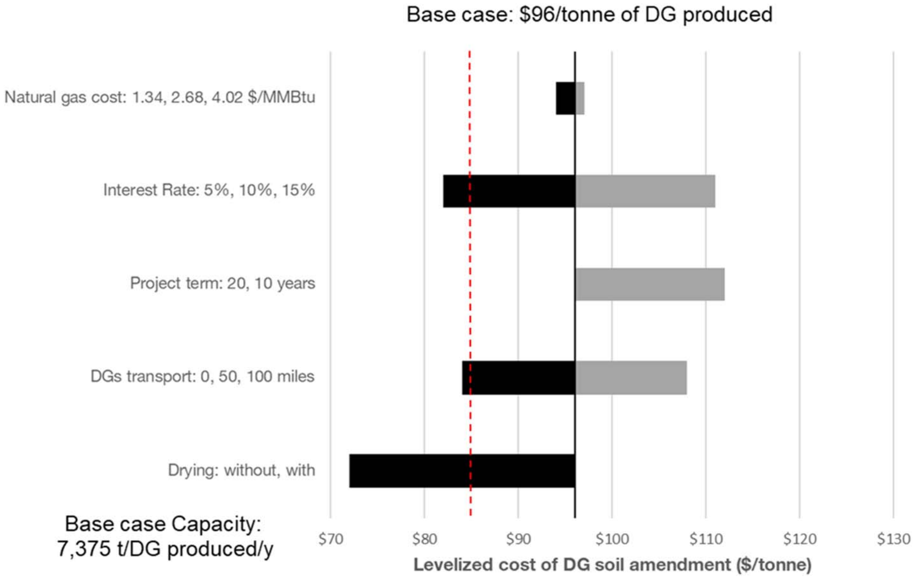


Fig. 11 Sensitivity analysis of the levelized cost of DGs soil amendment production as it is impacted by various parameters, the red vertical line indicating the current cost of aglime.

significantly reduce the capital and operating costs by removing the industrial dryer and relying on air drying instead, resulting in a levelized cost of just $72 per tonne of DGs in this optimistic scenario.[75] We developed a sensitivity analysis in order to analyze the impact of this proposed change, as well as other potential variable changes. A sensitivity analysis quantifies the impact of changes to the value of variables in a modelled process. We used a sensitivity analysis here to determine the impact of an optimistic and a pessimistic change to several assumptions. The assumptions we chose to alter and evaluate were the usage of industrial drying, the impact of increasing and decreasing the natural gas cost, the interest rate, the project term, and DGs transport distance. The base-case levelized cost of producing one tonne of DGs as a soil amendment was $96 per tonne (Fig. 11).

The removal of drying clearly has the largest impact on the cost of DGs production, reducing the levelized cost from $96 per tDGs to $72 per tDGs, below the cost of agricultural lime ($85 per tonne). However, pessimistic changes of these variables widen the gap between the two amendments. Therefore, it is critical for the cost and the emissions of the proposed process that air or greenhouse drying is employed rather than the capital and energy intensive process of industrial drying. There is limited research on the drying of minerals such as DGs in greenhouses, but the small particle size and low initial moisture content of DGs would likely allow for effective and relatively quick drying, albeit slower than industrial drying.[76] Assuming greenhouse drying cannot be employed due to land availability or climatic limitations, the relatively high levelized cost of $96 per tonne of DGs likely limits, but does not eliminate, the likelihood of pulp mills producing and growers purchasing DGs to replace agricultural lime.

4. Conclusion

Alkaline waste materials generated at biomass pulp mills, namely dregs and grits (DGs), closely resemble the chemical makeup of aglime, which is predominantly comprised of calcite ($CaCO_3$). In a 120 day soil incubation with 22 acidic soils from across the Southeastern United States, both the DGs and the calcite increased the soil pH by 1.94 pH units. We developed a random forest model and determined the most important variables for predicting pH change to be the CEC, initial pH of the soil, and density of the soil. The carbon content of the soils after the incubation revealed that the soils incubated with DGs experienced a statistically significant increase in soil carbon compared to those soils incubated with calcite. The presence of metals such as aluminum and iron within the DGs potentially reduced the microbial respiration in the soils, thus limiting the release of gaseous CO_2, whereas the pure calcite did not limit soil carbon losses.

Ongoing research is seeking to understand the impact on biomass growth for crops grown in soils amended with DGs as opposed to agricultural lime. Future research should include field trials wherein gas flux is measured, quantify the impact of DGs on the respiration rates of soils, and incorporate the use of rainwater containing carbonic acid in incubation trials.

We developed a technoeconomic and a life cycle analysis to calculate a levelized cost of DGs of $72 per tonne in the optimistic case and $96 per tonne in the base case. The use of an industrial dryer significantly drove up the cost of the process to $96 per tonne. Without industrial drying, $72 per tonne of DGs was favorable compared to the current cost of agricultural lime of $85 per tonne. We also calculated an emission factor of 0.35 tonnes of CO_2 for every tonne of DGs dried, crushed, and transported to agricultural fields, which compares favorably to the emission factor for agricultural lime of 0.86 tonnes of CO_2 emitted per tonne of lime produced. With air-drying, there is potential to remove CO_2 emissions from the atmosphere with a life cycle emission factor of -0.07 tonnes of CO_2 per tonne of DGs. Overall, when factoring in the increase in the soil pH, the change in soil carbon content, and the overall life cycle emissions, DGs prove to be a sustainable and potentially economical substitute for agricultural lime in acidic soils of the Southeastern United States.

Author contributions

Ethan Woods: writing – original draft, writing – review and editing, investigation, data curation, formal analysis, methodology. Andrew Trlica: writing – review and editing, data curation, formal analysis. Sean Bloszies: investigation, methodology. Perry Berlin: investigation, methodology. Alex Woodley: funding acquisition, methodology, resources, writing – review and editing. Rachel Cook: funding acquisition, methodology, resources, writing – review and editing. William Joe Sagues: conceptualization, funding acquisition, methodology, resources, supervision, writing – review and editing.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information is available. See DOI: https://doi.org/10.1039/d5su00814j.

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