



Scalable, economical, and stable sequestration of agricultural fixed carbon

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We describe a scalable, economical solution to the carbon dioxide problem. CO₂ is captured from the atmosphere by plants, and the harvested vegetation is then buried in an engineered dry biolandfill. Plant biomass can be preserved for hundreds to thousands of years by burial in a dry environment with sufficiently low thermodynamic “Water Activity,” which is the relative humidity in equilibrium with the biomass. Maintaining a dry environment within the engineered dry biolandfill is assisted by salt that preserves biomass, which has been known since Biblical times. A “Water Activity” <60%, assisted by salt, will not support life, suppressing anaerobic organisms, thus preserving the biomass for thousands of years. Current agricultural costs, and biolandfill costs, indicate US\$60/tonne of sequestered CO₂ which corresponds to ~US\$0.53 per gallon of gasoline. The technology is scalable owing to the large area of land available for nonfood biomass sources. If biomass production is scaled to the level of a major crop, existing CO₂ can be extracted from the atmosphere, and will simultaneously sequester a significant fraction of world CO₂ emissions.

carbon capture | sequestration | agriculture | anaerobic | biolandfill

For the past ~10,000 y, humanity has come to rely upon agriculture (1) for survival. Over the centuries, cultivation came down in cost, now requiring only a few percent of humanity, as farmers, to feed everyone. Agriculture is a form of solar energy. In 1980, Albert Rose, a television pioneer (2) and early solar energy advocate, pointed out that the commercial energy price, from combustion of cultivated plants, in dollars per 100MJ is competitive to the crude oil price in the same units (3). The competitiveness of agriculture is further validated by the commercialization of biofuels derived from farming (4).

In 1977, a famous physicist, Freeman Dyson, conjectured (5) that managed forestry could capture a significant portion of the CO₂ produced by the combustion of fossil fuels. In that early paper, Dyson did not invoke the most efficient crops. Moreover, the storage solution he proposed, a wet anaerobic peat bog, would unfortunately degrade to CO₂ and CH₄. Nonetheless, Dyson was among the first to recognize that carbon neutrality was insufficient; that we also need carbon negativity (6, 7). More recent proposals, that recognize the potential of agriculture (8), and forest lands to mitigate or offset global warming, involve harvesting woody biomass and burying it in trenches under a layer of soil (9), burying biomass in conventional landfills, or burying algae (10). All these approaches sequester biomass in anaerobic but wet environments. The biomass degrades to CO₂ and CH₄ by microorganisms that live in these anoxic and anaerobic environments (11). An inventory of greenhouse gas emissions shows that wet anaerobic storage would at best be greenhouse gas neutral (12), rather than negative. Amelse and Behrens (13) proposed low-cost biomass sequestration in conventional landfills in which some biomass degradation would occur, but had the insight that dry sequestration would slow anaerobic decomposition.

The most stable biomass component is lignin in wood, the remaining components being cellulosic carbohydrates which decompose more readily. Moist ancient wood, in anaerobic environments, loses much of its carbon, but sometimes retains much of its physical appearance. This preservation of physical appearance is due to the degradation resistance of the lignin component (20 to 40%), in wood. These conditions lead to at most a fractional sequestration credit, relative to dry Agro-Sequestration. This is discussed in *SI Appendix, section 1.4*.

It is now fully recognized that there is a pressing need for extraction of carbon from the atmosphere and for assured long-term sequestration for hundreds to thousands of years (14). Owing to the time urgency of climate change, we need low-cost carbon-negative solutions which can scale rapidly. Responding to this urgent need, nature-based solutions (15) such as forestation, management of existing forests, marine carbon, and carbon in soil and roots are being proposed to capture atmospheric CO₂. Nonetheless, there are significant questions concerning the duration of their long-term sequestration (16). Direct

Significance

This article presents a carbon-negative solution to the world's CO₂ emissions by stably sequestering carbon that has been photosynthetically fixed by cultivated plants. The technology buries salted biomass in a dry environment within an engineered biolandfill. The key to stable sequestration is the recognition that a dry environment assisted by salt preserves biomass. Preservation by salt has actually been known since Biblical times. Salt effectively reduces the relative humidity of the sequestered biomass, preventing decomposition for thousands of years. Current agricultural and biolandfill costs indicate US\$60/tonne of sequestered CO₂, corresponding to ~US\$0.53 per gallon of gasoline. A significant fraction of world emissions can be sequestered.

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Competing interest statement: Patents have been filed on certain aspects related to the sequestration procedure.

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air capture and CO₂ gas sequestration are in the early stage of development and deployment, with initial demonstrations having a ~US\$600/tonne cost for sequestered CO₂ (17). Many worldwide efforts are ongoing to lower this cost, one of which is the X-Prize Foundation's carbon removal prize competition which has already awarded 15 intermediate Milestone Prizes (18). Other comparisons of Agro-Sequestration favorable to a variety of carbon-negative technologies (including Bio-Energy Carbon Capture and Sequestration – BECCS) are presented in *SI Appendix, section 1.5*. Undoubtedly, many of the ideas being explored worldwide can work, and cost per tonne of sequestered CO₂ will become one of the key metrics. In this article, we present a technology with a projected cost of sequestered CO₂ ~US\$60/tonne, which we believe to be at the lower end of the range of costs for technologies being explored. Numerically (19), this translates to a premium of US\$0.53/gallon of gasoline. At this price, offsetting the world's CO₂ emissions would set back the world's economy by 2.4% (20).

The remainder of this article presents a technical option to extract CO₂ from the atmosphere, and to safely sequester it in biolandfills. About half the cost of this technology is based on the known agricultural economics of biomass farming as already worked out for the biofuel industry. The cultivation costs are further validated by real-life financial transactions every day at the Chicago Board of Trade (21). Of equal importance is the biomass sequestration step which will take place in dry engineered biolandfills located within the agricultural regions, minimizing shipping costs. A rough drawing of such a biolandfill is shown in Fig. 1.

Low humidity, parametrized by the thermodynamic quantity called *Water Activity*, is very important. If kept dry, anaerobic decomposition within the biolandfill is suppressed, and biomass can be preserved for hundreds to thousands of years. Biolandfills are designed using best principles for landfill construction to limit ingress of groundwater. One key innovation limiting *Water Activity* is salting the sequestered biomass. Salt limits the thermodynamic *Water Activity* (the biomass relative humidity), helping to keep the sealed biolandfill dry. In practice, there will be a trade-off between the salt cost and the cost of aggressive crop drying. Properly designed and operated biolandfills provide a way of verifiably and stably sequestering biomass captured from the atmosphere by cultivated plants.

Landfills are a widely practiced technology, whose costs are well known. Additional costs specific to biolandfill design are readily estimated. The biolandfill cost is roughly the same as the agricultural cost, each about US\$30/tonne of CO₂, leading to a total cost of US\$60/tonne, using known methods without technological risk.

Dryness and Salting to Eliminate Biomass Degradation

To eliminate biomass decomposition into CO₂ and CH₄, we follow principles used for long-term food preservation (22), namely sequestering biomass in dry conditions that prevent destructive microorganisms from growing. *Water Activity* (the relative humidity in equilibrium with the biomass) provides a measurement of the dryness level that prevents microorganisms from growing in sequestered biomass. In aerobic environments, a *Water Activity* above 0.95 will provide sufficient moisture to support the growth of bacteria, yeasts, and mold (23). Decreasing the *Water Activity* inhibits the growth of such organisms. For food stored in aerobic environments, if the *Water Activity* is controlled to be 0.85 or less in the finished product, the growth of organisms is sufficiently reduced so that it is not subject to the US Food and Drug regulations 21 CFR Parts 108, 113, and 114 (23). As the *Water Activity* further decreases, fewer and fewer life forms can grow (24) and their metabolic rate slows. Decreasing *Water Activity* below ~0.61 has been shown to extinguish life (25).

The reason that life forms suffer as *Water Activity* decreases is that living cells must transfer water-solubilized nutrients inward through the cell wall and water-solubilized waste materials out through the cell wall (26). Water content strongly bound to specific sites does not act as a solvent, while free mobile water can solubilize nutrients and waste. As *Water Activity* decreases, water only populates strongly bound sites such as hydroxyl groups of polysaccharides, the carbonyl and amino groups of proteins, as well as other sites where water can be held by hydrogen bonding, by ion-dipole bonds, or by other strong interactions (27). This binding action is referred to as sorption behavior and can be quantified by measuring water sorption isotherms, which is the equilibrium weight fraction of water taken up by biomass as a function of relative humidity. A plot of the equilibrium weight ratio of water to dry biomass, versus *Water Activity*, is shown in *SI Appendix, Fig. S1.2 A and B*. This same basic behavior also occurs in anoxic and anaerobic environments. Although the microorganisms that live in anoxic and anaerobic environments can be different from those living in aerobic environments, they still require the transport of water-solubilized nutrients and waste products across cell walls (28). As such, the water activity that will not support life will be similar for aerobic, anoxic, and anaerobic environments.

In a biolandfill, *Water Activity* will thermodynamically equilibrate, producing identical activities for water absorbed in the biomass, water vapor in the gas space, and water absorbed by salt. The ability of CaCl₂, MgCl₂, and several other salts to sorb water and lower *Water Activity* to below <60% is described in *SI Appendix, Fig. S2A–C*.

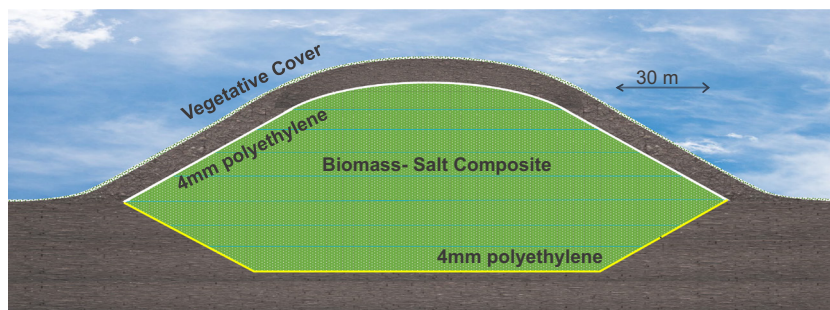


Fig. 1. A simplified version of the bio-landfill technology. It is essential to keep the biomass dry. A key role is played by dual layers of high density polyethylene adding up to 4 mm thickness, as a water diffusion barrier. In 1 y, <1.75 μm equivalent water thickness diffuses through. This rate of water diffusion can be accommodated for thousands of years by the dry salt-biomass mixture which can absorb the water without increasing its own relative humidity (*Water Activity*) above 60%. *Water Activity* remaining below 60% suppresses all life, and all bio-degradation. A more complete version of this biolandfill is in *SI Appendix, Fig. S2A–C*.

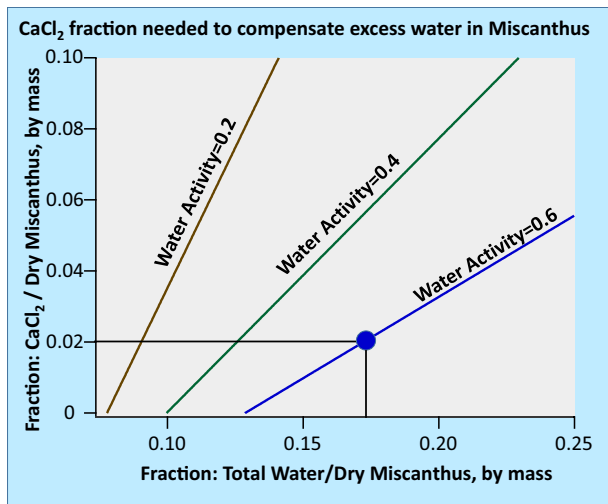


Fig. 2. A graph showing the CaCl_2 salt fraction needed to compensate the presence of excess water in *Miscanthus* biomass. This is derived from the water sorption isotherms shown in *SI Appendix, Fig. S1.2 A and S1.3 B*. The goal is to maintain a *Water Activity* < 0.6 , equivalent to 60% relative humidity, the blue line. If there is a 17% ratio by mass, total water in very wet *Miscanthus*, (represented by the big blue dot in Fig. 2), the required CaCl_2 salt/*Miscanthus* ratio required is 2%. CaCl_2 is an inexpensive road-de-icing salt, and would contribute $< \$3$ cost per tonne of biomass. To obtain a *Water Activity* of < 0.6 , drying *Miscanthus* to a ~ 0.12 water/*Miscanthus* mass ratio, eliminates the need for salt addition.

section 1.3. Water sorption isotherms for common salts are shown in *SI Appendix, Fig. S1.3 A and B*. We use these to determine the amount of salt that must be buried with the biomass to assure a low *Water Activity*, even in the presence of excess water, an example being shown for CaCl_2 in Fig. 2.

The lowest *Water Activity* that can be achieved with NaCl is 0.75, whereas with the two low-cost salts used for street de-icing (MgCl_2 and CaCl_2), *Water Activity* can be reduced below 0.6. Alternately, the salt can compensate for excessively moist biomass. A high treatment fraction of 2 wt.% CaCl_2 costing less than US\$3/tonne of biomass can easily obtain a *Water Activity* of less than 0.6. Significantly less CaCl_2 is needed with drier biomass (Fig. 2). Incorporating high fractions (such as 2 wt.%) of CaCl_2 into biolandfills would stretch world supplies, while options with drier biomass and less CaCl_2 can be scaled up without significantly impacting world supply (*SI Appendix, section 3*).

Biolandfill Design

Biolandfills stably store biogenic carbon by sequestering salted biomass composites within a dry tomb structure to prevent ingress of ground waters that would lead to decomposition, evolving greenhouse gases. Designs are based on current best municipal landfill practices, but with some notable enhancements.

Our reference design contains a dry tomb structure formed from layered ultra-low permeability base and cap structures (shown in *SI Appendix, Fig. S2 B and C*) that contain dual nested polyethylene water diffusion barrier geomembranes, surrounded by geosynthetic clays, geocomposites, and geotextiles. A schematic cross-section of a biolandfill with such a dry tomb structure sequestering dry salted biomass is shown in *SI Appendix, Fig. S2A* and a discussion of the design is presented in *SI Appendix, section 2*. The biolandfill remains dry because dual nested, 2 mm-thick layers of polyethylene diffusion barriers can limit water ingress to $< 1.75 \mu\text{m}$ of equivalent water thickness per year. Over a thousand year period, *SI Appendix, section 7* shows that the ingress of this amount of water is more than 400 times smaller than the amount of water that would have

originally been present in the dry biomass salt composite loaded into the biolandfill. *SI Appendix, section 2* also discusses landfill operational enhancements needed to temporarily store harvested biomass, chop, dry, and salt it, keeping the salted biomass composite dry while loading and compressing it in the biolandfill. In addition, *SI Appendix, section 2* discusses landfill design enhancements to make the biolandfill sequestration verifiable.

Owing to the 30 m biolandfill thickness, the landfill area is $\sim 10^{-4}$ of the agricultural area. Moreover, the top surface of the landfill can be restored to agricultural production.

Biomass Sources

Biomass can be purchased from independent farmers based on seasonal contracts. The potential to create new large-scale biomass sources has been investigated in connection with the creation of biofuels.

Crops suitable for agricultural carbon capture and sequestration include high productivity plants with dry biomass yields in a range from 4 to > 45 dry tonnes per hectare. Many of these are energy crops that the IPCC considered for generating biofuel feedstocks (29). A partial listing of candidate crops is presented in *SI Appendix, section 3.1*, and we estimate that in all, there are more than 50 potential high productivity crops. Examples that we used for our bottom-up technoeconomic analysis include *Miscanthus*, switchgrass, and loblolly pine.

The wide range of crops increases the breadth of applicability because feedstocks can be grown in diverse climates throughout the world. Many of these crops can be grown on marginal pasture and forest lands with reduced yields, but not competing with food production. The weight fraction of carbon in the dry biomass of plants considered ranges from $\sim 40\text{wt.}\%$ to $\sim 55\text{wt.}\%$, making it possible to sequester ~ 1.5 to 2 t CO_2 equivalent per tonne of dry biomass.

To provide comparisons, let us sequester approximately a half of the world's greenhouse gas emissions (~ 20 Gt of CO_2 equivalent per year). With the carbon content and yields/hectare given in the previous paragraph, biomass burial would require agricultural production of 4.8×10^8 ha, or equivalently 4.8×10^6 km^2 . This corresponds to 1/5th of the world's row cropland (30), or 1/15th the land area of all croplands, pastures, and forests (31, 32). More than half the IPCC's Shared Socioeconomic Pathway (SSP) models for greenhouse gas reduction by the year 2100 (33) call for comparable or larger land areas than 4.8×10^6 km^2 for biomass production.

There has been some difference of opinion about difficulties in creating such a large change in land use and its impact on food prices. In *SI Appendix, section 8*, we present a detailed regional analysis of Agro-Sequestration scalability including an analysis of how large biomass sources could be obtained in the United States. This assessment is based on the "DOE's 2016 Billion Ton Report" (34) and is deemed to be less controversial.

Carbon Efficiency and Economics for Stable Sequestration

We present an estimate of costs, based on both a bottom-up analysis of agricultural and sequestration costs and a different methodology based on observed prices in today's society. The detailed bottom-up estimate of costs for growing, harvesting, transporting, drying, and sequestering carbon in crops is in *SI Appendix, sections 4 and 5*. The simpler analysis based on observed current prices is as follows:

Agricultural costs can be estimated from data for yield and sale prices of major row crops, which are published by most national governments. In some countries, there are direct agricultural

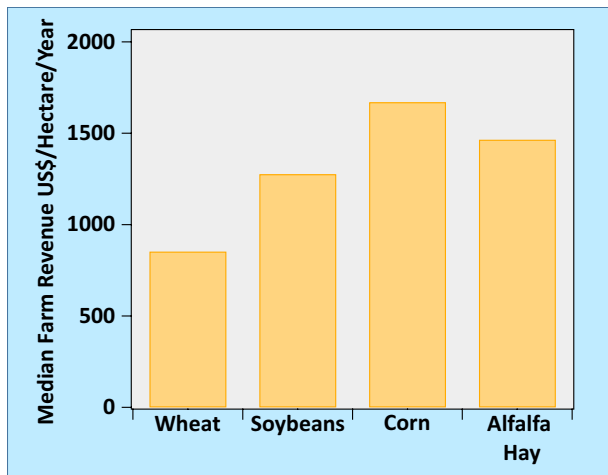


Fig. 3. We generally estimate costs through a bottom up analysis of all inputs. But there is a 2nd method shown here, the revenue received by farmers at the Chicago Board of Trade. We reason that the median prices received by farmers is an upper limit to the cost of production. Food attracts hungry parasites, requiring intensive farming, with costs of US\$850-US\$1450/hectare shown in the figure. Non-food biomass crops tend toward the lower end of that range.

subsidies, but in the United States, this takes the form of paying farmers not to farm. For the remaining farmers, the sale prices received must on average cover their costs. Fig. 3 is a bar chart of these costs reflected as median farm revenue per hectare for corn, wheat, soybeans, and hay. These are computed from the median of historical inflation-adjusted costs over the last 22 y for delivery of these crops to the Chicago Mercantile Exchange and crop yields per hectare. An estimate of agricultural direct air capture costs can be calculated from the median prices farmers received per hectare for row crops and then making the conservative assumption that energy crops would entail no extra costs relative to food crops. This methodology is detailed in *SI Appendix, section 6*. Using this methodology, we arrive at a representative agricultural direct air capture cost of US\$30/tonne of CO₂ for growing, harvesting, and shipping highly productive energy crops to local landfills.

Likewise, an estimate of biolandfill costs can be arrived at from an analysis of tipping fees at municipal landfills in the United States. The average tipping fee in 2021 was approximately US\$60/tonne (*SI Appendix, section 6*) with a variation ranging from +60% to -30% depending on location, licensing requirements, etc. A tonne of dry biomass contains carbon equivalent to ~1.83 tonne of CO₂. To compare costs for agricultural sequestration versus municipal landfills, differences in density of the fill must be taken into account. Compacted US landfills are reported to have a density in a range from 0.3 to 0.4 g/cc, while biolandfills can have densities >0.6 g/cc (35). As such, the biolandfill has lower cost per tonne of material buried compared to a municipal landfill. *SI Appendix, section 6* discusses the various factors in detail and arrives at a biolandfill cost of ~US\$30/tonne of CO₂. An estimate of sequestration costs would be ~US\$30/tonne agriculture + US\$30/tonne biolandfill = US\$60/tonne of CO₂ captured and sequestered.

The detailed bottom-up analysis for agriculture and biolandfills discussed in *SI Appendix, sections 4 and 5* supports these estimates and provides an estimate of the cost sensitivity that would likely be encountered. Fig. 4 shows results from the bottom-up analysis for agriculture and biolandfill cost structures for miscanthus, switchgrass, and loblolly pine.

Any carbon capture scheme must include corrections for the CO₂ emission during the sequestration process itself, such as fuel

burned in growing, harvesting, transporting, drying, and compacting biomass, as well as manufacturing the polyethylene landfill liner, etc. Moreover, a small credit must be given for carbon in soil and plant roots. These CO₂ emission penalties and credits are discussed in *SI Appendix, section 8*. This leads to the concept of carbon efficiency in Agro-Sequestration, which can range from 90 to 105% of the carbon sequestered in the biolandfill. Owing to the narrow range of uncertainty, further carbon efficiency precision would require analyzing specific projects.

Historical Evidence of Stable Agro-Sequestration

We are very fortunate that a natural experiment in Agro-Sequestration has emerged in the archeological record, confirming 2,000 y of safe sequestration. We have this information through the safe preservation and germination of ancient seeds (36). During the 1963 to 1964 excavations of Masada, an ancient fortress overlooking the Dead Sea (built in the second half of the first century Before the Common Era, BCE, but destroyed in 70CE), ancient seeds were discovered beneath the rubble, (37). On average, Masada has one of the driest climates on earth. Radiocarbon dating of the date seeds indicated that they were over 2,000 y old (36). The ancient seeds were planted. They germinated (36) and have now produced a number of date palm trees (38). A photograph of the tree called Methuselah is shown in Fig. 5. Preservation of other plant species is presented in *SI Appendix, section 1.1*.

Shortcomings and Next Steps

We have arrived at favorable conclusions regarding the cost, scalability, and long-term stability of Agro-Sequestration. Agro-Sequestration uses known existing technologies with known costs. It provides a practical path toward removing CO₂ from the atmosphere and solving the CO₂ problem.

Since farmers change crops from year to year, the scale-up time could be 1 y for some farmers, or longer, if ranchland has to be repurposed and redeveloped for growing bioenergy crops. These short timescales are important in view of the IPCC's call for immediate action. Agro-Sequestration can scale very quickly.

Nonetheless, there are shortcomings that need to be addressed:

a. The polyethylene moisture barrier must be carefully sealed around the landfill perimeter. This is easy in the laboratory, but quality control may be difficult in the field. Of course, the biolandfill will be thoroughly instrumented.

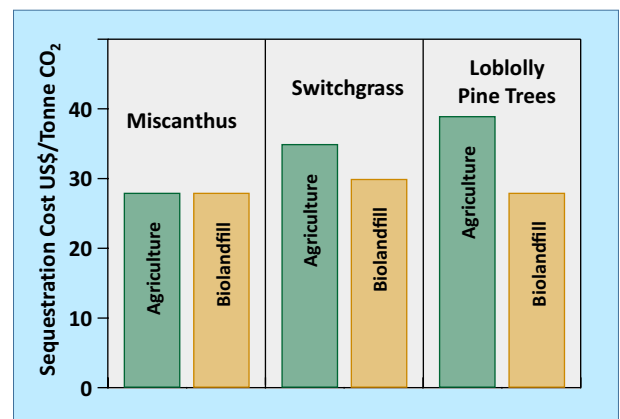


Fig. 4. A breakout of bottom-up farming cost and landfill cost for three different biomass crops, per tonne of CO₂ sequestered. The detailed analysis was done in *SI Appendix, sections 4 and 5*. A tonne of biomass sequesters ~1.83 t of CO₂.



Fig. 5. The Judean Date-Palm tree called Methuselah, of historical and cultural significance, germinated in 2005 from a 2,000-y-old seed, which had been stored in a dry location adjacent to the Dead Sea. (Photo acknowledgement, Guy Eisner).

b. Speedy biolandfill construction cycles and rain protection protocols have to be implemented to protect the dry biomass from atmospheric moisture. But, can they be implemented well enough?

c. It may take longer than expected for new crops to become part of the normal agricultural cycle, and impacts from large-scale repurposing of land will adversely affect large-scale deployment.

d. It is expected that successful biolandfills can be constructed with small additions of CaCl_2 ; however, if large amounts are deemed necessary, the supply would have to be significantly increased.

e. A regulatory framework must be adopted quickly, but will this be possible?

Some of the next steps are to reduce the unknowns in Agro-Sequestration technology which are listed below:

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