

This manuscript is a non-peer reviewed preprint submitted to EarthArXiv.

It is a working paper presenting the preliminary results of 2.5 years of enhanced rock weathering experiments in close-to-nature settings. We have shared this paper on our blog and decided to submit it to EarthArxIv because a formal scientific publishing process with proper peer-review usually takes at least half a year. Too long for new insights and scientific data that need to be shared quickly with the community of the dynamic carbon dioxide removal (CDR) research area of enhanced rock weathering (ERW).

Nevertheless, several scientists who are actively working on this topic as well as numerous experts from ERW companies have given their feedback as a form of peer review. Thanks to (in no particular order): Maria-Elena Vorrath, Karen Strassel, Benedikt Kratochwil, Jelle Bijma, Henry Liu, Adam Wolf, Lily Schacht, Johannes Barth, Philipp Swoboda, Mathilde Hagens, Philip Pogge von Strandman, Jacob Rietzler, Tim Jesper Suhrhoff, Mark Baum, Mary Yap, Dimitar Epihov, Jörg Rickli, Jens Hartmann and Thorben Amann for their input.

If you have comments or suggestions please contact us at <u>info@carbon-drawdown.de</u>.





Monitoring CO₂ Concentrations in Soil Gas: A Novel MRV Approach for Cropland-Based ERW?

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Abstract

This paper discusses the challenges we have faced in the past 2.5 years of field experiments trying to measure the speed of Enhanced Rock Weathering (ERW) in agricultural settings for removing CO_2 from the atmosphere.

Traditional measurements that allow quantification of carbon dioxide removal (CDR) in laboratory ERW tests include chemical analyses of the leached out soil water and monitoring with electronic pH and EC soil sensors. Unfortunately, application of these methods on our ERW field experiments has not given us any results, and it seems that it may take several more years for a relevant ERW signal to emerge. As an alternative quantification method, we used CO₂ sensors placed inside the soil in combination with automated CO₂ flux meters on top of the soil as a new way to assess the CDR effects resulting from ERW treatments in agricultural settings.

This novel approach could provide a faster way to assess the effectiveness of ERW in removing CO_2 from the atmosphere. And in turn, this would expedite ERW experiments as it could make large-scale/multi-variant experiments feasible and eventually become a new methodology for MRV (monitoring, reporting and verification) of ERW. This approach may also enable a faster collection of the large datasets that are needed for the creation, calibration and validation of accurate ERW models.



Part 1: How does CDR through ERW work?

Enhanced Rock Weathering (ERW) involves the use of rock dusts (such as basalt) on croplands to initiate Carbon Dioxide Removal (CDR). The fact that rock weathering pulls carbon dioxide (CO2) from the atmosphere is a well-known geological fact. For millions of years such rock weathering has shaped and stabilized the earth's climate. But this natural process typically happens on time scales of at least millenia. So for climate-relevant CDR we need to speed up this process to ideally a few decades and we should know how fast it can become under specific circumstances. It is however quite difficult to measure CDR resulting from ERW, especially outside of the lab. Nevertheless, CO2 removal through ERW can become one of the necessary steps to address the climate crisis if we succeed with both speeding it up and measuring it.

The climate-relevant effect of ERW on croplands is mostly an indirect CDR effect: rock-dust treated soils are expected to show a lower annual flux of CO_2 back to the atmosphere over their seasonal cycle. The amount of carbon that enters and leaves one square meter of cropland over one year is in the order of 750 g C. We expect a reduction in the annual C efflux by ERW to be in the order of 50 g C (pink and dark blue arrows in graph below). Measuring a CDR effect that is a reduction of less than 10% of the total carbon flux will be tricky given the countless heterogeneities and huge variances in a natural soil system.

For the following graph, data for "a hypothetical cornfield in a warm temperate region" was taken from Weil & Brady: "The Nature and Properties of Soils" (Chapter 12). Depending on the type of soil, climate, crop, irrigation, and farming methods the numbers can vary widely, but the basic concept of the soil carbon cycle remains the same. The graph below is from <u>our</u> blog.

An interesting fact is that the CO_2 concentrations inside biologically active soils are much higher (thousands of ppm, we measured over 40,000 ppm in some experiments) than in ambient air (currently at 420 ppm). This makes soils an excellent environment for enhanced rock weathering as the high CO_2 concentration speeds up the CDR process.



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The four step concept

Here is how CDR through ERW in soil works in four steps:

- Step 1: Rainwater mixed with CO₂ coming from the ambient air and produced inside the soil itself forms carbonic acid, which reacts with the rock dust and slowly dissolves the rock's components. The rock thereby releases its elements as ions that can now move and react with the other components of soil, water and air.
- Step 2: The ions change the soil chemistry and influence the soil processes which are affected by a cocktail of interconnected parameters, processes and players, including: grain size distribution, mineral content, waterflow and soil moisture, plant roots, fungi, bacteria, earthworms and other fauna, fertilizers, mineral precipitation and re-dissolution, DIC/DOC-balances, and - finally - the farmer's work on the field, e.g. plowing and tilling. Some of the ions interact with soil components - being adsorbed to clay particles or taken up by plant roots while other ions remain unbound and free in the soil water solution. Such free positively charged ions, the unbound cations, can neutralize dissolved carbon anions (mainly negatively charged bicarbonates) and effectively keep the carbon in solution while moving downward. This charge balance actively contributes to lowering the efflux of CO₂ gas from the soil to the atmosphere.
- Step 3: Eventually the "chemical signal" caused by rock dust amendments makes its way through the soil layers and can be observed in the leachate water below. Finding elevated levels of bicarbonate in this water is the rock solid proof that a CDR effect has occurred.
- Step 4: The bicarbonate anions coupled to rock dust derived cations are transported by soil-, groundwater and rivers to the oceans, where they are stored as marine alkalinity for tens to hundreds of thousands of years. In the long run, these added bicarbonates may also precipitate into solid carbonate minerals. Ultimately the whole process creates a permanent carbon sink that helps to solve the climate crisis.

Over longer time scales of decades to centuries, most of the rock's theoretical CDR potential that can be derived from stoichiometric calculations will have occurred. But for ERW to become a viable CDR business and to make a significant contribution to climate change mitigation, we need to know the actual speed of this process at a scale of years in order to create certificates that can be sold economically.

Within a specific soil and field, the interactions between all the players in step 2 are very complex and highly heterogeneous both in all three spatial dimensions and in time. The heterogeneity and variability between different fields or geographical regions thereby only increases. As these complex soil interactions are barely understood and subject of continued scientific research, one could argue that we may not be able to reach a thorough understanding fast enough to make ERW one of the potential solutions for the climate crisis.

For the MRV of CDR via enhanced rock weathering we need to know WHEN the actual permanent sequestration of CO_2 happens, i.e. when a CO_2 molecule is securely, when it is (at the human scale) permanently removed from the field's carbon cycle and cannot get back to the ambient air. Only then have we achieved true CDR that can be sold on the carbon markets.

Challenges of measuring

 For step 1 we can measure the loss of rock dust over a certain time using isotope dilution inductively coupled mass spectroscopy or ID-ICP-MS (e.g. LithosCarbon uses these measurements in their MRV approach, see this <u>preprint study</u> and some results below which already show results a few months after the rock amendment. But then we only measure how much rock has been dissolved without knowing if it dissolved due to carbonic acid, the combination of CO₂ and water, or any other acid that does not contribute to CDR. The theoretical, stoichiometrically calculated CO₂ capture potential of the already dissolved rock amendment is thus the upper limit of the actual CDR that can be achieved.

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- 2. In step 2 the rock's ions are still wandering around in the soil, "parking" in and out of the cation/anion exchange capacity (e.g. see "<u>Cartion park</u>"), interacting with fauna/flora and or precipitating/re-dissolving over time. But only those cations that are available in solution to associate with bicarbonate ions are accountable for CDR.
- 3. So only in step 3, when we measure elevated bicarbonate levels and/or elevated total alkalinity, we can be sure that the CDR effect has happened and that in step 4 the bicarbonate will reach the next river and eventually the ocean. The downstream losses in rivers and oceans are likely less important (see here and here).

So the information measured in step 1 can only give us a best case scenario estimation of the maximum amount of CDR. Then the processes in step 2 are hard or almost impossible to determine, and even if quantifiable they would require a lot of measurement points due to the soil's inherent heterogeneity. This is both too costly and impractical at large scale. In step 3, measurement of the ERW induced increase in cations and bicarbonates in the leachate water could give us a correct value of the CDR effect. Unfortunately, we do not know how long it takes until the chemical ERW signals become visible in step 3. It depends on several parameters and hence varies from one field to the next. It can take years, and in some cases we might not even be able to distinguish the chemical signature resulting from ERW from the strong background noise of the natural system's chemistry. As mentioned above, the urgency of the ongoing climate crisis does not give us years in order to understand if experiment variant A is performing better than variants B, C or D. We need much faster results of large-scale ERW experiments!

Let's solve it with a model (?)

Due to the complexity of the processes influencing ERW and the lack of data derived from actual field experiments, most of what we think we know about ERW is based on models. These models often include only a limited number of input parameters, involve lots of assumptions and simplifications and are barely validated through actual measurements – because there are hardly any large (public) ERW datasets for such verifications. Generating ERW datasets with lots of variations and replicas is therefore quite urgent and requires faster and cheaper measurement methods than the ones that have been available up to now.

What you are about to read

On the following pages we will show ERW data derived from

- 1. in situ field experiments,
- 2. large lysimeter experiments set up in open nature,
- 3. hundreds of smaller lysimeter experiments in a greenhouse (in Fürth/Germmany) and
- 4. simultaneous soil column experiments at the University of Hamburg.

These data will show you that after two years we are still not able to identify a clear "CDR signal" in soil water leachate chemistry and/or in EC/pH sensor data. A <u>sister experiment in Greece</u> showed quite similar results.

Although it is possible that we made unfortunate choices regarding our soils and rocks, the fact that we have the same situation of "no-signal" across several experiments makes this less plausible. Instead, it indicates that we need a much better understanding of ERW under (semi) field conditions. Our latest greenhouse experiment with - so far - 15 soils and 12 rock types is designed specifically to improve this understanding.

And there is also good news: our XXL lysimeter and greenhouse experiments seem to deliver a proper signal from CO_2 gas sensors buried at a depth of 15 cm into the soil. Even better: this signal becomes visible as early as days or weeks after rock dust is applied and mixed into the soils. Best of all? This ERW monitoring approach is available at a fraction of the cost of the still ongoing chemical analyses.



Based on initial observations, we seem to be able to qualitatively assess the weathering rate of different soil/rock variations and might be able to quantify the actual CDR rates in tons of CO_2 per hectare per year soon! By looking directly at the effect of ERW on the soil's carbon cycle – namely the reduction of CO_2 respiration from the soil – we circumvent the otherwise required understanding of the complex bio/geochemical soil processes and hopefully acquire a novel, straightforward MRV approach for ERW.

Part 2: Our experiments

Since we started working on ERW experiments in late 2020, we have built 4 major experiments: In March 2021 we started **our first open field experiment** in Fürth with two large plots treated with 40 t/ha and 240 t/ha basalt (<2000 microns) and an untreated control field:



In May 2022 we built **our XXL Lysimeter experiment**: 20 large water buckets (300 liters) were converted into large lysimeters and installed next to the older field experiment:



In May 2022 **a column experiment** with Fürth soil was set up at the University of Hamburg



In January 2023 we kicked off our greenhouse experiment with 350 lysimeter pots in Fürth:



In all these experiments we used a basalt that we call "Eifelgold", it is the product "Basalt-Urgesteinsmehl" from RPBL (<u>www.rpbl.de</u>) in two grain size distributions:

- Fine: Max. grain size 100 microns, 80% below 50 microns, 50% below 25 microns.
- Coarse: Max. grain size 2,000 microns, 80% below 850 microns, 50% below 400 microns.

As you can see most of our coarse material is much finer than 2,000 microns. The advantage of having 20% grains over 850 microns is that the material is easy to handle, it remains pourable, even if it gets wet. The fine Eifelgold clumps too much when it gets wet.

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Experiment 1: Field Experiment in Fürth 2021

Our longest running ERW experiment is a field trial which started in March 2021 and has been monitored throughout with electronic sensors (from vendor Dragino) for electrical conductivity (EC) and pH.

Below you see three circles, from left to right: Field C is the untreated control field. Field 1 received 40 t/ha in March 2021, Field 2 received 40 t/ha (plus some biochar) in March 2021 and an additional 200 t/ha in May 2022. Based on ERW literature studies and laboratory experiments, addition of the rock dust and its subsequent dissolution was expected to trigger an increase in both pH and EC. Until today, however, neither the EC nor the pH sensor data have shown any such signal. The fact that over time some sensors died, didn't really help, either.



Soil EC (uS/cm) and pH measured with soil sensors since the start of the field experiment. Different colors represent individual sensors within the same field (replicates); Field C: Control; Field 1: 40 t/ha in 2021; Field 2: 40 t/ha in 2021 plus 200 t/ha in 2022.





Experiment 2: XXL Lysimeter Experiment 2022

In our XXL lysimeter experiment we wanted to increase the chances of seeing an ERW chemical signature by using higher basalt amendment rates (100 t/ha, 200 t/ha and 400 t/ha). These rates are considerably higher than expected for agricultural deployments. We also tried to increase reproducibility of the data by (1) having 4 replicates for each treatment, (2) homogenizing the soil before it went into the pots, and (3) eliminating some of the variables linked to the open system of a natural field by putting the soil-rock amendments in large buckets.



This setup in so-called lysimeters furthermore allows collection of the soil water that has drained through the soil column. Such leachate waters were taken from the bottom of the tanks every 2 weeks for the first 3 months, and every 4 weeks afterwards. All water samples were analyzed for specific chemical parameters that could be indicative of ERW in the professional lab of Analytik Institut Rietzler, Fürth.

The graphs below show that none of these chemical parameters – the bicarbonate flux (which correlates with total alkalinity TA), the dissolved organic carbon (DOC) flux, the calcium (Ca) flux and the magnesium (Mg) flux as well as pH and EC – reflect a significant difference between the 4 treatments over the time period from May 2022 to February 2023.

Note 1: The spike in late August was caused by the first heavy rains after 8 weeks of drought (1st flush event after setup).

Note 2: "Flux" in these graphs is calculated by multiplying the measured concentrations in the leachate waters with the total amount of leachate water collected between sampling events. So it is actually "amount of stuff (in mg) that was flushed out from the soil column into the bottom of the lysimeters over 4 weeks".

As weathering of basalt releases bicarbonate, calcium and magnesium into the soil water, a simultaneous increase of these chemical parameters within the leachate waters of soils amended with rock dust would be the rock-solid proof of CDR happening. There is however no systematic increase of these elements, or a significant difference between the leachate water chemistry of the control and the rock dust amended soils. Yet! It is possible that it may still take several months, perhaps even years, for the desired "ERW signal" to reach the leachate water tank at 80 cm depth, due to inhibition of this chemical signal by cation exchange (adsorption-desorption of Ca and Mg to soil particles) and other buffering effects of the soil and/or precipitation somewhere on the way.

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XXL Lysimeter Project: chemical analyses of leachate waters; May 2022 - April 2023; shaded areas denote plus/minus 1 sigma of the variance of the replicas.

Of course, we also installed pH and EC sensors (Dragino) into each of the XXL lysimeters, 120 sensors in total. But when we study their data we can't identify an ERW signal yet, either. At least not a signal that we expected – an increase in pH and/or EC. The graphs below show that the pH at 30 cm soil depth in the

basalt amended lysimeters is similar or lower (instead of higher!) than that of the controls (shown with dots). Note: Some sensors broke down over the course of the experiment so their curves drop out of the chart.



Likewise, the electrical conductivity in the basalt treated lysimeters, which we would assume to increase, doesn't follow our expectations either: in most cases there is a lower EC in the basalt amended soils than in the controls (dotted).





Short Detour: Lithos method - How much basalt has dissolved so far?

Every few months we sent soil samples from our XXL lysimeter experiment to the team of our portfolio company <u>Lithos Carbon</u>. They analyze the soil samples using their novel isotope dilution ICP-MS method (<u>preprint study</u>).

These measurements tell us how much of the basalt has dissolved and "disappeared" from the top 10-15 cm of the soil. As CDR happens when basalt dissolves and resulting cations travel downwards in the soil, the measured amount of dissolved rock indicates the maximum amount of carbon removal that took place so far.

Our coarse Eifelgold is excellent for CDR through enhanced weathering: its stoichiometry reflects a CDR potential of over 400 kg of CO_2 per ton of basalt. It furthermore has a large fraction of small grains: less than 20% of the rock dust grains are larger than 850 micron. The CDR rates below, however, can not be assumed for just any similarly coarse basalt rock dust (up to 2.000 micron). In order to estimate a rock dust's CDR potential one needs to assess its chemistry, grain size distribution and mineralogy.



Lithos: Total tCO₂ CDR (error bars represent bootstrapped standard error)

The measurements by Lithos show that the upper bound of CDR effect, i.e. the maximum amount of potentially removed CO_2 , in the lysimeters is between 4 and 8 tons of CO_2 per hectare in 8 months. Interestingly, the 200 and 400 t/ha treatments do not show the double and quadruple of the CDR effect of the 100 t/ha treatment. So it seems that the ERW efficiency decreases when you apply more than 100 t/ha. But rates beyond 100 t/ha per year are not expected as usual ERW rates at scale anyway, we applied them just to make sure we would see "a CDR signal".



Lithos: Total CDR % completion (error bars represent bootstrapped standard error)

These first 8 months of weathering likely represent dissolution of only the fine fraction of the coarse bulk material. During this initial weathering, it is likely that originally jagged mineral grains become more smooth as they start dissolving. Weathering speeds – and hence CDR rates – can therefore not be expected to remain linear after the "initial weathering phase".

The XXL lysimeter experiment data show that over 8 months there was about 40-70 kg CO₂ removal per ton of added rock dust forthe 100t/ha treatment, approx 30-41 kg CO₂ removal per ton for the 200t/ha dose, and approx 18-20 kg CO₂ removal per ton of applied rock dust for the 400t/ha dose. This suggests that applying rock dust at four times the rate of 100t/ha is about $\frac{1}{3}$ as effective, and hence clearly not economical.



If we extrapolate the data of the 8 months long experiment to a full year we can assume a CDR effect in the order of 5-10 t/ha "potentially removed amount of CO₂" for our field in Fürth after a treatment of 100 t/ha coarse Eifelgold basalt. **We have a signal!** But where did the rock's dissolved components go? They have not yet reached the tank at 60 cm depth (see above). Did they precipitate in new minerals along the way? Were they temporarily stored in the soil's cation exchange capacity? So although over time this CDR effect will be consolidated, we do not know whether it is already permanent.

Experiment 3: Soil columns at University of Hamburg

In May 2022, Xuming Li and Jens Hartmann set up ERW column experiments with Fürth soil at the University of Hamburg. These soil columns were in ambient air and irrigated with a mix of tap water and deionized water at a rate of 800 mm/year to mimic an environment similar to the XXL lysimeter experiment.



The total amount of rainfall in 2022 at the XXL lysimeter project location was 600 mm. Since June 2022 we have measured 320 mm of rain and irrigated the experiment with another 160 mm of tap water.



Let's look at the graph showing the CO_2 consumption (" $CO_{2consrate}$ ") of the soil in tons of CO_2 per hectare per year: no significant difference between the treatments can be seen over 250 days, the treated soils even tend to show a lower CO_2 consumption rate than the control. Only the columns amended with the fine basalt seem to have a slightly higher CO_2 consumption rate than the control column without rock dust. Thanks to Xuming Li for sharing this graph with us.



Experiment 4: Carbdown Greenhouse Experiment 2023



We have built a greenhouse experiment that combines 15 German soils with 11 different rock dusts at various application doses (over 70 treatments, each in 4 replicas). Besides a setup that allows sampling of the leachate water for analyses we also installed soil CO2 sensors. To keep weathering rates high 365 days a year and to achieve an "ERW signal" as soon as possible we keep the ambient temperature above 18°C all year long, we irrigate the pots at a rate of 2000 mm/year or even 8000 mm/year and we use perennial plants (gras). This setup should allow us to identify the CDR effect as fast as possible and at lower application doses than in the XXL lysimeter experiment. This greenhouse experiment should furthermore enable us to understand the effects of different soils and rocks on the CDR effect. The greenhouse experiment is thus focused on speeding up and monitoring CDR through ERW, but it is not intended to mimic the processes in nature. Once it has shown that we can observe and actually measure CDR, we will return to nature with our ERW work.

As this experiment was set up only 4 months ago, there is only a little data of chemical analysis of the leachate water. Although it is too early to draw any conclusions from these data, it is interesting to note that seemingly the soils do not only differ in their physical characteristics but also in the chemistry of their respective leachate water.

More measurements and a longer monitoring period are however needed to confirm these initial observations. It takes a while for the soil column to settle, and all soil processes to be established after initial construction of the pots. As the soil's chemistry and physical processes are not stable in this initial phase, the first reliable results can only be expected in a few months at the earliest.

Our results: The unfortunate conclusion so far

Despite conducting field and lysimeter experiments for two years at Project Carbdown, we have not yet been able to identify significant differences in the dissolved load and leachate waters from soils treated with rock dust and the untreated controls (e.g. see <u>Schubert, C.</u> (2022). Counteracting global climate change-The <u>Carbon Drawdown Initiative</u>).

We thereby used various chemical parameters such as TA, pH, EC, cations, anions, DIC in leachate waters as well as electronic sensors for EC and pH. Even at high application rates, such as 400 t/ha basalt: no clear difference between controls and amended soils could be observed. The complexity of the different soil processes seems to stand in our way (e.g. see "<u>Cartion park</u>") and it could take years to see any ERW signal in the leachate waters. This poses an obvious problem for MRV, which in turn slows down the adoption and necessary scale-up of an all-new CDR industry that might become one of the solutions to the climate crisis.

Was our Fürth soil an unfortunate choice for our experiments? Did we select a "CDR blank", a soil/rock combination that doesn't work? Or didn't we wait long enough yet and the ERW signal will appear later on? Are other scientific groups doing ERW research luckier and do they see "ERW signals" due to more fortunate choices?

The point is: Just the fact that we have found this one soil/rock combination that either doesn't do CDR, or that takes too long to show a signal, is reason enough to fundamentally rethink the whole process so far. Everyone involved in the experiments' design, setup and execution expected at least some signal. As we only have speculations to explain our observations so far, maybe we just don't know enough about ERW in general?

No, wait...



Part 3: ...there is also good news: Hear our story about CO₂ sensors!

For the last 12 months we have been experimenting with CO_2 sensors in our ERW research. Initially we put a few in the leachate water tanks of the XXL lysimeters and they showed a first signal only a few days after we applied the basalt. Encouraged by these early results we put off-the-shelf CO_2 sensors into small "chambers", open at the bottom, and buried them 15 cm deep into the soil column of the XXL lysimeters. And these sensors also gave us a signal!

But let's start from the beginning.

The challenges of monitoring CO_2 in soil gas

Even though it seems a pretty straightforward approach to directly measure CO_2 in order to assess the CDR effect of rock dust on croplands, this hasn't been done extensively in the past – probably because of the following challenges:

- 1. ERW's CDR effect is indirect. Rather than directly removing CO_2 from the atmosphere, the rock reacts with CO_2 already present in high concentrations in the soil. Soil gas can contain several 10,000 ppm of CO_2 due to root respiration and bacterial decomposition of organic matter. In the soil, basalt, CO_2 and H_2O react and convert CO_2 into bicarbonates reducing the CO_2 level in the soil and with it the soil's CO_2 efflux (Fick's 1st Law, see below). What we actually need to measure is this reduction in CO_2 efflux (less CO_2 is respired from the soil when e.g. basalt is added) which has natural daily and seasonal variations. Quoting <u>Yan et al.</u> 2023: "CO₂ from respiration was sequestered".
- CO₂ concentrations in a soil can vary significantly within a day, week, month, season or year. Hourly measurements of a control pot and pots with different levels of basalt (100, 200, and 400 t/ha)

over seven days show that the data can vary by a factor of 2 within a day and from one day to the next (mostly due to the weather, changes in humidity or temperature).





As taking manual measurements every few days may not yield valid results and 24/7 continuous measurements are necessary, automated monitoring is required. To cancel out any weather/seasonal-related variations, it's thereby crucial to monitor controls and rock treatments simultaneously and use the relative difference between them as a signal.

- 3 Plants consume CO_2 throughout the dav (photosynthesis) while dark respiration of CO₂ goes on 24/7, with most uptake occurring above the soil and most respiration occurring at the roots. This process creates carbonic acid which dissolves rock, thereby making nutrients available to the plant's roots. Since these effects vary over 24 hours, using standard soil flux-meters does not seem to be optimal as they may not take these factors into account. Instead, we found that measuring CO2 concentrations in soil gas may be the better option.
- 4. Plant activity both above and below ground involves significant carbon transport, which can easily overwhelm the smaller CDR "signal". In one year, carbon in the order of 750 g can circulate through one square meter of soil, while the CDR effect is only about 50 g of carbon (see above and on our blog).

To tackle these challenges we use hundreds of automated, electronic CO_2 sensors that have been buried in the soil at 15 cm depth. The sensor's chamber-opening is thereby located at the bottom of



the soil-rock mix layer. These sensors send one data point every 60 minutes or every 5-15 minutes via LoRaWAN or WiFi (depending on sensor type), giving us at least 8760 data points per pot per year for each of the 350 pots.

Estimating the CO₂ efflux using soil CO₂ concentrations and Fick's First Law

We wanted to measure the CO_2 efflux of each of the 350 lysimeters in our greenhouse. However, working with 350 flux meters wouldn't be practical and the effects of plant activity, sunlight, etc. made this approach futile.

But one can also estimate the CO_2 efflux by looking at the CO_2 concentration in the soil gas, as discussed in the paper "Soil CO_2 in organic and no-till agroecosystems" (Lockhart et al. 2022).

If we neglect lateral CO_2 transport as it is generally minimal, we can use Fick's first law in one dimension. This law states that the flux (J) of a substance (in this case, CO_2) through a surface perpendicular to the direction of diffusion is proportional to the concentration gradient (dC/dy) of the substance in that direction:

J = -D * (dC/dy)

where:

J = flux [g/m²s] D = diffusion coefficient [m²/s] C = concentration [g/m³] y = distance in the direction of diffusion [m]

In our case we want to calculate the flux of CO_2 at the top of the soil and at a soil depth of 15 cm. Therefore, we need to know the concentration gradient of CO_2 between the top of the soil (where y = 0) and 15 cm (where y = 0.15 m). Assuming that the CO_2 concentration at 15 cm is C1 (in g/m³), and the CO_2 concentration at the top of the soil (i.e., x = 0) is C2,

the concentration gradient (dC/dy) is:

dC/dy = (C1 - C2) / (0.15 m - 0 m) = (C1 - C2) / 0.15 m

Therefore, the flux of CO_2 at the top of the soil (J) can be calculated as:

J = -D * (C1 - C2) / 0.15 m

and we have all necessary variables, except for D, to calculate the fluxes of controls and untreated variations:

$$J_{Treated} = -D \cdot \frac{(C1_{Treated} - C2)}{0.15 m}$$
$$J_{Control} = -D \cdot \frac{(C1_{Control} - C2)}{0.15 m}$$

where D is the diffusion coefficient of CO_2 in the soil (in m^2/s) which depends on various soil parameters and is hard to quantify, we will discuss D further below.

Qualitative comparison between controls/treated variations

For now we assume that D is the same for control and amended soil. This means we can calculate how much more/less the treated pots respirate relative to the control over a certain period of time (a qualitative comparison), as

$$Factor_{ERW} = \frac{J_{Treated}}{J_{Control}} = \frac{C1_{Treated} - 420 \, ppm}{C1_{Control} - 420 \, ppm}$$

i.e. the ratio only depends on the two CO₂ concentrations at 15 cm depth. For our carefully set up control/variation combinations and their replicas we assume for now that all other parameters – including D are (almost) the same for the related experiments (same soils, different rock treatments) so get canceled out and are constant over time (even the effects of plant activity).

If $Factor_{ERW}$ is below 1 for a given time span then the control has emitted more CO_2 than the treated pot, and therefore CO_2 or C has been stored either in the treated soil or in the water, as it can't go anywhere else (all other 5 directions except "up" are sealed off).



In our XXL lysimeter experiment we found that Factor_{ERW} is actually below 1 for all basalt-amended lysimeters over 9 months. Even in the recently set up greenhouse experiment we can already see distinct signals for some pots, with e.g. steel slag, after a few days or weeks (see data chapter below). This means: **We might already have measured the CDR effect by ERW** while our other measurements did not show a signal (yet) and might still take months/years to show this!

For a quantitative measurement we need the diffusion coefficient D and a little more work so we can calculate the actual amount of respirated CO_2 in g per surface area.

How do we get the diffusion coefficients D for all our pots?

The diffusion coefficient D depends, among other things, on the soil's grain size distribution and could therefore be changed by adding rock dust. Given the relatively small amount of rock dust added to the soil, our working hypothesis is that D changes minimally even for 400 t/ha amendment. The CDR effect from weathering of added rock dust is thus expected to be much stronger than the porosity change.

D can either be found by analyzing the soil or, and this will be our approach, by using flux measurements which will be done in our ongoing greenhouse experiment. This will give us J in the equation J = -D * (C1 - C2) / 0.15 m with which we can then solve the equation for D for various soil/rock combinations.

We did some initial flux measurement experiments to find out whether the CO_2 in the 400 t/ha coarse basalt pots is lower because its higher D would allow outgassing easier/more rapidly. We preliminary found that not to be the case (it would also contradict Fick's 1st law).

Due to natural intra-day variations of the soil CO_2 concentrations and day-to-day effects of the weather (sun, rain, temperature) on soil CO_2 concentrations, flux measurements need to be done automated 24/7 and must be run on controls and variations (4x control vs.

4x treated) at the same time (to cancel out any transient effects). Further and more sophisticated flux measurements will be done in the greenhouse to verify and quantify this.



The photos show our automated fluxmeter robot prototypes (V2, the one above, built with Lego; V3 was built with ESP32 computers and off-the-shelf IoT technology) in our greenhouse. They measure the flux 24/7 every 10 minutes and record the data into a cloud database.





Integrating/accumulating CDR data over time

As an MRV approach for ERW we need the cumulative effect of CO_2 efflux reduction over time. This can be calculated from the integral of the flux-difference of control and treated pots over time.

Based on $J = -D \cdot \frac{(C1-C2)}{0.15 m}$ we get m_{CDR} , the mass of collected CO₂ per field surface (in g/m²) as

$$m_{CDR} = A_{pot-surface} \cdot \int_{time} (J_{treated}(t) - J_{control}(t)) dt$$

which we can approximate as a summation like

$$m_{CDR} = 0,05147 m^2 \cdot \frac{24 h}{0.15 m} \cdot \sum_{1st \, day}^{last \, day} \left(-D_{treated} \cdot (C_{1treated}(day) - C_{ambient}) \right) - \left(-D_{control} \cdot (C_{1control}(day) - C_{ambient}) \right)$$

using our measured daily averages of CO_2 concentrations and $C_{ambient}$ (we calculate with 420 ppm). We can not use the 24 intra-day measurements as they are not sent synchronously by the sensors, so for now we use the daily averages until we have sensors that measure more often.

Variables are:

A = surface (m^2) J = flux (g/m^2s) D = diffusion coefficient (m^2/s) C = concentration (g/m^3) m = mass (g)

Our sensor measurements in ppm can be converted into g/m^3 using:

$$C1 [g/m^{3}] = C1 [ppm] \cdot 10^{-6} \cdot \frac{44.01 g/mol}{0.022454 m^{3}/mol}$$
$$= C1 [ppm] \cdot 0.00196 \frac{g}{m^{3}}.$$



Part 4: Actual CO₂ based data from our experiments

Now let's have a look at our CO2 data so far!

How we cheaply measure soil gas CO₂ concentrations in practice

We have been using the <u>Seeedstudio Sensecap CO₂</u> <u>LoRaWAN Gen 1 sensors</u> which are built around the <u>Sensirion SCD 30 sensor module</u> with a measurement range of 400 to 40,000 ppm and an advertised accuracy of $\pm(30 \text{ ppm} + 3 \text{ %MV})$. One Sensecap device costs ca. \in 100 and will deliver hourly data for years.

The Sensirion SCD 30 sensor module (\notin 45) is a dual channel NDIR CO₂ sensor which means it can be used over a long time without recalibration. It has a second chamber filled with a gas that is stable since its manufacture, does not change and whose infrared absorption properties are well known. By analyzing the subtle differences in the measurement results of that gas, in that chamber 2, the sensor firmware can calculate how much the measurements have deviated from what they should be and can apply a correction to the gas concentration measured in chamber 1.

Specifications

CO2		
Parameters	Condition	Value
Range	-	0 to 40000 ppm
Accuracy	400 to 10000ppm	±(30 ppm + 3 %MV)
Resolution	-	1 ppm
Temperature Stability	T = 0 to 50 $^{\circ}\mathrm{C}$	±2.5 ppm/°C

To understand the sensors' uncertainties, we put 8 uncalibrated sensors as they came from the factory in a chamber that started with a CO_2 concentration of 40,000 ppm and slowly dropped to 500 ppm over 18 hours. The resulting graphs below show that the differences between the sensor measurements are smaller at higher CO_2 concentrations. Between 40,000 and 8,000 ppm the difference between the sensors'

common average and any individual sensor was plus/minus 5-10% at most. For two sensors this relative difference increased further to 15% when CO2 concentrations continued to drop down to 2,000 ppm. At soil CO2 concentrations below 200 0ppm, the sensors' measurements can differ up to 25% and one would need to calibrate the sensors for quantitative work.

No pot in the greenhouse has shown soil CO_2 contents below 3,000 ppm and in the XXL lysimeters in the field we had readings down to 1,000 ppm only in winter time (when the differences between control, treated, ambient were very small anyway). Hence we believe that the uncertainties on the sensor measurements are not a problem for our project.



And as the lines on the graph are almost linear/straight, delta values between two values of the same sensor (e.g. for flux measurements) remain valid, regardless of how "off" the sensor is.

The issue with these particular soil sensors is that we can't recalibrate them once they are in the soil, we would need to destroy the soil column inside the lysimeter to get them out. So for the current sensor generation we need to make do with these uncertainties, for future experiments we will calibrate before using the sensors.

We generally have 4 replicas for each control/treated pot which will randomly include more and less accurate sensors, so we assume that their shared uncertainty is below plus/minus 10% for the rest of this paper. Further research with regards to the sensor measurement uncertainties will be done.





The sensor module was placed into a small container (covered by root fleece) which was then buried upside down into the soil (lower end at 15 cm). The soil gas CO_2 equilibrates with the chamber gas through the fleece and the sensor can pick up the signal.



The chamber with an open bottom retains an air bubble around the sensor in the event of flooding and protects the sensor from water this way.

Unfortunately the Seeedstudio SenseCAP Gen 1 sensors are no longer in production and Seeedstudio follow-up

Gen 2 product only goes up to 10.000 ppm CO₂ which isn't enough for soil gas monitoring. So we bought the remaining stocks of Gen 1 from shops all over Europe. In the future we will create our own soil gas CO₂ sensors.

To validate our cheap solution, a soil air specialist from environmental lab R & H Umwelt GmbH brought his high-end CO2 measurement instrument onto our field. He measured the amount of CO_2 in the soil at 10, 20 and 40 cm. The black bars show our time-synchronized electronic measurements in comparison to the high-end device's measurements (single measurements, no replicas involved):



We can see that our soil gas sensor at 15 cm (black bars) is easily in the range of plus/minus 10% of the professional measurement at 20 cm (yellow bars).

In order to stay in our budget, our concept is therefore to use many, cheap off-the-shelf sensors in multiple lysimeters and get CO_2 data every few minutes in a 24/7 manner (while keeping the related uncertainty in mind) rather than using just a few expensive, more accurate sensors that would not be able to cover all the pots simultaneously. The latter would simply not work with the transient nature of the soil gas CO_2 values anyway. We aim to beat uncertainty with a high number "n" of sensors, at least until we can show that this concept actually works.



CO₂ sensor data, part 1: XXL Lysimeter Concentrations

The XXL lysimeters were set up in May 2022, basalt was added late June 2022. The first 4 sensors were installed in the pots early August 2022 and another 16 were added in November 2022.

The following graphs show the daily concentration averages in ppm for the 4 replicas of each soil/rock treatment with their standard deviations (plus/minus 1 sigma/). Sigma thereby represents the variance of 24 measurements per pot over a 24 hour cycle, so it also covers intraday changes



The graphs show that the more basalt was applied, the lower the CO_2 concentrations in the soil are, which is to some extent caused by changes to the soil's porosity due to addition of the basalt grains. The differences between controls and amended lysimeters are thereby clearly higher than the 10% sensor uncertainty.

We can also see that, except for the green line for 100 t/ha and the blue line for control, the sensors' standard

deviation (= sigma, heterogeneity of the 4 replicas) areas are close to each other or even overlap. As over time the soil columns settle, the data seem to converge significantly and from here on we should be able to work just with the averages.

There are also CO_2 sensors installed in the bottom tanks of three lysimeters since June 2022 (albeit limited





to a max of 10.000 ppm) and these also indicate lower CO₂ contents for more basalt amendment.

CO2 sensor data, part 2: XXL Lysimeter CDR

In the first figure in this document we have shown that the annual amount of carbon respired by the soil in the form of CO_2 can be in the order of 576 g C per m², which equals 1,584 g/m²/a of CO_2 or 15,840 kg/ha/a. In a global comparison these CO_2 efflux values are however on the lower end of possible ranges, e.g. this <u>paper reports</u> ~80 t/ha/a with switchgrass and ~50 t/ha/a on bare soil at a humid subtropical location in Tennessee, USA.

To assess the reduction of this respiration due to ERW we would need to measure the actual CO_2 fluxes from the soil using fluxmeters throughout the year. This is rather hard to do – if at all possible.

Our hypothesis is that we can alternatively use the concentration of CO_2 in the soil at e.g. 15 cm as a proxy for this flux, following Fick's 1st law. But to estimate the soil CO_2 flux this way we need the diffusion coefficients of the rock dust treated soil. Since our fluxmeter measurements are not yet carried out, we do not have these diffusion coefficient data at present

So in order to demonstrate our concept we will work from here on with a diffusion coefficient D=0.008166

 $m^2/h = 0.00000230 m^2/s$ for our untreated soil, according to Jabro, et al. (2012): Estimation of CO₂ diffusion coefficient at 0–10 cm depth in undisturbed and tilled soils, Archives of Agronomy and Soil Science, (doi, PDF). Jabro also shows that values of D increase with higher porosity. For standard application rates of 5-100 t/ha we would not expect a notable change in D, but in our experiment we have also applied some exuberant amounts of rock dust. So whilst we assume an unchanged D for 100 t/ha, we consider a 5% higher D for the 200 t/ha treatments and a 20% higher D for the 400 t/ha dose (in the latter case ¼ of the top layer is our basalt dust, see photos).

So we can simplify the equation of m_{CDR} above and get a first estimate of the XXL lysimeters' CDR in g/m² and t/ha from the formula below:



XXL lysimeters, untreated soil



XXL lysimeters, soil with 400 t/ha treatment



$$\begin{split} m_{CDR}[\frac{g}{m^{2}}] &= 0,48 \ m^{2} \ \cdot \frac{24^{*60^{*}60 \ s}}{0.15 \ m} \ \cdot \ 0,00000230 \frac{m^{2}}{s} \ \cdot \ 0,00196 \ \frac{g}{m^{3}} \ \cdot \ \sum_{1st \ day}^{last \ day} (- \ (C_{1treated}(day) \ - \ 420 \ ppm) \ + \ (C_{1control}(day) \ - \ 420 \ ppm)) \\ &= 0,0012463 \ \frac{g}{m^{2}} \ \cdot \ \sum_{1st \ day}^{last \ day} (- \ (C_{1treated}(day) \ - \ 420 \ ppm) \ + \ (C_{1control}(day) \ - \ 420 \ ppm)) \\ &= m_{CDR}[\frac{kg}{ha}] \ = \ m_{CDR}[\frac{g}{m^{2}}] \ / \ 1000 \ \frac{g}{kg} \ * \ 10000 \ \frac{m^{2}}{ha} \ = m_{CDR}[\frac{g}{m^{2}}] \ * \ 10 \end{split}$$

Now let's look at the results from both daily and over time accumulated graphs. We have a reduction of CO_2 efflux when the blue areas (treatment) of the graphs are smaller than the red areas (=controls). The yellow graph shows the difference between the two, the daily (left column) and accumulated (right column) CDR effect.





Up to February, both the 100 and 200 t/ha experiments had relatively stable CDR amounts (yellow areas above) but since then these treated pots seem to increase their CO₂ respiration earlier than the controls (plants/bacteria?) – resulting in a diminished accumulated CDR effect in the past few months. As we do not know how this will further evolve and play out in the second year of the XXL lysimeter experiment, the importance of year-round CO₂ measurements is clear. The following graph compares the estimated effluxes and CDR effects based on soil CO_2 concentration to the potential CDR effects based on the rock dust dissolution measurements from Lithos Carbon.



Over 8 months we estimate a CO_2 efflux from the soil into the atmosphere of 9,265 kg for the controls, which is in the right order of magnitude of what we had expected (10-50 t/ha/a). The CO_2 efflux we estimate for the 100 t/ha treatments over the same time period was 8,669 kg, so the difference is the CDR effect of 596 kg/ha in 8 months.

The CDR effects estimated from the soil measurements by Lithos Carbon are about an order of magnitude higher than those derived from the CO₂ effluxes. This could reflect that the Lithos CDR potential calculated from the dissolved rock measurements is not yet fully delivered as a CDR effect. But although the CDR effects derived from these two methods are not in agreement, the true CDR effect is expected to be somewhere in between. Lithos' CDR potential calculated from dissolution rates has quite likely not been fully delivered as a CDR effect yet.

Do keep in mind that without reliable values for D and/or flux measurements, our above CDR effect values are rough estimates. The main uncertainty right now in our CO_2 efflux calculation approach are the D values, but we are in the process of defining these through measurements with CO_2 fluxmeters.



And there are some other aspects which we have simplified/disregarded for now but which may also impact the calculated amounts of collected CO₂:

- When converting ppm to g/m³ the day specific atmospheric pressure and the temperature are not yet included in the calculations (but we have this information in our database).
- We did not measure the actual D coefficients as this still has to be done using flux meters. And our estimated fluxes should also be verified by fluxmeter measurements.
- The accuracy of the sensors is plus/minus 30 ppm and 3% of the measured value.
- Changes in bacterial life due to a fertilizing effect from the basalt rock dust could also influence the CO₂ efflux, this needs further investigation.

In any case it is important to note that this CDR signal is already visible right after installation of the sensors into the lysimeters (August 5th), which is mere weeks after the basalt was applied on June 22th 2022. This is an exceptional find given that so far no other measurement approach has delivered any significant results from the XXL lysimeters over almost one year.

Could differences in plant growth explain the differences in CO₂?

In early October we measured the height of plants on the lysimeters (5 cosmea plants per lysimeter) and counted the blossoms to get a quick, first order estimate of the differences in biomass. We found no significant differences in the height of the plants (but treated pots seemed to have somewhat higher plants) and the biomass and stem width didn't seem much different either. The number of blossoms, however, was higher for most treated pots. But this can not explain a difference of up to 50% less CO_2 respiration from the 400 t/ha pots.



Total number of Blossoms (per Pot) Min, 20% and 80% Percentile, Max







CO₂ sensor data, part 3: Greenhouse pot experiment

Although the experiment in the greenhouse is just 12 weeks old at the time of writing, we can already show some interesting data from the CO_2 sensors. In this sneak peek we only discuss the relative difference in CO_2 concentration between treated pots and their respective control pots.

Percent values below zero indicate a reduction in CO_2 respiration (CO_2 sink), values above zero indicate that we may have created a CO_2 source. Every deviation above plus/minus 20% can be considered significant.

The first graph shows the data of nine variations containing soils from the Ortenau area in Germany.

These soils were taken less than 40 km apart from each other, some are even from the same farmer. They were all treated with the same dose of 40 t/ha basalt and it is astonishing how different these pots have reacted to the treatment so far in comparison to their untreated controls. For three soils we measure a steady increase of CO_2 respiration compared to an untreated control (much higher than sensor variance could explain), so these seem to be CO_2 sources. Three other pots show a clear sign of respiration reduction, also improving over time, and the remaining three don't show a significant signal. Although we don't have a clear explanation for this yet, we think farming practices and recent fertilizer activity may be involved.

Indicative CDR effect per variation (% delta of soil CO2 concentration compared to control)

Monthly Data: Feb 1st - May 10th (values<0 = CDR has happened, values>0 = we created at CO2 source)





The following 9 graphs show with more detail the intraday CO_2 measurements inside these pots with the farmers' soils for days 103-110 of the experiment. The gray areas with dotted lines show the CO_2 values in the untreated control pots. The yellow lines show the CO_2 values in the treated pots. When the yellow line is above the gray area the treated pot has a higher efflux of CO_2 , suggesting we might have created a CO_2 source by adding rock dust.

This large variation of desired and undesired outcomes from the same treatment is concerning and can not be fully explained by porosity changes. As long as we can not explain why this happens, it will be hard to avoid the same undesired outcomes in field deployments. Although we do not know how this situation will further develop over the next weeks and months, further investigation is necessary. Because these early results seem to indicate that under certain conditions ERW might result in CO_2 sources - quite the contrary of the intended CDR.





The next graph shows data for 4 different soils (with 2000 mm/a irrigation) and one set of highflow variants (8000 mm/a). The blue, red, yellow and green bars show the data for months January through to May and reflect the soil settling processes throughout this period of time.

As shown above, we currently assume a sensor uncertainty of ca. plus/minus 20% (to be further investigated). At only 40 t/ha application rate we do not expect changes in porosity alone to explain what we see here.



Indicative CDR effect per variation (% delta of soil CO2 concentration compared to control) Monthly Data: Feb 1st - May 10th (values<0 = CDR has happened, values>0 = we created at CO2 source)

The bars of this graph indicate once again that the type of soil makes a massive difference, e.g. the pots treated with Sibelco olivine on 5 different soils (x.7.x pots) show +75%, -6%, -34%, -10% and +36% of CO_2 concentration versus controls in the 4th month. As with the pots where the same basalt dose was added to different soils (previous graphs), we do not have an explanation yet as to why basalt rock addition seems to result in both carbon sinks and sources.

The greenhouse experiment furthermore includes three soil/rock treatments with 40, 100 and 200 t/ha basalt which so far suggest decreasing CO_2 concentrations with increasing basalt amounts. Finally, the steel slag treatments effused virtually no CO_2 in the first month (we measured soil CO_2 concentrations below 100 ppm for a few weeks after building the experiment!). However, later on two of them (5.10 and 8.8) have turned into CO_2 sources.



The final set of graphs below shows in more detail the intraday measurements in all the pots with Fürth soil and the LUFA soils from day 103 through to day 110 of the experiment. Coloured lines (treatments) above the gray areas (control pots) point to CO_2 sources.

As the experiment is just 100 days old, we can only see the early effects of ERW in these graphs. Nevertheless, it seems important to note that more than half of our greenhouse pots have been CO_2 sources since their setup. It will be very interesting to watch how this evolves over the next months.





Next Steps

There are a lot of unanswered questions. Our immediate next steps are:

- In the greenhouse we will use self-built automated robotic flux meters that measure controls and rock dust-treated pots in parallel for several days at a time so we can assess whether the soil CO₂ concentration is a valid proxy for the actual respiration rate.
- These measurements should also give us valid data for the D coefficients of our experiments, and this data should also indicate how it changes with different soil/rock treatments. This detail is especially crucial for the suggested calculations.
- Over time it should be possible to correlate this data with other MRV measurement methods such as assessing the remaining rock dust in the soil and leachate water analysis.
- We will need to gather many months of data in the greenhouse and in the field and with as many variations/replicas as possible. So we need many more soil gas CO₂ concentration sensors (self-built) and an IT system to gather and analyze data.

Other questions are:

- Is the rock dust added to the soil changing aspects of the system, e.g. oxygen levels, pH, that could also explain some of the CO₂ reduction in soil gas with reduced microbial activity (lack of O₂) or different CO₂ dissolution rate in the pore water (due to changed pH)? Are these effects permanent with regards to the CDR effect?
- How much of the changes in CO₂ concentrations is caused by more powerful/healthy plants and/or more active bacteria due to fertilization effects of the rock (as we see on steel slag pots with stronger plants)? If this is the case, are these effects permanent in a MRV perspective, or is the CO₂ reduction/increase only transient?

As soon as this proposed CO₂ sensor based approach proves to be giving us correct results, large scale experiments with lots of rock/soil combinations should be considered in open fields and in greenhouses. Also the potential effects of fertilizers, farming practices, etc. could be studied at a much faster speed than with traditional methods. Ideally this would help to build a large database of ERW data that can be used to construct, train and validate developing ERW models, so that in the more distant future most ERW treatments can be assessed with only model-based methods while still maintaining rigorous MRV standards.

All of this would help to speed up establishing the new ERW CDR industry, would make ERW carbon removal certificates more trustworthy and eventually support ERW in becoming an important asset in the fight against the climate crisis.