

Report on Permanence of Biochar

Perspectives from two recent publications and EBI conclusions

Bier, H. & Lerchenmüller, H. (2024)

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Foreword

Science is key to mitigating the climate crisis and the knowledge about carbon is key to understanding carbon removals.

As ground-breaking **new scientific papers** on **permanence of biochar** have been published in recent months, we considered it useful to initiate a broader discussion process on this topic to facilitate a common understanding between scientists from different disciplines, CDR policy makers and commercial CDR stakeholders.

We have therefore delved deeply into the topic of biochar durability and produced this report, which has undergone a **multi-step review process**. Many thanks to the members of the Review Board for their valuable feedback and constructive discussions.

With the publication of this report, we now invite a broader circle of **scientists**, **CDR policy makers** and **CDR stakeholders** to scrutinize our arguments and conclusions.

Harald Bier & Hansjörg Lerchenmüller March 2024







This report was prepared under involvement of a Review Board, responsibility for the content lies solely with the authors

Review Board

- Dr. Elias Azzi
- Dr. Alice Budai
- Dr. Anna Lehner
- Prof. Ondrej Masek
- Dr. Pål Jahre Nilsen

- Prof. Hamed Sanei
- Hans-Peter Schmidt
- Dr. Cecilia Sundberg

Scope of the Draft Review

- Challenging the line of thinking and arguments
- Support the authors in drawing conclusions and making recommendations on how to further communicate on this topic

Timeline

- 24.01.2024 draft report (V0.3) sent to Review Board
- 30.01.2024 1st video call with the Review Board with request to subsequent feedback
- 12./13.02.'24 discussions with Prof. Johannes Lehmann
- 20.02.2024 2nd video call with the Review Board; revised draft (V0.4) sent to Review Board
- 14.03.2024 publication of this report

Authors of this Report (EBI)

- Harald Bier
- Hansjörg Lerchenmüller

Responsibility for the content of this report is **solely** with the authors.

The individual **Review Board** members **provided** verbal and/or written **feedback** to earlier drafts. This participation does **not imply endorsement** of the report by the individuals concerned or their associated institutions. IPCC AR6 WGIII: CDR Factsheet



Carbon Dioxide Removal



CARBON DIOXIDE REMOVAL (CDR) refers to technologies, practices, and approaches that remove and durably store carbon dioxide (CO₂) from the atmosphere. CDR is required to achieve global and national targets of net zero CO₂ and greenhouse gas (GHG) emissions. CDR cannot substitute for immediate and deep emissions reductions, but it is part of all modelled scenarios that limit global warming to 2° or lower by 2100. Implementation will require decisions regarding CDR methods, scale and timing of deployment, and how sustainability and feasibility constraints are managed.

Centuries to millennia <i>(in soils and sediments)</i>								
CDR METHOD	Afforestation, Reforestation, Improved Forest Management		Soil carbon sequestration	Biochar	Bioenergy with Carbon Capture and Storage (BECCS)	Direct Air Carbon Capture and Storage (DACCS)	Enhanced rock weathering	Peatland and wetland restoration
IMPLEMENTATION OPTIONS	Agroforestry; tree planting, silviculture; timber in construction; bio-based products		Agricultural practices; pasture management	Cropping and forestry residues; urban and industrial organic waste; purpose-grown biomass crops		Solid sorbent; liquid solvent	Spreading crushed silicate rock	Rewetting; revegetation
STORAGE TIMESCALE	Decades to centuries (in vegetation, buildings, soils)		Decades to centuries (in soils, sediments)	Centuries to millennia (in soils and sediments)	10,000+ years (in geological formations)	10,000+ years (in geological formations)	10,000+ years (in minerals)	Decades to centuries (in vegetation, soils, sediments)
FINANCIAL COST (\$ per tonne of CO ₂)	Afforestation/ reforestation: \$0-\$240	Agroforestry and forest management: not enough data	-\$45-\$100	\$10-\$345	\$50-\$200	\$100-\$300	\$50-\$200	Not enough data
TRADE-OFFS and RISKS	Afforestation/ reforestation: Inappropriate deployment at large scales can increase competition for land (limiting land for biodiversity conservation and food)	 Agroforestry: limited impacts on agricultural crop production Forest management: if fertiliser use and introduced species are involved, risks include: reduced biodiversity, increased eutrophication, and upstream GHG emissions 	 Increasing carbon sequestration can occur at the expense of production Sequestration contribution per hectare is small and hard to monitor 	Negative impacts from dust Competition for biomass	Growing energy crops increases competition for land (limiting land for biodiversity conservation and food)	High energy requirement could lead to growing competition for low-carbon energy or increased GHG emissions. Some DACCS processes require water.	 Dust emissions Potential for increased GHG emissions from energy generation 	Some peatlands are used for food production, so could result in competition for land

https://www.ipcc.ch/report/ar6/wg3/downloads/outreach/IPCC_AR6_WGIII_Factsheet_CDR.pdf

This document has not been subject to the procedural IPCC review processes and has not been endorsed by the IPCC.



In **IPCC reports**, as well as in recent CDR reports and scientific papers, **biochar** has so far mostly been classified to be permanent for centuries to millennia, ... yet some publications talk about decades to centuries

We know of carbon that is millions of years old and carbon that clearly shows degradation in soils, how does this fit together?



At the surface of the earth crust one can find **fossil carbon** that is **millions of years old** and **charcoal** from ancient human settlements **thousands of years old** ...



... on the other hand, there is solid science showing that certain **biochars show relevant degradation** in surface soil environments.



The answer to the **question** of how to assess the durability of biochar can be found in two recently published scientific review papers

Two groundbreaking papers on permanence of biochar (Jan '24)



• Study shows 76% commercial biochar as pure inertinite,

• R_o analysis: reveals carbon pools, calculates

carbonization temperature for biochar stability.

exceeding benchmark.

 Non-linear models improved correlations between BC₁₀₀ estimates and properties.

GEODERMA

- A dataset of biochar incubations is made available along with code to analyse it.
- Curve fitting and data selection are key steps in biochar persistence modelling.
- The H/C ratio remains the main indicator of biochar

The Hydrogen to Carbon ratio (H/C-ratio) is an established proxy to determining Biochar's inertness



Hydrogen and Carbon are two elements found in all organic matter and their ratio is a key metric for biochar

The **Appendix on H/C** to this report provides some deeper insights on H/C ratios



1

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Capabilities and limitations of incubation experiments

2

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Capabilities and limitations of incubation experiments

Message from Azzi et al. (2024): dataset includes key gaps



- Azzi et al. (2024) includes a comprehensive analysis on all available incubation experiments made so far:
 - 134 individual observations
 - > 8,000 data points
- The supporting information includes a valuable compilation of all the measurement data and results of existing incubation experiments
- The authors conclude that the dataset on incubation shows significant gaps, notably lacking chars with
 - higher pyrolysis temperatures and
 - low H/C ratios
- The authors also highlight the **lack of** advanced characterization

H/C distribution of 79 datasets of historic publications (Azzi et al. 2024) shows high H/C values and a broad distribution



- 134 datasets were made available by Azzi et al. (2024), 49 datasets were excluded from further analysis (*low fit quality, short incubation time, ...*)
- After excluding incubations with dried biomass (references for incubation experiments), **81 datasets** remain
- For two datasets no H/C ratio was available, so the histogram on the right is on **79 biochars**
 - 10 of these biochar samples (13%) show an H/C ratio < 0.40
 - 69 samples (87%) of the samples were above this threshold
 - 50 samples >0.4 and <0.7
 - 19 samples even >0.7
 (clearly **not biochar**)

H/C distribution of 27 commercial/industrial biochars (Sanei et al. 2024) shows lower H/C values and a narrower distribution



- From the 64 biochar samples analyzed by Sanei et al. (2024) those biochars that are produced at commercial & industrial scale have been selected (excluding sewage sludge chars as well as R&D, pilot plant and lab samples)
- This leaves us with 27 biochars
- 25 of these biochar samples (93%) show an H/C ratio < 0.40 and only 2 samples were slightly above this threshold often used as a proxy for permanence



Most of the incubation experiments published so far, have been carried out with **carbonized material**, not representative of biochar produced commercially worldwide today



Moreover, the incubation experiments published so far were made with **biochar not** further characterized to obtain the proportions of different biochar fractions

The data from past publications (Azzi et al. 2024) also reveal that the biochar was pyrolyzed at moderate temperatures



QEBI

- At pyrolysis temperatures below 500 °C it is getting increasingly difficult to reach a high degree of carbonization, particularly when residence times are short or variable
- Pyrolysis temperatures were partly estimated and, if measured, based on the temperatures in the reactor. They do not reliably represent the particle temperatures (which are systematically lower)

HTT stands for Highest Treatment Temperature as reported by the producer of the Biochar (BC)

[•] Temperature relative to H/C values of the 79 biochars selected from Azzi et al. (2024) collection of 134 datasets

The data from Sanei et al. (2024) reveal that commercial/ industrial biochars are mostly processed at 500°C or above



E B

 Temperature relative to H/C values of the 27 commercial & industrial biochars out of the 64 samples from Sanei et al. (2024)

 Biochar that has been exposed to high temperatures (>500°C) for long enough time show a high degree of carbonization and aromatization

HTT stands for Highest Treatment Temperature as reported by the producer of the Biochar (BC)

Direct comparison of the data shows how different the two data bases of the analyzed/incubated biochars are



Pyrolysis Temperature relative to H/C

- Average H/C for 27 datasets from Sanei et al. (2024) is H/C = 0.20 (representative of industrially produced biochars)
- Average H/C for 79 datasets from Azzi et al. (2024) is H/C = 0.60 (part of the samples representative of industrially produced biochar)

HTT stands for Highest Treatment Temperature as reported by the producer of the Biochar (BC)



I would have difficulties to create these high H/C biochars on purpose with our systems

Helmut Gerber (founder and CTO of PYREG GmbH) when confronted with datasets of biochar used in incubation experiments

Narrowing the selection down to look at comparable data still shows a significant difference in biochar properties



Pyrolysis Temperature relative to H/C

- With the aim of looking at results of incubation tests selected by Azzi et al.
 (2024) that are closest to today's industrial and commercial biochars, further exclusions have been made
 - all sets of data with HTT < 400°C and H/C > 0.44 were excluded
 - experiments with field tests were excluded (4 sets of data) to have better comparable results
- This leaves 16 experiments from 5 papers (Zimmermann 2010, Singh 2012, Wu 2016, Santos 2021, Aubertin 2021)
 - average H/C is **0.38**
 - average HTT is 540°C

Incubation experiments with biochar representative of biochar produced commercially show marginal degradation rates



QEBI

- For the selection H/C < 0.44 (implicitly HTT > 400°C; 16 datasets) the average remaining carbon after 1 year is 99.4% (H/C average is 0.38)
- As highlighted by Azzi et al.. (2024), the biochars used in past incubation experiments lack advanced characterization. Based on data from Sanei et al. (2024) on biochars with comparable properties, it has been estimated that the biochars used in the experiments contain typically 2.0% (0.5 3.5%) of labile fractions (Appendix).
- The observed degradation is consistent with the fact that labile fractions are prone to microbial degradation

With higher H/C values, degradation rates increase, which is consistent with the assumed larger proportion of labile fractions



- For the selection 0.44 < H/C < 0.70 (and HTT > 400°C) the average amount of remaining carbon after
 1 year is 99.0% (H/C average is 0.56)
- The higher degradation of these biochars is consistent with the fact that such biochars with a higher H/C ratio generally have a significantly higher proportion of labile fractions

If biochars contain highly durable fractions, degradation of such fractions can not be evidenced with incubation experiments



- How realistic is it, that the long-term degradation of the durable biochar fractions can be quantified in incubation experiments over 1, 2 or 3 years?
 - Assuming a 10,000 years half-life for highly durable carbon fractions (above graphs) shows that it would require **measurement accuracy** in the range of **0.001%**
 - It seems unrealistic to reach such measurement accuracies in the presence of labile hydrocarbon fractions, which degrade within few years
- Experimental proof of the degradation of such durable biochar fractions based on incubation experiments would require measurements to run over very long times (at least centuries)
- Thus, incubation experiments are fundamentally unsuitable to determine degradation rates of durable biochar fractions as these are beyond the level of detection due to their low degradation rate

Summary and conclusions on capabilities and limitations of incubation experiments

- Biochar consists of different fractions with fundamentally different chemical properties and therefore, show completely different resistance to microbial decomposition
- Bulk characterization methods of biochars do not give the full picture as they do not provide the proportions of differently stable fractions
- Most of the incubation experiments published so far have been carried out with carbonized material that is not representative of biochar produced commercially worldwide today and with biochar not further characterized to obtain the proportions of different biochar fractions
- In incubation experiments with biochars representative of today's industrial biochars (16 datasets), the **average carbon degradation after one year is 0.6%** (0.25% 1.2%)
- These degradation rates correspond well with the amount of labile fractions, which usually make up 2.0 % (0.5 - 3.5 %) in such biochars and are known to be easily degradable
- These incubation experiments therefore do not allow to make statements on degradation of highly durable fractions. And even further, incubation experiments are fundamentally not suitable to determine degradation of the such durable fractions.



2

Advanced characterization of carbon fractions in biochar

Messages Sanei et al. (2024): (i) R_o of 2% proposed as "Inertinite Benchmark" (IBR_o2%) (ii) fully carbonized biochar = inertinite



Most of the samples of today's commercial biochars showcase R_o distributions entirely above IBR_o2%, denoting pure Inertinite Biochar

- Sanei et al. (2024) includes detailed analyses on 64 biochars:
 - samples cover a broad range of feedstocks, heating treatments and industrial production equipment
 - from the 64 samples, 27 samples are representative of volume produced commercial and industrial biochar
- The authors propose random reflectance (R_o) of 2% as the "Inertinite Benchmark" (IBR_o2%)
- The authors apply R_o measurement to quantify the permanent fraction of carbon in a biochar. The analysis shows that most of the analyzed samples have their entire R_o distribution range well above IBR_o2%

BEBI

Snapshot Random Reflectance (R_o): a calibrated method for determining the proportions of different fractions in biochar



- Fully carbonized biochars consist of large portions of biochar fractions chemically/structurally equivalent to a material know in geology as **inertinite**
- Inertinite Biochar (IBR_o2%) is regarded as the most thermodynamically stable form of organic carbon, analogous to the stability of carbonates (mineral carbon)
- The amount of Inertinite Biochar in a sample can be determined using
 Random Reflectance (R_o), a well-established characterization method used to determine the maturity of coal
- Random Reflectance is the only analytical method, which is calibrated by millions of samples throughout the earth's crust and has a temporal record supporting inertinite's residence time

Biochar consists of different fractions with fundamentally different chemical properties that can be quantified



- Different biochar fractions show completely different behaviors in terms of mineralization by microbial activity
- The different fractions can not be identified with bulk characterization methods
- Characterization methods well established in petrology (including R_o) allow for quantification of the different fractions:
 - Inertinite Biochar (R_o > IBR_o2%): most stable form of organic carbon (geological timeframes)
 - Semi-inertinite (1.2 < R_o < 2%): very high resistance to degradation over significant time scales (centuries to millennia)
 - Liptinite (0.2 < R_o < 1.2%): degradable (years to decades)
 - **Free Hydrocarbons**: highly degradable (weeks to months)



Biochar Carbonization Concrete Ball, Peanut Butter, and Jam Analogy



Increase in Random Reflectance (R_o)



Pyrogenic fractions in fossil coal and pyrolyzed biomass show great similarities in their structural composition



Particle of pyrogenic carbon (fusinite), 300 million years old, stored under highly oxic environment in Australian permian coals



Freshly pyrolyzed pieces of algae

Two natural ways to permanently removal carbon: the <u>inorganic</u> pathway and the <u>organic</u> pathway





Inertinite is a highly carbonized organic matter found abundantly throughout most sedimentary rocks, that is regarded as the end product of organic carbon carbonization processes (organic maturation or wildfire rapid carbonization) in the earth system.

Inertinite Biochar is a highly carbonized material that has undergone extensive thermal alteration, resulting in a substance that is chemically and structurally equivalent to inertinite as assessed by light microscopy, structural analysis, and geochemistry. Inertinite Biochar is defined by a R_o > 2%, the "inertinite benchmark" (IBR_o2%).

Today's industrial & commercial biochars are in large parts chemically/structurally identical to inertinite



- The graph shows data from Sanei et al. (2024):
 - Orange data points refer to the above-mentioned selection of 27 fully industrial & commercial biochars
 - **Blue data points** are R&D samples produced on industrial pyrolysis equipment to deliberately test the limits
- Each R_o-value consists of 500 spatially resolved measurement points over the probe

Today's industrial & commercial biochars are in large parts chemically/structurally identical to inertinite



- The graph shows the data-points including the 90% confidence interval
 - Statistically half of the measurements outside the confidence interval show higher values for random reflectance
 - So only 5% of the measurements outside the confidence interval are below the lower bar
- For almost all commercial biochar samples, not only the mean value but almost the entire R_o distributions are above the inertinite benchmark of R_o = 2.0



EB



Sample #25 consists almost entirely of Inertinite Biochar

* In manual scans the percentages of different carbon fractions do not precisely represent the mass balance see <u>Appendix</u>



EB



Sample **#26** consist mostly of **Inertinite Biochar** with smaller fractions of **semi-inertinite** and **liptinite**





Sample #36 consists almost entirely of Inertinite Biochar



EB



Sample **#37** consists mostly of **semi-inertinite** with a small fraction of **inertinite** and very small fraction of **liptinite**

Exemplary R_o distributions for R&D samples





R&D sample **#4*** consists mostly of **inertinite** and **semiinertinite** with a smaller fraction liptinite

Exemplary R_o distributions for R&D samples



EB



R&D sample **#1*** consists mostly of **semi-inertinite** and **liptinite** with a smaller fraction **inertinite**. H/C as sole proxy for permanence might overestimate durability compared to the previous sample #4*.



3

Summary



Most **biochars** are **heterogeneous** and consist of **various fractions** of different stability



Biochar fractions that have been exposed to high temperatures for sufficient time are chemically/structurally equivalent to inertinite



Inertinite is the most stable form of organic carbon and has long been accepted as a benchmark for permanence in earth science



There is **no of scientific evidence of microbial degradation** of **Inertinite Biochar**. Yet there is still the hypothesis of (relevant) microbial degradation

of inertinite.



If there is **no evidence** for degradation of Inertinite Biochar, but overwhelming evidence of inertinite's persistence over geological time scales, it seems logical to accept Inertinite Biochar as a benchmark for permanence



Inertinite Biochar fractions will not relevantly degrade in soils within climate-relevant periods

(in any case beyond 1,000 years, realistically well beyond 10,000 years)



Whether Inertinite Biochar is microbially degraded over several 10,000 years or if it remains over millions of years is a scientifically interesting question, but is not relevant for climate protection



The analysis in Sanei et al. (2024) of **commercially produced biochars** shows that they **mainly consist of Inertinite Biochar**



Over 95% of the carbon of such **commercial biochars** will not relevantly degrade in soils within climate-relevant periods

(in any case beyond 1,000 years, realistically well beyond 10,000 years)



Yet it can **not be deduced** from the above that <u>all commercial/industrial biochars</u> are generally permanent over climate-relevant periods



The proportion of Inertinite Biochar has to be verified by suitable characterization methods (e.g. random reflectance) and ensured by certification

The Inertinite Biochar fraction in any given type of biochar qualifies for permanent carbon removal



Appendix





R

H/C ratio and R_o as proxies for permanence

EBI

Deep-dive on H/C ratio as a proxy for inertness

H/C measurements as proxy for inertness

- For homogenous biochar, H/C is a valid and, above all, established method to characterize biochar and it is a good proxy for the level of carbonization. However, the hydrogen index H-index might be an even more suitable bulk proxy for inertness.
- H/C mostly correlate with %R_o results. Though bulk values can lead to misleading results when considering very heterogenous biochars.
- Biochars are composed of fractions of differently inert material/molecules. For biochar that might have inhomogeneous material properties a more differentiated approach is recommendable. Sanei et al. (2024) proposed Random Reflectance (%R_o) as a proxy.



- Hydrogen and Carbon are two key elements found in all organic matter
- H/C ratio is controlled by (i) the type of biomass and (ii) the degree of carbonization. Terrestrial biomass (wood) starts with a lower H/C than aquatic biomass (algae). By pyrolysis, hydrogen (H) is driven out of the feedstock as carbonization advances (C remains).

Estimation of the labile fraction for the 16 selected datasets from Azzi et al. (2024)

reactive organic carbon (%)



Sanei et al. (2024) - filtered <750°C & 0.20 < H/C < 0.44)





- For 16 datasets from Azzi et al. (2024) (Ø H/C = 0.38 Ø temp = 540°C) we have no information about the share of the labile fractions in the biochar
- To **estimate** this proportion of **labile fractions**, the reactive carbon fraction from Sanei et al. (2024) was analyzed for the data with the following filter
 - H/C between 0.20 and 0.44
 - and < 750°C
- The selection provides a dataset with an average temperature of 580°C and average H/C of 0.32 (still higher grade of carbonization than the 16 datasets from Azzi et al. (2024)
- To make an estimate on the labile fractions the selection was further narrowed down to the range 0.32 – 0.44

Estimation of the labile fraction for the 16 selected datasets from Azzi et al. (2024)

reactive organic carbon (%)



1,00 0,50 0.00

0,15

0,20

0,25

Sanei et al. (2024) - filtered <750°C & 0.32 < H/C < 0.44*



0,30

H/C

0,35

0,40

0,45

0,50

- The rage 0.32 0,44 provides 11 datasets with an average temperature of 535 °C and an average H/C 0.40 (very close the 16 datasets from Azzi et al. 2024)
- The **resulting reactive organic carbon** fraction shows an **average of 2.0%** ranging from 0.5 – 3.5%
- Based on this result, labile fractions for a set of 16 datasets from Azzi et al. (2024) was estimated to 2.0% (0.5 – 3.5%)

^{*} Note, even biochars pyrolyzed at high temperatures can include higher portions light hydrocarbons trapped in the porous carbon matrix

Example for bulk values not providing the full picture

Looking at two different biochar samples

- Sample A (industrially produced) shows H/C = 0.39,
 Sample B (R&D test run on pilot-line) H/C = 0.13.
 Judgment on permanence based on H/C ratio would suggest that Sample B is more durable.
- The graphs to the right show R_o measurements for the two samples. While mean values for R_o differ only slightly, the distribution is totally different. For Sample A 95% of the fractions surpass the IBR_o2% threshold and for Sample B this is only 49%. Judgment on permanence based on R_o would suggest that Sample A consisting of 95% Inertinite Biochar.
- Even a mix of hard wood and well carbonized biochar can yield results below the commonly accepted threshold for biochar-based carbon removals of H/C<0.4



Quantification of the four different fractions of biochar





Manual and automatic R_o scan of a biochar sample to determine the proportion of inertinite and semi-inertinite in the fixed carbon

- The share of free hydrocarbons (FHCs) is quantified by re-pyrolysis at 300°C
- The share of liptinite-derived hydrocarbons (LHCs) is measured during ramp-up of temperature from 300°C to 650°C
- The amount of residual carbon or fixed carbon (composed of semiinertinite and Inertinite) is determined during ramp up of temperature from 150°C to 850°C in oxygen environment
- Random reflectance measurements R_o are made to quantify the split between inertinite and semi-inertinite
 - according to ISO 7404-5, 2009 R_o is measured with manual scans,
 - additionally automatic scans are used to verify the results



ii

Further Questions

Questions regarding potential use of Inertinite Benchmark (IBR_o2%) in carbon removal standards and certification schemes

How representative are the measurements on R_o?

- What is the variation of the output of a pyrolysis machine?
- How representative are the samples taken?
- Building on afore mentioned question: What would be a reasonable protocols for sampling and (re)testing requirements?
- Are there enough labs conveniently available to all biochar producers that are able to do random reflectance testing in sufficient number & at affordable price points?
- How will key standards like EBC, Puro and Verra deal with permanence in the future?

Questions beyond the Inertinite Biochar fraction

How about degradation rates of semi-inertinite?

- To which extent is it adequate to use this fraction for 1,000 yr CO₂-certificates?
- How about degradation rates of Liptinite Hydrocarbons (LHCs)?
 - In CO₂-certifications for permanent CDR this should not be used though
 - Scientifically and to explain historic incubation experiments this is still interesting



iii

Literature

Literature references for two papers analyzed for this report

- Sanei, H., Rudra, A., Przyswitt, Z.M.M., Kousted, S., Sindlev, M.B., Zheng, X., Nielsen, S.B., Petersen, H.I., 2024. Assessing biochar's permanence: An inertinite benchmark. International Journal of Coal Geology <u>https://doi.org/10.1016/j.coal.2023.104409</u>
- Azzi, E.S., Li, H., Cederlund, H., Karltun, E., Sundberg, C., 2024. Modelling biochar long-term carbon storage in soil with harmonized analysis of decomposition data. Geoderma. <u>https://doi.org/10.1016/j.geoderma.2023.116761</u>