

CRANFIELD UNIVERSITY

DAVID LEFEBVRE

LIFE CYCLE ASSESSMENT OF LAND-BASED GREENHOUSE GAS
REMOVAL TECHNOLOGIES

SCHOOL OF WATER, ENERGY, ENVIRONMENT AND AGRIFOOD

PhD

Academic Year: 2018 - 2021

Supervisor: Dr Adrian Williams

Associate Supervisor: Prof Guy Kirk

June 2021

CRANFIELD UNIVERSITY

SCHOOL OF WATER, ENERGY, ENVIRONMENT AND AGRIFOOD

PhD

Academic Year 2018 - 2021

DAVID LEFEBVRE

Life cycle assessment of land-based greenhouse gas removal technologies

Supervisor: Dr Adrian Williams
Associate Supervisor: Prof Guy Kirk
June 2021

This thesis is submitted in partial fulfilment of the requirements for the
degree of PhD

© Cranfield University 2021. All rights reserved. No part of this
publication may be reproduced without the written permission of the
copyright owner.

ABSTRACT

Greenhouse gas removal technologies (GGRT) are needed to meet the UNFCCC aim to limit the global average temperature increase to 2°C above pre-industrial levels. GGRTs vary in carbon sequestration potential, readiness level, scalability, cost, required surrounding environment and related environmental and social effects. Quantifying these components in every context is critical to ensure maximum greenhouse gas removal efficiency while minimising negative effects.

In this thesis I use life cycle assessment (LCA) to assess the carbon sequestration potential of three GGRTs: enhanced weathering, biochar and reforestation. I use case studies in São Paulo, Brazil for biochar and enhanced weathering, and in the Peruvian Amazon for reforestation. In addition, I use LCA to identify the most important processes in each system and to determine context-specific limits that switch the systems from net GHG sequestration to net GHG emission.

Overall, this work promotes the use of LCA to consider GGRTs in their entirety and predict their context-specific carbon capture potential, along with their limitations and potential caveats to guide both the science and policy communities.

This thesis was made possible thanks to funding through the SOILS-R-GGREAT (NE/P019498/1) project of the greenhouse gas removal (GGR) program. The GGR program is financed by the UK Natural Environment Research Council (NERC), Engineering and Physical Science Research Council (EPSRC), Economic and Social Science Research Council (ESRC) and the UK department for Business, Energy and Industrial Strategy (BEIS).

Keywords: LCA; GGRT; CDR; NET; Enhanced Weathering; Biochar; Reforestation

ACKNOWLEDGEMENTS

I am grateful to my supervisors: Dr Adrian Williams, Prof Guy Kirk, Dr Jeroen Meersmans and Dr Pietro Goglio for their support, sound advice, and mentorship throughout, or during part, of this journey. I extend this gratitude to the broader Cranfield personnel and members of the Soil-R-GGREAT project for their availability, support, and constructive criticism.

I am thankful to my family and friends for their support on cloudy days as well as their presence when the sun shone.

Finally, I express my upmost gratitude to Eloise for her unparalleled support, communicable confidence, trust, and selflessness.

TABLE OF CONTENTS

ABSTRACT	i
ACKNOWLEDGEMENTS	iii
LIST OF FIGURES.....	vii
LIST OF TABLES.....	xi
LIST OF EQUATIONS	xiii
LIST OF ABBREVIATIONS	xiv
CHAPTER 1.....	1
INTRODUCTION.....	3
1.1 <i>Paris Agreement</i>	3
1.2 <i>Carbon Dioxide Removal Practices</i>	3
1.3 <i>GGRT Needs</i>	5
1.4 <i>Limitations and Concerns of GGRTs</i>	5
1.5 <i>Land-Based GGRTs</i>	6
1.6 <i>Life Cycle Assessment</i>	10
1.7 <i>Aims and Objectives</i>	11
1.8 <i>Thesis Structure</i>	12
1.9 <i>References</i>	14
CHAPTER 2.....	23
ASSESSING THE POTENTIAL OF SOIL CARBONATION AND ENHANCED WEATHERING THROUGH LIFE CYCLE ASSESSMENT: A CASE STUDY FOR SÃO PAULO STATE, BRAZIL	25
2.1 <i>Abstract</i>	25
2.2 <i>Introduction</i>	26
2.3 <i>Methodology</i>	27
2.4 <i>Results</i>	39
2.5 <i>Discussion</i>	46
2.6 <i>Conclusion</i>	53
2.7 <i>Acknowledgments</i>	54
2.8 <i>References</i>	54
CHAPTER 3.....	65
MODELLING THE POTENTIAL FOR SOIL CARBON SEQUESTRATION USING BIOCHAR FROM SUGARCANE RESIDUES IN BRAZIL	67
3.1 <i>Abstract</i>	67
3.2 <i>Introduction</i>	67
3.3 <i>Results and Discussion</i>	70
3.4 <i>Method</i>	78
3.5 <i>References</i>	84
3.6 <i>Acknowledgments</i>	92
CHAPTER 4.....	93

AN ANTICIPATORY LIFE CYCLE ASSESSMENT OF THE USE OF BIOCHAR FROM SUGARCANE RESIDUES AS A GREENHOUSE GAS REMOVAL TECHNOLOGY	95
4.1 Abstract.....	95
4.2 Introduction	96
4.3 Method.....	97
4.4 Results.....	105
4.5 Discussion.....	108
4.6 Conclusions	112
4.7 Acknowledgments	112
4.8 References.....	112
CHAPTER 5.....	123
ASSESSING THE CARBON CAPTURE POTENTIAL OF REFORESTATION PROJECTS	125
5.1 Abstract.....	125
5.2 Introduction	126
5.3 Results.....	128
5.4 Discussion.....	132
5.5 Methods	136
5.6 Acknowledgements.....	142
5.7 References.....	142
CHAPTER 6.....	151
DISCUSSION	153
6.1 Key results and testing of hypothesis.....	153
6.2 Intellectual contribution and potential impacts of findings.....	154
6.3 Limitation of the work.....	155
6.4 Limitations to the deployment of GGRTs and future challenges.....	157
6.5 An opportunity for localized actions	160
6.6 Future of GGRTs	161
6.7 References.....	162
CHAPTER 7.....	167
CONCLUSION	169
APPENDICES.....	171
Appendix A Supplementary Information – Chapter 2: Assessing the potential of soil carbonation and enhanced weathering through Life Cycle Assessment: a case study for São Paulo State, Brazil.....	171
Appendix B Supplementary Information – Chapter 3: Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil.....	183
Appendix C Supplementary Information – Chapter 4: An anticipatory life cycle assessment of the use of biochar from sugarcane residues as a greenhouse gas removal technology.....	203
Appendix D Supplementary Information – Chapter 5: Assessing the carbon capture potential of reforestation projects	235

LIST OF FIGURES

Figure 2.1: Overview of the rock weathering process, the two pathways (carbonation and EW) considered in this study and the sequestration potential of Brazilian basalt for each considered path.	31
Figure 2.2: São Paulo state with buffer zones (red shade) around São Paulo's basalt quarries overlapping grassland and cropland areas.....	32
Figure 2.3: System boundaries for extraction, comminution, transport and spreading of basalt considered in this study.	33
Figure 2.4: Crushing process stages considered to reach 5 mm diameter particles of basalt based on Rosado <i>et al.</i> (2017) and Metso (2015).	36
Figure 2.5: Impact categories contribution analysis considering the quarry to field distance of 65km.	41
Figure 2.6: Effect of an increasing quarry to field distance on the process stages considering GHG balance [kg CO ₂ e] impact category and an application rate of 5 t/ha.....	42
Figure 2.7: Share increase of transport linked with increasing quarry to field distance [km] considering total GHG balance impact category and an application rate of 5t/ha..	43
Figure 2.8: Effect of Emission Factor (truck type) and W value on the offsetting distance. The W value of 1 and 1.79 represent Carbonation and EW, respectively. The error bars represent the propagation of the uncertainty on emissions associated with all the other processes as well as the uncertainty on sequestration potential according to the W value.	51
Figure 3.1: Comparison of the soil C stock increase after 5 years of biochar addition (10 applications of biochar) between measured values (black dots ± SD) and simulated (red and green lines) using the modified RothC model described in this manuscript. The red line accounts for the loss of 5.2% year ⁻¹ of the biochar via leaching.	71
Figure 3.2: Predicted SOC stock in sugarcane fields across São Paulo. At steady state (A) and after 5, 10, and 20 year of biochar addition for Scenarios 1 (B, C, and D respectively), 2 (E, F, G), and 3 (H, I, J). For Scenarios 1, 2, and 3, the biochar additions are 2.46, 1.23, and 0.62 t C ha ⁻¹ year ⁻¹ respectively and the fresh C inputs are 6.57, 9.74, and 11.1 t C ha ⁻¹ year ⁻¹ , respectively as trash, bagasse, roots, root exudates, and applications of vinasse and filter cake.....	73
Figure 3.3: Effect of FAO soil type on predicted SOC stock up to 20 year. Dots are means across sugarcane fields at the indicated times (year 0 is the steady state value); ribbon width is 1 SD due to geographical climate variability; lines indicate general trends and not intermediate points. The SD is small for Soil 7 because they account for only a few sugarcane fields concentrated in one particular region (Supplementary Table B.3). The land area associated with each soil type is shown in parenthesis for each plot.	74

Figure 3.4: Priming effect of biochar on SOC under the three addition scenarios. Dots are mean values across sugarcane fields at the indicated times (year 0 is the steady state value); ribbon width is due to climate and soil variability (1 SD). The downward arrow indicates the % decrease in SOC stock after 20 year compared to no priming.	75
Figure 3.5: São Paulo State C sequestration potential. The number in bold above each bar represent the State total C sequestered after 20 years of biochar addition in Mt of C (10^6 t C).	77
Figure 3.6: Soil carbon model. RothC is combined with a sub-model for the decomposition of fresh biochar. Details in text.	80
Figure 3.7: Scenario flowchart representing the fate of the bagasse and trash C produced per hectare per year on a sugarcane field in São Paulo. ‘CHP’ stands for ‘combined heat and power plant’	83
Figure 4.1: Comparison between the baseline system and the assessed scenario. The orange box represents the process or products susceptible to change from the baseline to the biochar producing scenario and, hence, are assessed for their carbon footprint.	98
Figure 4.2: Contribution analysis considering NG as marginal energy and 100% allocation of available sugarcane residues to BC production. The upper outlined numbers report the exact figures in t C ha ⁻¹ for each process. The under outlined numbers report the share of each process within each group (emission or emission reduction). The ribbon and error bars are $\pm$ 1 SD. The dotted line represents the net sequestration value.	106
Figure 4.3: Combined results of all scenarios. Section A, B and C represent the production of 100%, 50% and 25% of the maximum biochar production. The remaining residues in section B and C are treated in a CHP similarly to the baseline. The black dots and labels are the net sequestration with associated error bars representing $\pm$ 1 SD. .	107
Figure 4.4: Biochar soil carbon sequestration efficiency: the remaining carbon capture potential of biochar in soils of after accounting for all the processes, in percent. Section A, B and C represent the production of 100%, 50% and 25% of the maximum biochar production. The remaining residues in section B and C are treated in a CHP similarly to the baseline. The error bars are $\pm$ 1 SD.	108
Figure 5.1: Descriptive schematic figure of the processes and stocks considered. a Vegetation and soil C stocks. b Processes included in the LCA and their relative impact. c effect of the planting density.	128
Figure 5.2: Changes in carbon stocks following transplanting. a Vegetation and soil. b Total, with $\pm$ 1 SD indicated by the ribbon.	129
Figure 5.3: Grouped contribution analysis of the emission and emission reduction associated with reforesting one hectare from our case study plot including one year maintenance. The error bars represent $\pm$ 1 SD.	130

Figure 5.4: Time needed in years (labels) to reach estimated claim of 100 kg CO ₂ e captured per tree using our model considering all C pools (black), above and below-ground C pool only (yellow), and above-ground C pool only (blue). The red label represents the time in years necessary to offset the emissions associated to set up the plot. The ribbon around the model carbon capture rate is ± 1 SD.	131
Figure 5.5: Effect of the planting density on the plot-scale carbon capture rate. No manual thinning and 100% seedling transplant survival are assumed. The blue dot represents the planting density of 1,111 seedling ha ⁻¹ (where our model reach 100 kg CO ₂ captured per tree 4.1 years after planting).....	132
Figure 5.6: Case study location. Generated using Google Earth (Google, 2018)	137
Figure 5.7: System boundaries of the study	138
Figure 5.8: Above-ground carbon model (black line) and boxes representing above-ground carbon values from the literature of forest plot nearby our case study. Forest plots represented by the boxes on the right hand side were assumed to be a mature forest (≈ 80 years) (Becknell, Kissing Kucek and Powers, 2012; Requena Suarez et al., 2019; S. L. Lewis et al., 2019; T. Lewis et al., 2019).....	141
Figure A.1: Average emissions and standard deviation from extraction to application of one tonne of basalt for different type of trucks and considering an average quarry to field distance of 50km in straight line.....	175
Figure A.2: Above: sequestration potential trend in t CO ₂ e /t of basalt for increasing value of ω . Bottom: distribution curve with $\omega = 1$ (left) and $\omega = 1.79$ (right) representing carbonation and enhanced weathering, respectively. The Dotted purple line represents the 25 & 75th percentile and the red line dotted line, the 2.5 & 97.5th percentile.....	177
Figure A.3: Effect of Emission Factor (truck type) and ω value on the offsetting distance. The ω value of 1 and 1.79 represent Carbonation and EW, respectively. The error bars represent the propagation of the uncertainty on emissions associated with all the other processes as well as the uncertainty on sequestration potential according to the ω value.	179
Figure B.1: The study area (State of São Paulo – Brazil) with; (i) the 7 different soil types according to the Harmonized World Soil Database (FAO, 2012), (ii) the location of the meteorological stations considered in the study (FAO, 2006) and (iii) the sugarcane fields according to the CANASAT project (Theodor Rudorff, 2014). Map generated using ArcGIS 10.5.1 (ESRI, 2011).....	185
Figure B.2: Soil carbon stock values at steady state obtained through the R (green) and the Windows (blue) version of RothC. The boxplot combines the values obtained for the different meteorological stations relevant to each soil type. The black boxplot represents the spread of the soil carbon stock under sugarcane cultivation in São Paulo found in the literature (De Oliveira Bordonal et al., 2013; Maia et al., 2014; Carvalho et al., 2017). The single line for Soil 7 is due to the only meteorological	

station attributed to this soil type (see Table B.3). The purple dotted line represents the averaged value when all relevant soils and climate station combinations are considered. The end of the upper (lower) whiskers represent the largest (smallest) value within 1.5 times interquartile range above (below) 75th (25th) percentile, as defined by the ggplot2 package of R software version (3.5.1) (Core Team, 2018).

..... 189

Figure B.3: Comparison between measured (Rogovska et al., 2014) (black dots $\pm$ SD) and simulated (red line) values using our modified RothC model. The soil carbon measured values originates from Rogovska et al. (2014) two years after biochar application..... 193

Figure B.4: Carbon stock (t ha^{-1}) 2 yr after biochar addition. Comparison between measured values (Rogovska et al., 2014) (black dots $\pm$ SD), simulated (red line) values using our modified RothC model, simulated values accounting for potential leaching of biochar particles (green line), and simulated values accounting for potential downward (leaching) and lateral migration (erosion) of biochar particles (yellow line). 195

Figure B.5: Biochar-C remaining as percent of biochar-C added over time. Our simulated values (red line) are based on meta-analysis values (Wang et al., 2016) applied on São Paulo conditions. The black dots are data points collected from literature (Kuzyakov et al., 2009; Fang, Singh and Singh, 2014; Kuzyakov, Bogomolova and Glaser, 2014; Dharmakeerthi et al., 2015; Fang et al., 2019) studying biochar mineralization in various condition through incubation experiments. The model reflects the general trend of these disparate experiments, which show, not-surprisingly, substantial scatter. Without longer term incubation experimental data, we cannot conclude if the model shows systematic bias nor do the experimental data contradict the model's predictions. 196

LIST OF TABLES

Table 1.1: Status of paper-based chapters	13
Table 2.1: Database and process used in the LCI of basalt extraction, comminution, transport and spreading	34
Table 2.2: Energy requirements of crushing process equipment, as metered electricity.	37
Table 2.3: LCIA results for three application rates. Results are shown per hectare and per tonne of CO ₂ e removed according to the two paths (EW & Carbonation).....	40
Table 2.4: Slope and constant of each impact category for linear prediction.	43
Table 2.5: Emissions and net sequestration in kt CO ₂ e for the implementation of the practice considering the area included under each buffer zones (with application rate of 5t/ha).....	45
Table 2.6: São Paulo (SP) CO ₂ e sequestration potential through carbonation and EW as compared with 2016 SP, Brazilian, and global CO ₂ e emissions. Expressed in Mt CO ₂ e.....	45
Table 4.1. Brazilian energy mix in 2016 and predicted for 2021 (Barros, Piekarski and De Francisco, 2018) and emission factors (EF) (Wernet et al., 2016).	100
Table 4.2. Summary table of the electricity mixes and emission factors considered for each scenario.	101
Table B.1: Main characteristics of the soil type selected as given by the harmonized world soil database (FAO, 2012).	183
Table B.2: Latitude, longitude, yearly average temperature and monthly precipitation values of the different meteorological stations considered from ClimWat database (FAO, 2006).....	184
Table B.3: The actual existing soil type/meteorological station combinations present in the state of São Paulo.	184
Table B.4: Parameter settings for soil carbon and biochar production calculations. ...	186
Table B.5: Evaluation dataset.....	190
Table B.6: Additional evaluation dataset	191
Table C.1: System information.....	207
Table C.2: Electricity baseline mill	209
Table C.3: Electricity pyrolysis.....	210
Table C.4: Electricity biochar processing.....	211

Table C.5: Additional processes.....	212
Table C.6: Yearly addition of N containing amendment with their nitrogen content and the total N applied per hectare per year	212
Table C.7: Emission factors.....	213
Table C.8: Average amendment application per hectare per year taking into account 4/5 years of ratooning (Cardoso <i>et al.</i> , 2019).....	214
Table C.9: Pyrolysis plant construction material	214
Table D.1: Latitude, longitude, yearly average temperature and monthly precipitation values from the ClimWat database (FAO, 2006) of the meteorological station considered in this study. These meteorological values were assumed in repetition for the modelling activities.	235
Table D.2: Soil characteristics of the case study plot and the reference forest (Román-Dañobeytia et al., 2021).	236
Table D.3: Elements needed for the growth of 1000 seedlings in the nursery (Farfan, 2020).	236
Table D.4: Data for soil and biomass carbon modelling activities.....	237
Table D.5: Data and emission factors used for the LCA.....	238
Table D.6: Contribution analysis of the LCA related to the setup of CINCIA's reforestation plot.	241

LIST OF EQUATIONS

(2-1) 28

(2-2) 28

(2-3) 28

(2-4) 28

(2-5) 29

(2-6) 39

(3-1) 80

(A-1) 178

(B-1) 187

LIST OF ABBREVIATIONS

AR5	Assessment Report #5
BC	Biochar
BECCS	Bioenergy with Carbon Capture and Storage
C	Carbon
CDR	Carbon Dioxide Removal
CHP	Combine Heat and Power
CO ₂	Carbon dioxide
CO ₂ e	Carbon dioxide equivalent
COP	Conference of the Parties
DAC	Direct Air Capture
DM	Dry Matter
EF	Emission Factor
ESM	Earth System Model
EW	Enhanced Weathering
GGR	Greenhouse Gas Removal
GGRT	Greenhouse Gas Removal Technology
GHG	Greenhouse gas
Gt	Gigaton
GWP	Global Warming Potential
ha	Hectare(s)
IAM	Integrated Assessment Model
IPCC	Intergovernmental Panel on Climate Change
kWh	kilo Watt hour
LCA	Life Cycle Assessment
Mt	Megaton
NCS	Natural Climate Solutions
NET	Negative Emission Technology
NG	Natural Gas
SD	Standard Deviation
SDGs	(United Nation) Sustainable Development Goals
SOC	Soil Organic Carbon
SP	São Paulo

tBC	ton of Biochar
TWh	Tera Watt hour
UNFCCC	United Nations Framework Convention on Climate Change
yr	Year(s)

CHAPTER 1

Introduction

For every complex problem there is an answer that is clear, simple, and wrong.

H. L. MENCKEN

INTRODUCTION

1.1 Paris Agreement

At the 21st Conference of the Parties (COP) of the United Nations Framework Convention on Climate Change (UNFCCC) in Paris in 2015, it was agreed to limit the global temperature increase to well below 2°C above pre-industrial levels and to strive to stay under a 1.5°C increase (UNFCCC, 2015). Article 4 of the agreement stipulates that the objective is to be reached by reducing emissions “*so as to achieve a balance between anthropogenic emissions by sources and removals by sinks of greenhouse gases*” (UNFCCC, 2015, p.4).

Soon after the agreement, several studies concluded that it would be necessary to remove carbon dioxide (CO₂) from the atmosphere for virtually all pathways to limit the global temperature increase to 2°C (Anderson and Peters, 2016; Smith *et al.*, 2016). The Intergovernmental Panel on Climate Change (IPCC) Special Report on Global Warming of 1.5°C, published in 2018, followed by stating that all pathways “*that limits global warming to 1.5°C with no or limited overshoot*” will require the removal of between 100 and 1000 Gt of CO₂ in the 21st century, depending on the pathway (IPCC, 2018, p.16). This observation was maintained in their following special report on climate change and land and corroborated by additional studies since then (Hansen *et al.*, 2017; Kriegler *et al.*, 2018; Rogelj *et al.*, 2018; IPCC, 2019; Brack and King, 2020). Even when worldwide extreme mitigation measures (including lifestyle change, additional reduction of non-CO₂ GHG, diet change, high agricultural yield, electrification) are modelled, the need for CO₂ removal is significantly reduced but not eliminated (Van Vuuren *et al.*, 2018). Hence emission reductions alone will not be sufficient to reach the objectives of the COP 21.

This realisation is strengthened by the acknowledgement that many mitigation measures taken by participating countries are inevitably subject to implementation delays. This means it is even more unlikely the Paris agreement can be reached with mitigation strategies alone (Brown *et al.*, 2019).

1.2 Carbon Dioxide Removal Practices

Carbon dioxide removal (CDR) is defined by the IPCC as “*Anthropogenic activities removing CO₂ from the atmosphere and durably storing it in geological, terrestrial, or*

CHAPTER 1

ocean reservoirs, or in products. It includes existing and potential anthropogenic enhancement of biological or geochemical sinks and direct air capture and storage, but excludes natural CO₂ uptake not directly caused by human activities.” (IPCC, 2018, p.26).

Along with CDR, NET (negative emission technology) or GGRT (greenhouse gas removal technology) are also used throughout the literature. As stated in the definition above, CDR includes all anthropogenic actions that permanently remove CO₂ from the atmosphere (IPCC, 2018). A NET is similar, with the addition that it describes a technology able to reach a net removal of greenhouse gases (GHGs), meaning that its capacity to remove GHGs is greater than the emissions associated with its deployment (Brander *et al.*, 2021). This adds a notion of GHG balance to CDR. A GGRT is yet broader than a NET since it covers any technology able to remove any of the most important GHGs but also includes technologies able to transform a GHG with a high global warming potential to a GHG with a lower global warming potential (e.g. methane to carbon dioxide) (Goglio *et al.*, 2020). In this thesis, the term GGRT is used as it is the broadest term of the three. However, CDR and NET are sometimes used when reporting results from published studies.

In the scientific literature, the first discussions about GGR are associated with the first integrated assessment models, associating bio- and geo-physical with socio-economic models to simulate mitigation pathways (Carton *et al.*, 2020). Soon after, debates and political disagreements about the types of sequestration practices and how to measure their efficacy demonstrated the need for further understanding of the practices and accounting methods (Carton *et al.*, 2020). Since then, the literature on the subject has been growing at an exponential rate. A review of the literature has shown that the number of publications on NET totalled 2,900 between 1991 and 2016 from which, almost 500 were published in 2016 alone (Minx *et al.*, 2017). Greenhouse gas removal (GGR) practices now encompass a variety of techniques, from well-known and practiced, to still at concept stages with important uncertainties and doubts about their scalability (Cox, Spence and Pidgeon, 2020; Ng *et al.*, 2020). Nevertheless, the use of GGRTs as a strategy for reducing the risks associated with climate change is currently partly accepted by the public (Cox *et al.*, 2020) and research is more active than ever.

1.3 GGRT Needs

Gasser *et al.* (2015) calculated that, assuming conventional mitigation strategies, between 0.5 and 3 Gt C would need to be captured per year to reach the 2°C target. This number rose to between 7 and 11 Gt C yr⁻¹ when they allowed for delayed and less-effective mitigation strategies (Gasser *et al.*, 2015).

Compatible with 1.5°C pathways and assuming no overshoot, the IPCC 1.5°C Special Report estimates that, for example, bioenergy with carbon capture and storage (BECCS) and afforestation/reforestation practices would together need to capture between 0 to 6, 1 to 19 and 1 to 21 Gt CO₂ yr⁻¹ in 2030, 2050 and 2100, respectively (IPCC, 2018). These numbers are constantly evolving and depend on the carbon budget left (estimated at 1200 Gt CO₂ from 2015 onward for the 2°C pathway – Fuss *et al.*, 2014), the evolution of the CO₂ emissions rate, mitigations endeavours, and GGRT implementation year (Gasser *et al.*, 2015; Kriegler *et al.*, 2018; Mace *et al.*, 2021).

However, most agree that in the business-as-usual scenario, these numbers are substantial and that the risks, costs, environmental and social pressures, and limitations associated with the deployment of GGRTs to such scale, jeopardize their capacity to reach the necessary level (Gasser *et al.*, 2015; Hansen *et al.*, 2017; Fuss *et al.*, 2018; Mace *et al.*, 2021).

1.4 Limitations and Concerns of GGRTs

Originally, the main concern related to GGRT was that they would justify the continuation of a business-as-usual scenario in terms of emissions, legitimate the push back of new mitigation measures, or be utilized to cover ineffective mitigation actions (Carton *et al.*, 2020).

However, due to the massive amount of CO₂ capture needed even for optimistic mitigation strategies (overviewed in Section 1.3), GGRT advocates emphasise the importance of mitigation strategies and their major impact on the potential success of the UNFCCC 2°C agreement, even when combined with GGRT most optimistic scenarios (Gasser *et al.*, 2015; Mace *et al.*, 2021). Similarly, the idea of a specific GGRT as a silver bullet capable of offsetting most human induced emissions is long gone and the literature promotes the use of the entire GGRT portfolio along with drastic mitigation measures

CHAPTER 1

(Mace *et al.*, 2021) or as Anderson *et al.* (2019, p 933) wrote it “...*incorporating NCS [natural climate solutions, a sub-group of GGRTs] with energy and industrial mitigation in the climate portfolio should not be “either/or” but “yes, and.”*”

McLaren *et al.* (2019) go further and state that it is critical to separate emission reduction and GHG capture in net zero model pathways because carbon capture can be subject to potential future leakage while emission reduction have lasting impact on CO₂ atmospheric concentration. They argue that separating the two allows a critical evaluation of the relevance of each (McLaren *et al.*, 2019).

More-recent concerns about GGRTs are the inherent limitations associated with each. Some rely on extensive changes in land use and threaten biodiversity and food security (Minx *et al.*, 2017; Forster *et al.*, 2020). Others are currently prohibitively expensive, are reversible, or of limited capacity, or all of these (Hansen *et al.*, 2017; Carton *et al.*, 2020). In addition, there is the accountability challenge of GGRT and the need to ensure adequate measurement of impact. Proper accountability is critical for the acceptance of GGRT by policymakers, investors and the general public (Brander *et al.*, 2021). Exhaustive impact assessments of each technology are needed, but are highly location- and context-specific, and it is sometimes difficult to quantify sink capacities and potential leakage (Brandão *et al.*, 2013; Brack and King, 2020; Ruseva *et al.*, 2020; Brander *et al.*, 2021).

Hence, not only is it agreed that implementation of GGRTs cannot replace emission reductions, but also that delays in reducing emissions increase the reliance on GGRTs with known limits, leakage potentials and questions about accounting methods.

1.5 Land-Based GGRTs

Land-based GGRTs include afforestation and reforestation, wetland restoration, soil carbon sequestration, biochar, BECCS and enhanced weathering. Each of these has advantages, limitations and risks (Fuss *et al.*, 2018). BECCS, for instance has an important capture potential but is costly and has a significant land use impact which can limit its development (Smith, 2016). A recent study found that all land-based GGRTs contributed positively to at least one of the United Nation Sustainable Development Goals (SDGs). Some, however, also contributed negatively to these SDGs, mainly through land competition for which the authors suggested further research and demonstration projects

(Smith *et al.*, 2019). This work focuses on three land-based GGRTs, enhanced weathering, biochar, and reforestation. An overview of each practice and their estimated global GGR potential follows.

1.5.1 Enhanced weathering and carbonation

Enhanced weathering relies on one of the mechanisms responsible of regulating Earth's climate over geological time scales (Berner and Kothavala, 2001). This mechanism is the weathering of silicate minerals by carbonic acid (H_2CO_3) resulting from the dissolution of CO_2 in rainwater. In enhanced weathering the speed of the reaction is increased by crushing the rock and thereby increasing the reactive surface area, and applying it on land and thereby exploiting CO_2 capture and release by vegetation (Taylor *et al.*, 2017). So-called 'carbonation' differs from 'enhanced weathering' in the fate of the weathering products and the amount of CO_2 sequestered. The greatest CO_2 sequestration is achieved if the carbonate and bicarbonate ions produced in silicate weathering are transferred to the oceans where they have very long residence times. Whereas under carbonation, the ions are re-precipitated as carbonate minerals in the soil, resulting in half the CO_2 capture per unit rate of silicate weathering (Manning *et al.*, 2013; Washbourne *et al.*, 2015; Kolosz, Sohi and Manning, 2019).

Enhanced weathering and carbonation have high GGR potential. It is estimated that up to 1 Gt of carbon dioxide equivalent (CO_2e) yr^{-1} could be sequestered by carbonation worldwide (Fuss *et al.*, 2018; Minx *et al.*, 2018). Taylor *et al.* (2016) go further by stating that up to 2 to 5 Gt C yr^{-1} (depending on application rate) could be captured via enhanced weathering if two thirds of the global available agricultural land was amended with basalt dust (a silicate rock). Overall, its potential is dependent on the available silicate rock, the area available for its application without adversely affecting biodiversity or food production, the factors affecting rates of silicate rock dissolution, and economics (Beerling *et al.*, 2020).

In addition to CO_2 capture, enhanced weathering may come with side benefits such as improved crop yield by providing K, P or other mineral nutrients, increased plant resistances to pests and diseases, and improved soil alkalinity as long as the increases in soil pH are not excessive (Beerling *et al.*, 2018). However, uncertainties about the dissolution rate, the availability of rock and the actual impacts on vegetation and the

CHAPTER 1

environment (Renforth, 2012; Moosdorf *et al.*, 2014; Beerling *et al.*, 2018) warrant further research. In addition, enhanced weathering is not yet widely accepted by the general public. There are concerns about effects on the oceans and the potential opening of mines for fighting climate change (Cox *et al.*, 2020)

1.5.2 Biochar

Biochar is a carbon rich product of the thermochemical conversion of biomass in a low oxygen atmosphere – pyrolysis (Smith *et al.*, 2019). Pyrolysis modifies the biomass both physically and chemically and forms (along with syngas and synoil) a carbon rich material highly resistant to biodegradation, remaining stable in soil for an average of 556 years (Wang *et al.*, 2016).

The use of biochar as a GGRT is quite recent proposition but interest in its potential is growing (Kamali *et al.*, 2020). The IPCC first acknowledged biochar as a GGRT in Assessment Report 5 (AR5) but recognized that there was still a lack of knowledge surrounding the practice (IPCC, 2014). In their special 1.5°C report however, four years after the AR5, the evidence of biochar as a GGRT was considered “*robust*” (de Coninck and Revi, 2018, p.343)

Estimates of its global potential differ largely, depending on assumptions. Lehmann *et al.* (2006) estimated that around 2 Gt CO₂e yr⁻¹ could be captured if biochar was produced using crop and forestry residues, with the biomass normally combusted for slash and burn agriculture, and using pyrolysis to produce the current bioenergy production. Laird *et al.* (2009) reached between 1.5 and 3.3 Gt CO₂e yr⁻¹ considering crop and aboveground forestry residues, forests degraded by disease and the transfer of 50% of human induced biomass burning to pyrolysis. Various global potentials are claimed in the literature from 0.65 to 35 Gt CO₂e yr⁻¹, depending on sources and quantities of available biomass (Tisserant and Cherubini, 2019).

Along with its GGRT potential, studies have shown that biochar interacts with the native soil organic carbon (Wang *et al.*, 2016; Han Weng *et al.*, 2017; Ding *et al.*, 2018), impacts soil microorganisms’ diversity and density (Domene, 2016), and modifies soil physical properties (Bamminger *et al.*, 2016; Zhang *et al.*, 2021) and chemical properties (Hagemann *et al.*, 2017; Liu *et al.*, 2018, 2019; Gao *et al.*, 2019; Zhang *et al.*, 2021).

Biochar is generally seen as beneficial for soil quality, fertility, yield and risk of soil degradation (IPCC, 2020). However, its impacts on the environment are complex and work through intricate mechanisms. Considering its application should be case specific and necessitate prior assessment (Tisserant and Cherubini, 2019).

1.5.3 Reforestation

Carbon capture through afforestation and reforestation was already considered in the Kyoto protocol (UNFCCC, 1997). Since then, in part due to belief in the cost-effectiveness of the practice, reforestation became, along with BECCS, one of most referenced GGRT in the in IPCC Assessments (Carton *et al.*, 2020). Compared to other GGRT, reforestation is affordable and requires unsophisticated and established technology (Griscom *et al.*, 2017). As a consequence, the number of tree planting projects has increased massively in the past decades (Schwartz *et al.*, 2017; Vidal, 2018; Barkham, 2019; Holl and Brancalion, 2020; The Bonn Challenge, 2020). However, ensuring the success and hence benefits of reforested areas is far more expensive and time demanding than was once thought and project failure rates are high (Mansourian *et al.*, 2006; Fagan *et al.*, 2020; Holl and Brancalion, 2020).

Nevertheless, Planting trees remains an effective GGRT (Bastin *et al.*, 2019; Lewis *et al.*, 2019; Hong *et al.*, 2020) and reforestation and afforestation together are estimated to capture around 1.1 Gt C yr⁻¹ (Smith *et al.*, 2016; Fuss *et al.*, 2018). Reforestation has many side benefits, such as water regulation, improved air quality, biodiversity conservation and income generation (Smith *et al.*, 2019). But it can also have negative consequences if poorly executed, such as increased wildfire risks, aggravating inequalities and damage to nearby crops (Brack and King, 2020).

Although being one of the oldest GGRT, reforestation practices are still discussed and debated, mainly about the form they can take (e.g. plantations, natural parks) (Brack and King, 2020), their GGR potential, and the methodology to account for their delayed capture, reversibility and “sink saturation” (in this case, the tree decrease C uptake rate as they mature (Hansen *et al.*, 2017)). Indeed, forest based GGRTs are characterised by a low permanence dependability and high saturation, even compared to biochar and enhanced weathering, and wood harvest and end uses have major implications on its GGRT potential (Fuss *et al.*, 2018; Ferreira *et al.*, 2020). Hence, although the efficacy of

reforestation as a GGRT is agreed upon, its impact and lasting effect are highly dependent on management options, accounting methodology, and environmental and socio-political changes (Brandão *et al.*, 2013; Anderson *et al.*, 2019; Brack and King, 2020; Fagan *et al.*, 2020).

1.6 Life Cycle Assessment

Life cycle assessment (LCA) is a tool used to evaluate the environmental impacts of products or systems throughout their life cycle (International Organization for Standardization, 2006a, 2006b). An LCA study is composed of four phases: (1) the statement of the objectives and boundaries of the study (goal and scope definition); (2) the collection of inputs and outputs data of the system under study (life cycle inventory analysis); (3) life cycle impact assessment, consisting of the analysis of the processes inside the product or system studied to better understand their environmental impacts; and (4) the interpretation phase, summarizing and discussing the results from phase two and three and providing recommendations based on the goal defined in phase one (International Organization for Standardization, 2006a).

Although LCA has been increasingly used in the past few years, its application for GGRTs is challenging. For instance, there are no agreements yet on accounting methodologies regarding delayed emissions or temporary C storage in climate change related impact categories (Brandão *et al.*, 2013; Goglio *et al.*, 2020). In addition, as LCAs typically rely on available data, and since all GGRTs are at different stage of technological readiness, comparisons between practices can be complicated and unreliable (Goglio *et al.*, 2020). These challenges can lead to miscalculation of the impacts of the assessed technology and have far-reaching consequences if the compromised LCA results are handled as argument in a decision-making process (Tanzer and Ramírez, 2019). However, despite the concerns and difficulties, assessment of GGRT using LCA has been deemed more effective than using other methodologies, in particular because of its potential to assess multiple environmental impact simultaneously (Goglio *et al.*, 2020).

Tanzer and Ramirez (2019) postulated that there are four necessary conditions to know whether or not a GGRT results in net negative emissions:

1. *“Physical greenhouse gases are removed from the atmosphere.”*
2. *“The removed gases are stored out of the atmosphere in a manner intended to be permanent.”*
3. *“Upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance.”*
4. *“The total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere.”* (Tanzer and Ramírez, 2019, p1216)

While each GGRT is recognised to capture CO₂, they all rely on components or processes requiring some form of energy, releasing GHG (Tanzer and Ramírez, 2019). In addition, as these components are merged into systems, their uncertainties and the risk of attendant impacts increase (Tanzer and Ramírez, 2019; Honegger *et al.*, 2020) and care should be taken to ensure that these complex systems indeed act as net carbon sink. However, a 2019 systematic review of NET related literature showed that, in the 286 papers analysed, only five acknowledged that all emissions associated with the implementation and use of the NET are needed to state if the practice truly reduces levels of atmospheric CO₂ (Tanzer and Ramírez, 2019).

Despite current debates and criticisms, the holistic view of LCA grants the practitioner the ability to observe the systems or products assessed in their entirety accounting for all inputs and outputs, which allows for more accurate representation of environmental impacts (or net carbon capture potential in the case of GGRTs) and the identification of the most important processes. A recent publication considers its use for identifying suitability and sustainability of context specific GGRT projects a key policy challenge, along with the acceleration of conventional abatement actions and the acceleration of the development and deployment of these identified sustainable GGRTs (Brack and King, 2020).

1.7 Aims and Objectives

The work presented in this thesis aimed to apply life cycle assessment to assess the net carbon capture potential of three land-based greenhouse gas sequestration practices (i.e.

CHAPTER 1

enhanced weathering, biochar, and reforestation) applied to specific contexts, discuss their net capture potential and uncover limitations and/or caveats.

Specific hypothesis for each of the GGRT assessed, were as follows:

- Enhanced weathering
 1. The environmental impacts associated with the setup of the practice reduces the sequestration estimates reported in the literature.
 2. Enhanced weathering using basalt rock in São Paulo state, Brazil, has a high carbon capture potential.
- Biochar
 3. Biochar produced from available sugarcane residues in São Paulo state, Brazil, has a high carbon sequestration potential.
 4. Replacement of combined heat and power units by pyrolysis plants producing biochar and energy lowers the carbon sequestration potential of the practice.
- Reforestation
 5. The act of planting trees emits GHG
 6. The immediate and permanent carbon GHG capture credits attributed to reforestation projects are not a representative nor a reliable accounting method

The overall hypotheses encompassing this work are

- The use of LCA as a GHG accounting method allows for the identification of the most emitting processes within the system studied.
- The net GGR potential of a system is highly context specific due to the location dependant emission factors of the processes necessary for its implementation.

1.8 Thesis Structure

The main chapters of this thesis are presented as a series of papers. The papers are presented identically to the accepted or submitted versions except for the formatting styles, which were adapted for consistency. An overview of the paper-based chapters is presented in Table 1.1.

Chapter 2 focuses on a life cycle assessment of the practice of enhance weathering in the state of São Paulo, Brazil. It compares the environmental cost and carbon sequestration

potential of the practice, uncovers its limitations, and offers a carbon capture potential values applicable to the state of São Paulo.

Chapter 3 investigates the carbon capture potential of sugarcane residues when turned into biochar and applied back to the soil over the state of São Paulo, Brazil. Biochar impact on soil carbon stock was integrated in a modified version of a soil carbon model, and applied on a spatially explicit way over all sugarcane fields of the state, accounting for differences in climate and soil type.

Chapter 4 builds upon Chapter 3 and assess the carbon footprint of the changes necessary to produce biochar from sugarcane residues. The scenarios are based on different marginal electricity mixes for São Paulo State. This chapter refines the values calculated during Chapter 3 and provides a more objective estimate of the carbon capture potential of turning the available sugarcane residues into biochar and apply it back onto the sugarcane fields.

Chapter 5 assesses the carbon footprint of reforesting a hectare of Amazonian rainforest in southeast Peru. The LCA is coupled with a soil carbon model and an above- and below-ground carbon model to represent the capture potential of the practice. The results are then compared with carbon capture claims made by a hypothetical reforestation platform.

Table 1.1: Status of paper-based chapters

Chapter	Paper	Title	Journal	Status	Reference
2	1	Assessing the potential of soil carbonation and enhanced weathering through Life Cycle Assessment: A case study for São Paulo State, Brazil	Journal of Cleaner Production	Published	(Lefebvre <i>et al.</i> , 2019)
3	2	Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil	Scientific Reports	Published	(Lefebvre <i>et al.</i> , 2020)
4	3	An anticipatory life cycle assessment of the use of biochar from sugarcane	Journal of Cleaner Production	Published	(Lefebvre <i>et al.</i> , 2021)

Chapter	Paper	Title	Journal	Status	Reference
5	4	residues as a greenhouse gas removal technology Assessing the carbon capture potential of reforestation projects	Nature Communications	Submitted	/

1.9 References

Anderson, C. M. *et al.* (2019) ‘Natural climate solutions are not enough’, *Science*, 363(6430), pp. 933–934. doi: 10.1126/science.aaw2741.

Anderson, K. and Peters, G. (2016) ‘The Trouble With Negative Emissions’, *Climate Change*, 354(6309), pp. 182–184. doi: 10.1126/science.aah4567.

Bamminger, C. *et al.* (2016) ‘Short-term response of soil microorganisms to biochar addition in a temperate agroecosystem under soil warming’, *Agriculture, Ecosystems and Environment*. Elsevier B.V., 233, pp. 308–317. doi: 10.1016/j.agee.2016.09.016.

Barkham, P. (2019) ‘Can planting billions of trees save the planet?’, *The Guardian*. Available at: <https://www.theguardian.com/world/2019/jun/19/planting-billions-trees-save-planet> (Accessed: 26 June 2020).

Bastin, J. F. *et al.* (2019) ‘The global tree restoration potential’, *Science*, 364(6448), pp. 76–79. doi: 10.1126/science.aax0848.

Beerling, D. J. *et al.* (2018) ‘Farming with crops and rocks to address global climate, food and soil security’, *Nature Plants*, 4(3), pp. 138–147. doi: 10.1038/s41477-018-0108-y.

Beerling, D. J. *et al.* (2020) ‘Potential for large-scale CO₂ removal via enhanced rock weathering with croplands’, *Nature*. Springer US, 583(7815), pp. 242–248. doi: 10.1038/s41586-020-2448-9.

Berner, R. A. and Kothavala, Z. (2001) ‘GEOCARB III; A Revised Model of Atmospheric CO₂ Over Phanerozoic Time’, *American Journal of Science*, 301(1), pp. 182–204. doi: 10.2475/ajs.294.1.56.

Brack, D. and King, R. (2020) ‘Managing Land-based CDR: BECCS, Forests and Carbon

Sequestration’, *Global Policy*. doi: 10.1111/1758-5899.12827.

Brandão, M. *et al.* (2013) ‘Key issues and options in accounting for carbon sequestration and temporary storage in life cycle assessment and carbon footprinting’, *The International Journal of Life Cycle Assessment*, 18(1), pp. 230–240. doi: 10.1007/s11367-012-0451-6.

Brander, M. *et al.* (2021) ‘Carbon accounting for negative emissions technologies’, *Climate Policy*. Taylor & Francis, 0(0), pp. 1–19. doi: 10.1080/14693062.2021.1878009.

Brown, C. *et al.* (2019) ‘Achievement of Paris climate goals unlikely due to time lags in the land system’, *Nature Climate Change*. Springer US, 9(3), pp. 203–208. doi: 10.1038/s41558-019-0400-5.

Carton, W. *et al.* (2020) ‘Negative emissions and the long history of carbon removal’, *WIREs Climate Change*, 11(6), pp. 1–25. doi: 10.1002/wcc.671.

de Coninck, H. and Revi, A. (2018) ‘Chapter 4 - Strengthening and implementing the global response’, in *Global Warming of 1.5 C an IPCC special report*. Available at: https://www.ipcc.ch/site/assets/uploads/sites/2/2019/05/SR15_Chapter4_Low_Res.pdf.

Cox, E., Spence, E. and Pidgeon, N. (2020) ‘Public perceptions of carbon dioxide removal in the United States and the United Kingdom’, *Nature Climate Change*. Springer US, 10(8), pp. 744–749. doi: 10.1038/s41558-020-0823-z.

Ding, F. *et al.* (2018) ‘A meta-analysis and critical evaluation of influencing factors on soil carbon priming following biochar amendment’, *Journal of Soils and Sediments*. Journal of Soils and Sediments, 18(4), pp. 1507–1517. doi: 10.1007/s11368-017-1899-6.

Domene, X. (2016) ‘A Critical Analysis of Meso- and Macrofauna Effects Following Biochar Supplementation’, in *Biochar Application: Essential Soil Microbial Ecology*, pp. 268–292. doi: 10.1016/B978-0-12-803433-0.00011-4.

Fagan, M. E. *et al.* (2020) ‘How feasible are global forest restoration commitments?’, *Conservation Letters*, 13(3), pp. 1–8. doi: 10.1111/conl.12700.

Ferreira, J. *et al.* (2020) ‘Life Cycle Assessment of Maritime Pine Wood: A Portuguese Case Study’, *Journal of Sustainable Forestry*. Taylor & Francis, 00(00), pp. 1–15. doi:

CHAPTER 1

10.1080/10549811.2020.1768871.

Forster, J. *et al.* (2020) 'Mapping feasibilities of greenhouse gas removal: Key issues, gaps and opening up assessments', *Global Environmental Change*. Elsevier Ltd, 63(May), p. 102073. doi: 10.1016/j.gloenvcha.2020.102073.

Fuss, S. *et al.* (2014) 'Betting on negative emissions', *Nature Climate Change*. Nature Publishing Group, 4(10), pp. 850–853. doi: 10.1038/nclimate2392.

Fuss, S. *et al.* (2018) 'Negative emissions — Part 2: Costs , potentials and side effects', *Environmental Research Letters*, 13 063002(063002). doi: 10.1088/1748-9326/aabf9f.

Gao, S., DeLuca, T. H. and Cleveland, C. C. (2019) 'Biochar additions alter phosphorus and nitrogen availability in agricultural ecosystems: A meta-analysis', *Science of the Total Environment*. Elsevier B.V., 654, pp. 463–472. doi: 10.1016/j.scitotenv.2018.11.124.

Gasser, T. *et al.* (2015) 'Negative emissions physically needed to keep global warming below 2°C', *Nature Communications*. Nature Publishing Group, 6. doi: 10.1038/ncomms8958.

Goglio, P. *et al.* (2020) 'Advances and challenges of life cycle assessment (LCA) of greenhouse gas removal technologies to fight climate changes', *Journal of Cleaner Production*. Elsevier Ltd, 244, p. 118896. doi: 10.1016/j.jclepro.2019.118896.

Griscom, B. W. *et al.* (2017) 'Natural climate solutions', *Proceedings of the National Academy of Sciences of the United States of America*, 114(44), pp. 11645–11650. doi: 10.1073/pnas.1710465114.

Hagemann, N. *et al.* (2017) 'Organic coating on biochar explains its nutrient retention and stimulation of soil fertility', *Nature Communications*. Springer US, 8(1), p. 1089. doi: 10.1038/s41467-017-01123-0.

Han Weng, Z. *et al.* (2017) 'Biochar built soil carbon over a decade by stabilizing rhizodeposits', *Nature Climate Change*, 7(5), pp. 371–376. doi: 10.1038/nclimate3276.

Hansen, J. *et al.* (2017) 'Young people's burden: requirement of negative CO₂ emissions', *Earth System Dynamics*, 8(3), pp. 577–616. doi: 10.5194/esd-8-577-2017.

Holl, K. D. and Brancalion, P. H. S. (2020) ‘Tree planting is not a simple solution’, *Science*, 368(6491), pp. 580–581. doi: 10.1126/science.aba8232.

Honegger, M., Michaelowa, A. and Roy, J. (2020) ‘Potential implications of carbon dioxide removal for the sustainable development goals’, *Climate Policy*. Taylor & Francis, 0(0), pp. 1–21. doi: 10.1080/14693062.2020.1843388.

Hong, S. *et al.* (2020) ‘Divergent responses of soil organic carbon to afforestation’, *Nature Sustainability*. doi: 10.1038/s41893-020-0557-y.

International Organization for Standardization (2006a) *Environmental management - Life cycle assessment - Principles and framework - ISO 14040*. Available at: <https://www.iso.org/fr/home.html>.

International Organization for Standardization (2006b) *Environmental management Life cycle assessment Requirements and guidelines - ISO 14044*. Available at: <https://www.iso.org/fr/home.html>.

IPCC (2014) *Climate Change 2014: Mitigation of Climate Change, Working Group III Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. doi: 10.1017/CBO9781107415416.

IPCC (2018) *Global Warming of 1.5°C - Special Report - Summary for Policymakers*. Edited by Intergovernmental Panel on Climate Change. Available at: https://www.ipcc.ch/site/assets/uploads/sites/2/2018/07/SR15_SPM_High_Res.pdf (Accessed: 2 January 2019).

IPCC (2019) *Climate Change and Land - An IPCC Special Report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems - Summary for Policymakers, IPCC*. doi: 10.4337/9781784710644.00020.

IPCC (2020) *Climate Change and Land - Summary for policy makers, Climate Change and Land: an IPCC special report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems*. Available at: https://www.ipcc.ch/site/assets/uploads/sites/4/2020/02/SPM_Updated-Jan20.pdf.

CHAPTER 1

Kamali, M. *et al.* (2020) ‘Scientometric analysis and scientific trends on biochar application as soil amendment’, *Chemical Engineering Journal*. Elsevier, 395(November 2019), p. 125128. doi: 10.1016/j.cej.2020.125128.

Kolosz, B. W., Sohi, S. p. and Manning, D. A. C. (2019) ‘CASPER: A modelling framework to link mineral carbonation with the turnover of organic matter in soil’, *Computers and Geosciences*. Elsevier Ltd, 124(January), pp. 58–71. doi: 10.1016/j.cageo.2018.12.012.

Kriegler, E. *et al.* (2018) ‘Pathways limiting warming to 1.5°C: a tale of turning around in no time?’, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 376(2119), p. 20160457. doi: 10.1098/rsta.2016.0457.

Laird, D. A. *et al.* (2009) ‘Review of the pyrolysis platform for coproducing bio-oil and biochar’, *Biofuels, Bioproducts and Biorefining*, 3(3), pp. 574–562. doi: 10.1002/bbb.169.

Lefebvre, D. *et al.* (2019) ‘Assessing the potential of soil carbonation and enhanced weathering through Life Cycle Assessment: A case study for Sao Paulo State, Brazil’, *Journal of Cleaner Production*. Elsevier Ltd, 233, pp. 468–481. doi: 10.1016/j.jclepro.2019.06.099.

Lefebvre, D. *et al.* (2020) ‘Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil’, *Scientific Reports*. Nature Publishing Group UK, 10(1), p. 19479. doi: 10.1038/s41598-020-76470-y.

Lefebvre, D. *et al.* (2021) ‘An anticipatory life cycle assessment of the use of biochar from sugarcane residues as a greenhouse gas removal technology’, *Journal of Cleaner Production*, p. 127764. doi: 10.1016/j.jclepro.2021.127764.

Lehmann, J., Gaunt, J. and Rondon, M. (2006) ‘Bio-char sequestration in terrestrial ecosystems - A review’, *Mitigation and Adaptation Strategies for Global Change*, 11(2), pp. 403–427. doi: 10.1007/s11027-005-9006-5.

Lewis, S. L. *et al.* (2019) ‘Restoring natural forests is the best way to remove atmospheric carbon’, *Nature*, 568(7750), pp. 25–28. doi: 10.1038/d41586-019-01026-8.

- Liu, Q. *et al.* (2018) 'How does biochar influence soil N cycle? A meta-analysis', *Plant and Soil*. Plant and Soil, 426(1–2), pp. 211–225. doi: 10.1007/s11104-018-3619-4.
- Liu, X. *et al.* (2019) 'Impact of biochar application on yield-scaled greenhouse gas intensity: A meta-analysis', *Science of the Total Environment*. Elsevier B.V., 656(127), pp. 969–976. doi: 10.1016/j.scitotenv.2018.11.396.
- Mace, M. J. *et al.* (2021) *Governing large-scale carbon dioxide removal: are we ready? - an update*. New York. Available at: <https://www.c2g2.net/governing-large-scale-carbon-dioxide-removal-are-we-ready/>.
- Manning, D. A. C. *et al.* (2013) 'Carbonate precipitation in artificial soils produced from basaltic quarry fines and composts: An opportunity for passive carbon sequestration', *International Journal of Greenhouse Gas Control*. Elsevier Ltd, 17, pp. 309–317. doi: 10.1016/j.ijggc.2013.05.012.
- Mansourian, S., Vallauri, D. and Dudley, N. (2006) 'Forest restoration in landscapes: beyond planting trees', *Landscape Ecology*, 22(3), pp. 477–479. doi: 10.1007/s10980-006-9029-7.
- McLaren, D. P. *et al.* (2019) 'Beyond “Net-Zero”: A Case for Separate Targets for Emissions Reduction and Negative Emissions', *Frontiers in Climate*, 1(August), pp. 1–5. doi: 10.3389/fclim.2019.00004.
- Minx, J. C. *et al.* (2017) 'Fast growing research on negative emissions', *Environmental Research Letters*, 12(3), p. 035007. doi: 10.1088/1748-9326/aa5ee5.
- Minx, J. C. *et al.* (2018) 'Negative emissions—Part 1: Research landscape and synthesis', *Environmental Research Letters*, 13(6), p. 063001. doi: 10.1088/1748-9326/aabf9b.
- Moosdorf, N., Renforth, P. and Hartmann, J. (2014) 'Carbon Dioxide Efficiency of Terrestrial Enhanced Weathering', *Environmental science & technology*, 48, pp. 4809–4816. doi: 10.1021/es4052022.
- Ng, W. Y. *et al.* (2020) 'Ranking negative emissions technologies under uncertainty', *Heliyon*, 6(12), p. e05730. doi: 10.1016/j.heliyon.2020.e05730.
- Renforth, P. (2012) 'The potential of enhanced weathering in the UK', *International*

CHAPTER 1

Journal of Greenhouse Gas Control. Elsevier Ltd, 10, pp. 229–243. doi: 10.1016/j.ijggc.2012.06.011.

Rogelj, J. *et al.* (2018) ‘Scenarios towards limiting global mean temperature increase below 1.5 °C’, *Nature Climate Change*. Springer US, 8(4), pp. 325–332. doi: 10.1038/s41558-018-0091-3.

Ruseva, T. *et al.* (2020) ‘Rethinking standards of permanence for terrestrial and coastal carbon: implications for governance and sustainability’, *Current Opinion in Environmental Sustainability*, 45, pp. 69–77. doi: 10.1016/j.cosust.2020.09.009.

Schwartz, N. B. *et al.* (2017) ‘Land-use dynamics influence estimates of carbon sequestration potential in tropical second-growth forest’, *Environmental Research Letters*, 12(7), p. 074023. doi: 10.1088/1748-9326/aa708b.

Smith, P. *et al.* (2016) ‘Biophysical and economic limits to negative CO₂ emissions’, *Nature Climate Change*. Nature Publishing Group, 6(1), pp. 42–50. doi: 10.1038/nclimate2870.

Smith, P. (2016) ‘Soil carbon sequestration and biochar as negative emission technologies’, *Global Change Biology*, 22(3), pp. 1315–1324. doi: 10.1111/gcb.13178.

Smith, P. *et al.* (2019) ‘Land-Management Options for Greenhouse Gas Removal and Their Impacts on Ecosystem Services and the Sustainable Development Goals’, *Annual Review of Environment and Resources*, 44(1), pp. 255–286. doi: 10.1146/annurev-environ-101718-033129.

Tanzer, S. E. and Ramírez, A. (2019) ‘When are negative emissions negative emissions?’, *Energy & Environmental Science*, 12(4), pp. 1210–1218. doi: 10.1039/C8EE03338B.

Taylor, L. L. *et al.* (2016) ‘Enhanced weathering strategies for stabilizing climate and averting ocean acidification’, *Nature Climate Change*, 6(4), pp. 402–406. doi: 10.1038/nclimate2882.

Taylor, L. L. *et al.* (2017) ‘Simulating carbon capture by enhanced weathering with croplands: An overview of key processes highlighting areas of future model development’, *Biology Letters*, 13(4). doi: 10.1098/rsbl.2016.0868.

The Bonn Challenge (2020) *The Bonn challenge - About the challenge*. Available at: <https://www.bonnchallenge.org/about> (Accessed: 22 March 2021).

Tisserant, A. and Cherubini, F. (2019) 'Potentials, Limitations, Co-Benefits, and Trade-Offs of Biochar Applications to Soils for Climate Change Mitigation', *Land*, 8(12), p. 179. doi: 10.3390/land8120179.

UNFCCC (1997) *United Nations Kyoto Protocol*. Available at: <https://unfccc.int/sites/default/files/resource/docs/cop3/107a01.pdf>.

UNFCCC (2015) *Paris Agreement, UNFCCC*. Available at: https://unfccc.int/files/essential_background/convention/application/pdf/english_paris_agreement.pdf.

Vidal, J. (2018) 'A eureka moment for the planet: we're finally planting trees again', *The Guardian*, 13 February. Available at: <https://www.theguardian.com/commentisfree/2018/feb/13/worlds-lost-forests-returning-trees> (Accessed: 9 March 2019).

Van Vuuren, D. P. *et al.* (2018) 'Alternative pathways to the 1.5 °C target reduce the need for negative emission technologies', *Nature Climate Change*. Springer US, 8(5), pp. 391–397. doi: 10.1038/s41558-018-0119-8.

Wang, J. *et al.* (2016) 'Biochar stability in soil: Meta-analysis of decomposition and priming effects', *GCB Bioenergy*, 8(3), pp. 512–523. doi: 10.1111/gcbb.12266.

Washbourne, C. L. *et al.* (2015) 'Rapid removal of atmospheric CO₂ by urban soils', *Environmental Science and Technology*, 49(9), pp. 5434–5440. doi: 10.1021/es505476d.

Zhang, Y., Wang, J. and Feng, Y. (2021) 'The effects of biochar addition on soil physicochemical properties: A review', *CATENA*. Elsevier B.V., 202(October 2020), p. 105284. doi: 10.1016/j.catena.2021.105284.

CHAPTER 2

Enhanced Weathering LCA

“However beautiful the strategy, you should occasionally look at the results.”

WINSTON CHURCHILL

ASSESSING THE POTENTIAL OF SOIL CARBONATION AND ENHANCED WEATHERING THROUGH LIFE CYCLE ASSESSMENT: A CASE STUDY FOR SÃO PAULO STATE, BRAZIL

David Lefebvre^a, Pietro Goglio^{a,b}, Adrian Williams^a, David A. C. Manning^c, Antonio Carlos de Azevedo^d, Magda Bergmann^e, Jeroen Meersmans^a, Pete Smith^f

^a School of Water, Energy and Environment, Cranfield University, College Road, Bedford, MK43 0AL, UK

^b Wageningen Economic Research, Wageningen University & Research, Leeuwenborch, Hollandsweg 1, 6706KN Wageningen, The Netherlands

^c School of Natural and Environmental Sciences, Newcastle University, Newcastle upon Tyne NE1 7RU, UK

^d USP/ESALQ/LSO, Av. Padua Dias, 11, 13415900-Piracicaba-SP, Brasil

^e Geological Survey of Brazil (CPRM), Rua Banco da Provincia 105, Porto Alegre, CEP 90840-030 Rio Grande do Sul, Brazil

^f Institute of Biological and Environmental Sciences, University of Aberdeen, 23 St Machar Drive, Aberdeen AB24 3UU, UK

2.1 Abstract

Enhanced silicate rock weathering for long-term carbon dioxide sequestration has considerable potential, but depends on availability of suitable rocks coupled with proximity to suitable locations for field application. In this paper, we investigate the established mining industry that extracts basaltic rocks for construction from the Paraná Basin, São Paulo State, Brazil. Through a Life Cycle Assessment, we determine the balance of carbon dioxide emissions involved in the use of this material, the relative contribution of soil carbonation and enhanced weathering, and the potential carbon dioxide removal of São Paulo agricultural land through enhanced weathering of basalt rock.

Our results show that enhanced weathering and carbonation respectively emit around 75 and 135 kg carbon dioxide equivalent per tonne of carbon dioxide equivalent removed (considering a quarry to field distance of 65km). We underline transportation as the principal process negatively affecting the practice and uncover a limiting road travel distance from the quarry to the field of 540 ± 65 km for carbonation and 990 ± 116 km for enhanced weathering, above which the emissions offset the potential capture. Regarding São Paulo State, the application of crushed basalt at 1 t/ha to all of the State's

12 million hectares of agricultural land could capture around 1.3 to 2.4 Mt carbon dioxide equivalent through carbonation and enhanced weathering, respectively.

This study suggests a lower sequestration estimate than previous studies and emphasizes the need to consider all process stages through a Life Cycle Assessment methodology, to provide more reliable estimates of the sequestration potential of greenhouse gas removal technologies.

2.2 Introduction

Scientists agree that, by 2100, the annual extraction of an average of 3.3 Gt of carbon equivalent per year (>12 Gt carbon dioxide equivalent [CO₂e] per year) from the atmosphere will be necessary to limit the increase in global average temperature to 2°C relative to pre-industrial levels (Fuss *et al.*, 2014; Smith *et al.*, 2016; Williamson, 2016). To mitigate this, it is not only necessary to prevent greenhouse gas (GHG) emissions, but also to remove GHG from the atmosphere. A greenhouse gas removal technology (GGRT) or negative emission technology is one capable of removing GHG from the atmosphere (Williamson, 2016; Williamson and Bodle, 2016; EASAC, 2018; Fuss *et al.*, 2018).

Enhanced weathering (EW) has received increasing interest in the past few years (Renforth *et al.*, 2011; Martin, 2017); its global potential has been addressed by Beerling *et al.* (2018). It is defined as the “*process by which CO₂ is sequestered from the atmosphere through the dissolution of silicate minerals on the land surface*” (Renforth, 2012, p.229) and is regarded as a potential GGRT. Similarly, carbonation is a process where the formation of carbonate minerals in soils is promoted artificially, mimicking natural pedogenic carbonate formation to produce a measurable permanent sink for atmospheric CO₂ (Manning *et al.*, 2013; Washbourne *et al.*, 2015; Kolosz, Sohi and Manning, 2019).

Weathering of silicate rock has played a significant role over geological time in the regulation of Earth’s climate (Berner and Kothavala, 2001). However, due to the average slow rate of natural rock weathering, its effect on a human time scale is too slow to compensate for artificial changes in atmospheric composition (Hartmann *et al.*, 2009; Taylor *et al.*, 2017). To accelerate the process, EW and carbonation rely on the

comminution of the rock, increasing its reactive surface, hence promoting its dissolution and thus its CO₂ capture rate (Taylor *et al.*, 2017).

Although studied, the magnitude of worldwide carbon capture potential through EW varies depending on the application area considered, type of rock used, and application rate (Moosdorf, Renforth and Hartmann, 2014; Beerling *et al.*, 2018). Life Cycle Assessments (LCA) of the CO₂ sequestration potential of silicate rock exist, however the process studied involves direct fluxes of CO₂ in a reactor at high temperature and pressure (Khoo *et al.*, 2011; Nduagu, Bergerson and Zevenhoven, 2012), which clearly differs from the natural dissolution process under ambient Earth-surface conditions studied here. No study has yet assessed in detail the environmental costs and impacts linked to the natural dissolution processes of this soil-based GGRT. São Paulo State (Brazil) was selected as a case study, because of a combination of unique factors: the geographical colocation of the Paraná flood basalts (Hartmann and Moosdorf, 2012) with large scale agricultural production that is amenable to application of rock dust within the Brazilian regulatory system for remineralizers (Manning and Theodoro, 2018). Our findings can be applied to other regions where suitable rocks are spatially associated with appropriate land management (Beerling *et al.*, 2018).

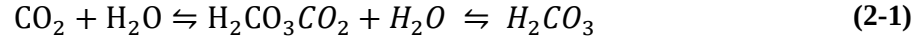
This study i) assesses the environmental impacts of the practice using basalt rock in São Paulo state conditions including the emissions associated with the rock extraction, comminution, transport and application through a Life Cycle Assessment approach; ii) estimates the contribution of the different processes involved with soil carbonation and enhanced weathering and iii) assesses the potential CO₂ removal of São Paulo agricultural land through soil carbonation and weathering of basalt rock. A large number of active mining operations occur within the Paraná flood basalts. These supply aggregate for construction; in providing low-cost products using existing technology, they represent a well-established mining sector that is expected to grow with increasing urbanisation.

2.3 Methodology

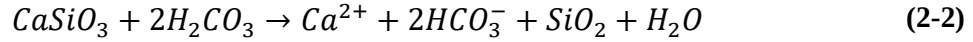
2.3.1 Chemical aspects

The process of rock weathering involves a series of chemical reactions. First, the hydration of CO₂ (Equation (2-1)) forms carbonic acid (Martin, 2017).

CHAPTER 2



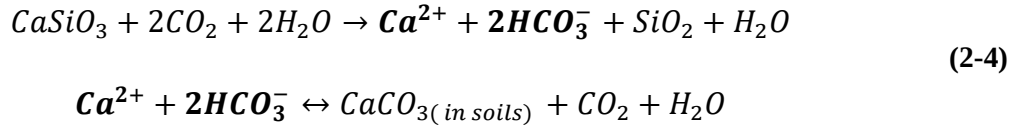
This weak acid breaks down silicate rock, which releases base cations (e.g. Ca^{2+} , Mg^{2+}) and bicarbonate and carbonate ions (Beerling, 2017) in proportion depending on pH. The weathering of silicate minerals is commonly represented using the calcium silicate mineral wollastonite (CaSiO_3), and releases bicarbonate ions (Equation (2-2)).



Following Equation (2-2), when the dissolved bicarbonate ions are then transferred to the ocean and contribute to its alkalinity, two moles of CO_2 are captured per mole of calcium (content depending on the rock). This path will be herein referred to as enhanced weathering (EW) (Equation (2-3)).



However, when soil conditions are favourable, some of the calcium and magnesium cations can precipitate to form carbonate minerals (Equation (2-4)), which accounts for half the moles of CO_2 captured during the weathering of the rock. The latter path will be herein referred to as carbonation (Equation (2-4)).



The capture potential of each path (i.e. Carbonation and EW) depends on the type of rock used. Minerals in ultrabasic rocks, such as olivine (Mg_2SiO_4), weather relatively quickly and show a high potential for CO_2 sequestration (Goldich, 1938; O'Connor *et al.*, 2005). However, several studies suggest that the weathering of olivine-bearing rocks could lead to the release of chromium (Cr) and nickel (Ni) into the environment, which if sufficient, suppresses calcium uptake by plants (ten Berge *et al.*, 2012; Renforth, Pogge von Strandmann and Henderson, 2015) and could be toxic, although they are essential to life in small amounts. Importantly, current mining of olivine-rich rocks is limited to a small number of locations globally.

In addition, basic rocks, such as basalt, occur widely and show great potential for enhanced weathering, despite their lower sequestration potential than olivine (0.3t CO₂e/t for basalt and 0.8t CO₂e/t for olivine in the case of EW) (Beerling *et al.*, 2018; Strefler *et al.*, 2018). Basalt is globally available in large quantities, has a fast weathering rate, and has lower concentrations of Cr and Ni and a higher concentration of phosphorus, thus posing fewer environmental risks through its application (Bryan and Ernst, 2008; Kantola *et al.*, 2017; Beerling *et al.*, 2018). It is widely mined for construction, and used in agriculture for remineralization (Manning and Theodoro, 2018).

2.3.2 Sequestration potential of Brazilian basalt rock

The potential CO₂ capture of basalt rock is calculated using Equation (2-5) from Renforth, (2012).

$$R_{CO_2} = \frac{M_{CO_2}}{100} \left(\frac{\%CaO}{M_{CaO}} + \frac{\%MgO}{M_{MgO}} \right) * \omega \quad (2-5)$$

Where R_{CO_2} is the sequestration potential in tonne CO₂e per tonne of rock; M_{MgO} , M_{CaO} , and M_{CO_2} , as the molecular masses of MgO, CaO and CO₂, respectively; %CaO and %MgO the percent in weight of CaO and MgO in the rock, respectively; and ω , a factor that accounts for the additional sequestration that occurs when the cations are transported to the oceans and remain dissolved.

The potential CO₂ capture through carbonation is calculated using the CaO and MgO content of the rock and assuming the precipitation of the associated cations (i.e. Ca²⁺ and Mg²⁺) into carbonate minerals (i.e. calcite and magnesite). Since no cations are transferred into the ocean, the value of ω for carbonation is one. Average MgO and CaO contents were computed from 48 measurements of Brazilian basalt (Baggio, Hartmann and Bello, 2016; Braz Machado *et al.*, 2018; Moraes, Seer and Marques, 2018) and averaged as 4.87 and 9.14 % by weight, respectively, leading to a capture potential through carbonation of 0.125 t CO₂e /t of basalt.

CO₂ capture through enhanced weathering assumes that all the cations, resulting from the dissolution of the rock, remain dissolved and are transferred to the ocean. Only ω changes in equation (2-5). According to Kheshgi, (1995), when all cations remain dissolved, the value of ω reaches 1.79 which means that 1.79 moles of CO₂ is sequestered per mole of

CHAPTER 2

CaO or MgO. Using the value of 1.79 for ω , the potential sequestration through EW reaches 0.225 t CO₂e /t of basalt. A ω value of 1.7 was used in other publications on enhanced weathering (Harvey, 2008; Renforth, 2012). Overall, the widely used sequestration value through EW of 0.3 t CO₂/ t of basalt (Kantola *et al.*, 2017; Beerling *et al.*, 2018; Streffler *et al.*, 2018) results from equation (2-5) using different average values for CaO and MgO content and a value of 1.7 for ω .

This study considers both the carbonation and EW pathway. Although carbonation is likely after the application of the rock on the field (Manning and Renforth, 2013), EW is expected to happen simultaneously and further dissolution of the calcite and magnesite produced through carbonation may also occur (Landi, Mermut and Anderson, 2003). Therefore, the sequestration potentials through carbonation and EW presented here represent lower and upper limits, respectively. The actual sequestration value after the application of one tonne of basalt on a field is likely to be between these two limits. The sequestration rate through carbonation or EW depends on the rock weathering rate that varies greatly depending on various factors. Although not studied here, the matter is discussed later.

Figure 2.1 provides an overview of the weathering process as well as the two considered paths and their sequestration potentials for Brazilian basalt rock.

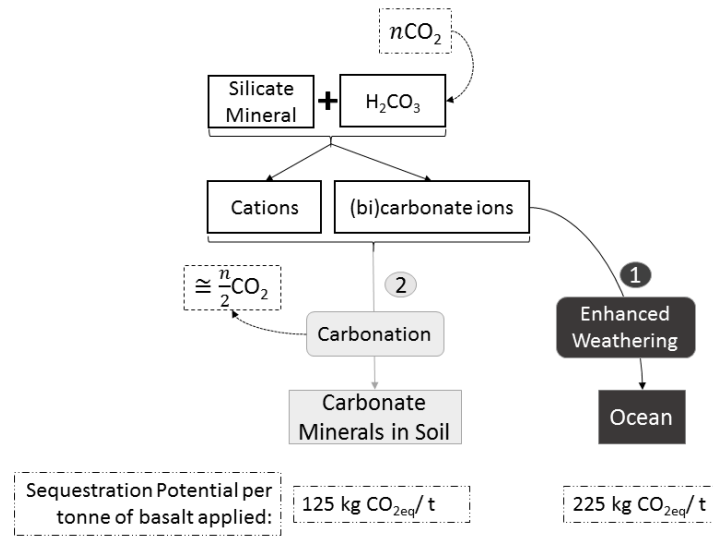


Figure 2.1: Overview of the rock weathering process, the two pathways (carbonation and EW) considered in this study and the sequestration potential of Brazilian basalt for each considered path.

2.3.3 Study area

We selected São Paulo (SP) State due to its situation on an area with large basalt deposits (Bryan and Ernst, 2008; Hartmann and Moosdorf, 2012), the resulting occurrence of basalt quarries (Perrotta *et al.*, 2006; DNPM, 2018), and the climatic and soil conditions for rapid rock weathering (Edwards *et al.*, 2017; Meysman and Montserrat, 2017). Further, the practice is already used by farmers in the area as a soil remineralizer.

Figure 2.2 shows SP with the locations of basalt quarries, cropland, and grassland inside the state. Concentric buffer zones, centred on the quarries, represent straight-line distances between quarries and agricultural areas. The distance increments were selected arbitrarily. The maximum distance of 250 km was selected because it covers the totality of São Paulo's agricultural area.

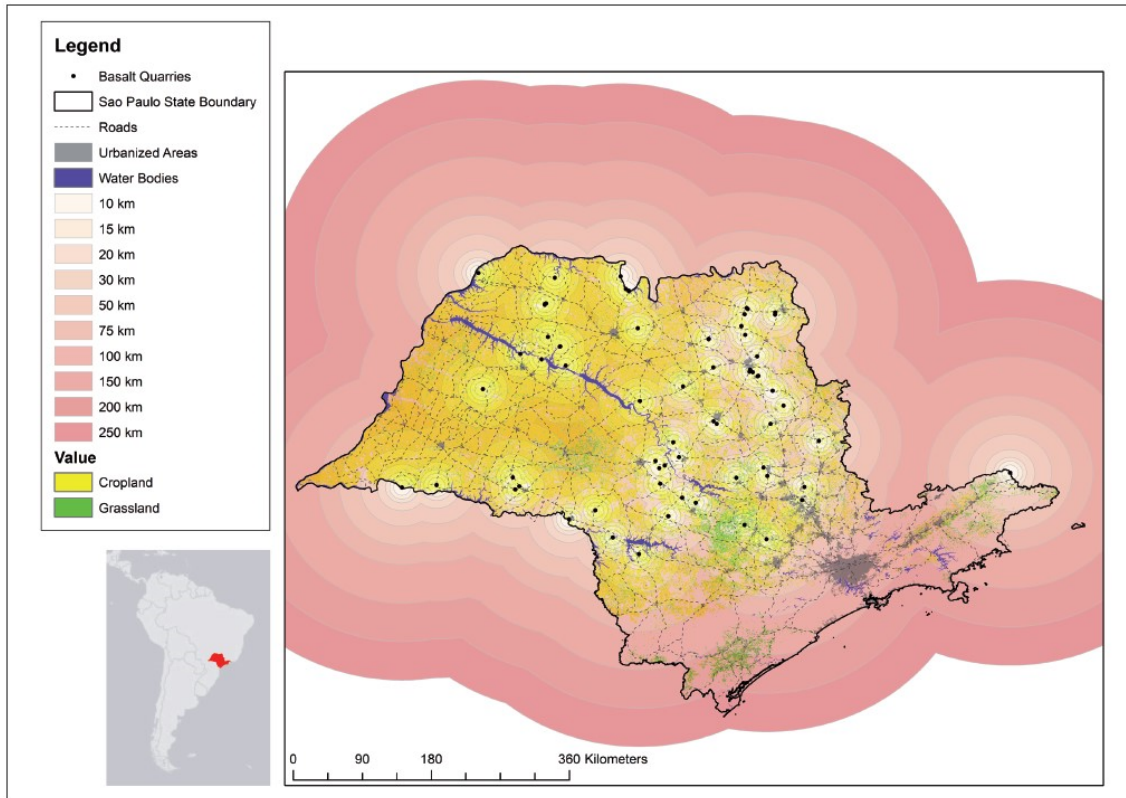


Figure 2.2: São Paulo state with buffer zones (red shade) around São Paulo's basalt quarries overlapping grassland and cropland areas.

The average quarry to field distance of 50 km was calculated by summing the product of each distance increment by the agricultural area it covers, and dividing the result by the total agricultural area of SP.

Data on land use was provided by the “ESA Climate Change Initiative” (UCLouvain, 2017) and the area of cropland and grassland reported is consistent with the Brazilian Institute of Geography and Statistics (IBGE, 2017). The location of the basalt quarries in SP was provided by the “National Geology Program of Brazil” (Perrotta *et al.*, 2006).

2.3.4 Objectives and system boundary

To assess the environmental impacts of basalt rock application the following processes were considered within the system boundary: the extraction of the material, its transport to the grinding facility, its comminution into particles of <5 mm, its transport from quarries to the fields, and its spreading on the field using agricultural spreaders (Figure

2.3). This LCA does not take into account any soil or crop response following the field application. Each process stage is described below.

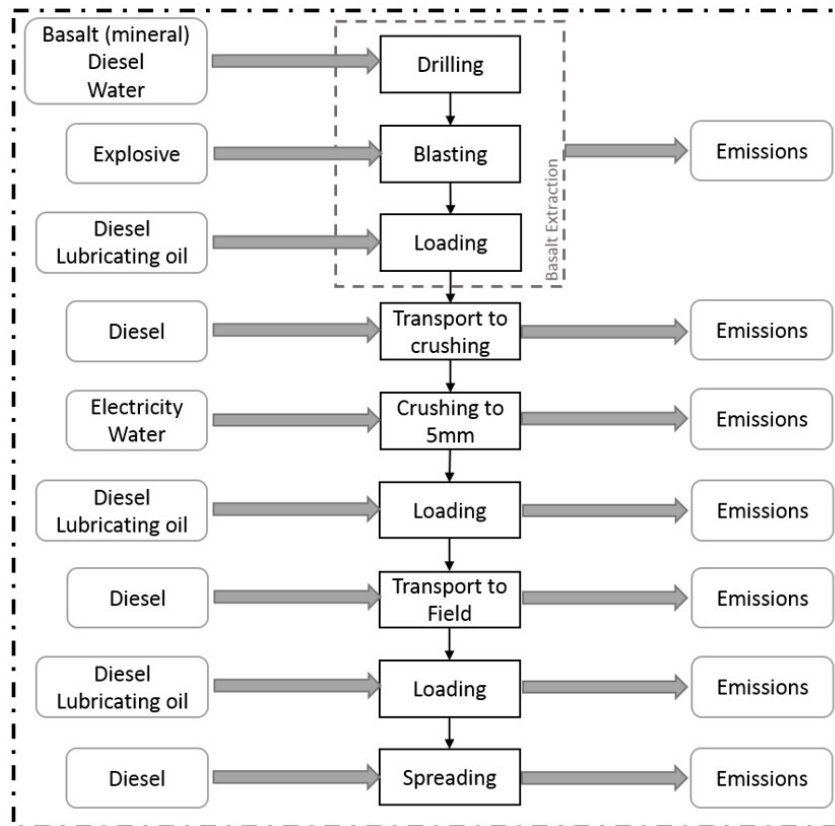


Figure 2.3: System boundaries for extraction, comminution, transport and spreading of basalt considered in this study.

The functional units (FU) of this LCA are i) per hectare of SP agricultural land amended by <5mm basalt particles, and ii) per tonne of CO₂e removed.

Distances from quarry to field are represented nominally in straight lines in our map (Figure 2.2). Road tortuosity ratio is a coefficient used to consider the tortuosity of the road network of an area. Multiplying the coefficient by the linear distance between two points gives a more accurate estimate of journey length through the road network.

We found a tortuosity ratio of 1.3 by comparing 30 actual road (randomly selected) and straight-line distances inside the state. We then multiplied every linear distance by this ratio to calculate the transport impacts. The average quarry to field distance of 50 km discussed earlier becomes 65 km once accounting for the road tortuosity of São Paulo state.

CHAPTER 2

We considered three basalt application rates: 5, 50, and 20 t/ha. The first application rate is representative of what is currently spread by some farmers in São Paulo (according to two local experts) as well as the minimum recommended rate according to two UK rock dust retailers (REMIN, no date; Ramezani *et al.*, 2013). The second application rate of 50 t/ha represents the maximum dose considered in the literature for carbon sequestration purposes (Beerling *et al.*, 2018). The intermediate application rate of 20 t/ha was selected because it has already been used in previous field research in São Paulo and corresponds to the recommended application rate according to a UK rock dust retailer (REMIN (Scotland) Ltd, n.d.).

2.3.5 Software, database & data processing

Most of the activity data regarding quarry processes in our Life Cycle Inventory are based on Rosado *et al.* (2017). The values used for our analysis are given in Table 2.1.

Table 2.1: Database and process used in the LCI of basalt extraction, comminution, transport and spreading

Process Stage	SimaPro 8.3.0.0 Process	Updates	Unit	Amount [Mg ⁻¹]
Basalt Extraction				
Basalt Drilling	Loader operation, large, ^a	Diesel and oil consumption	hour	0.00600
Blasting	Blasting ^b processing ^c	Mass of explosive	kg	0.145
Loading Operation				
Front Loader	Loader operation, large, ^a	Diesel and oil consumption	hour	0.00600
Transport to Crushing				
Lorry	Transport, freight, lorry 16-32 metric ton, EURO4 ^b ^c	/	tkm	1
Crushing to 5mm				
Electricity	Electricity, medium voltage {BR} electricity voltage transformation from high to medium voltage ^c	/	kWh	5.45

Process Stage	SimaPro 8.3.0.0 Process	Updates	Unit	Amount [Mg ⁻¹]
	Electricity, medium voltage {BR} market for ^c			
Loading Operation				
Front Loader	Loader operation, large, ^a	Diesel and oil consumption	hour	0.00600
Transport to field				
Lorry	Transport, freight, lorry 16-32 metric ton, EURO4 ^b ^c	/	tkm	65
Loading Operation				
Front Loader	Loader operation, large, ^a	Diesel and oil consumption	hour	0.00600
Spreading on field				
Diesel in Agri Spreader	Diesel, low-sulfur ^b market for ^c	/	kg	1.73

^a Northeast/North Central (NE/NC) region of the US (NE-NC/RNA - USLCI database)

^b "Rest-of-the-World" geographical consideration (RoW - EcoInvent 3 database)

^c Allocation default unit process (Alloc Def, U - EcoInvent 3 database)

Background information was collected using SimaPro 8.3 databases (EcoInvent 3 and USLCI), while SimaPro software was used to compute the Life Cycle Inventory (LCI) and Life Cycle Impact Assessment (LCIA) calculation. We used ArcGIS desktop (ESRI, 2011) to create the map and buffer areas, and Google Earth (Google, 2018) to calculate the tortuosity ratio of São Paulo state.

2.3.5.1 Rock extraction

The basalt quarry and production facilities studied here typically have a production capacity of 167 tonnes of aggregate per hour (Rosado *et al.*, 2017). The extraction of the rock starts with drilling to insert explosives (Rosado *et al.*, 2017). The shattered basalt is then loaded in a truck, using a front loader, and transported one km away from the pit, to the processing facility (Rosado *et al.*, 2017).

2.3.5.2 Rock comminution

For this study, we selected a particle size of 5 mm, corresponding to the high end of what is widely referred to as fine aggregate (Mitchell, Mitchell and Pascoe, 2008). In practice,

CHAPTER 2

the <5 mm crushed fraction has a wide range of particle size, thus including fine dust. Its production corresponds, in part, to the process of manufacturing construction aggregates, in which all costs are minimised to provide competitive low-cost products in large quantities. Thus, the process inherently minimises energy consumption, and utilises widely used machinery.

The blasted pieces of rock are ground through successive crushers to reach 5mm. The primary crusher (i.e. jaw crusher) with output particle size of 50 mm is as specified by Rosado *et al.* (2017). The number and model of additional crushers were selected according to their reduction ratios, feed size acceptance, and crushing rates using the guidelines provided by Metso (2015). Two additional crushers are required, a secondary crusher GP100S and a tertiary cone crusher HP200. The different stages of the crushing process are shown in Figure 2.4. All machines are powered by electricity; their energy requirements can be found in Table 2.2.

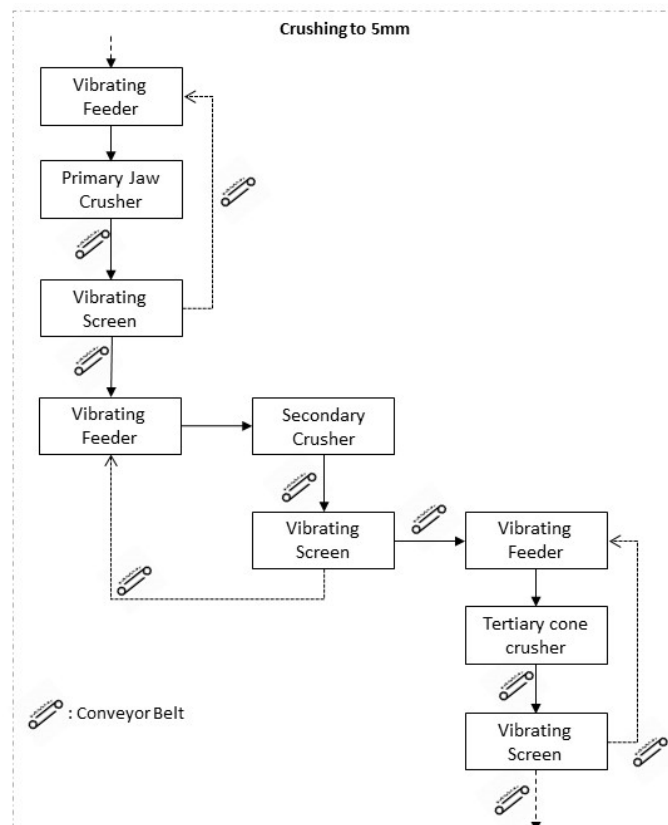


Figure 2.4: Crushing process stages considered to reach 5 mm diameter particles of basalt based on Rosado *et al.* (2017) and Metso (2015).

Table 2.2: Energy requirements of crushing process equipment, as metered electricity.

Item	Number of units	Electricity per unit (kWh / t of rock)	Data Origin
Vibrating feeder	3	0.176	Rosado <i>et al.</i> (2017)
Conveyor Belt	9	0.264	Rosado <i>et al.</i> (2017)
Vibrating Screen	3	0.176	Rosado <i>et al.</i> (2017)
Primary Jaw Crusher	1	0.659	Rosado <i>et al.</i> (2017)
Secondary Crusher GP100S	1	0.539	Metso (2015)
Tertiary Cone Crusher HP200	1	0.820	Metso (2015)
Total	18	5.450	

2.3.5.3 Rock transport

After comminution, the ground basalt is loaded in a truck using a front loader and transported to the field. The rock transport is assumed to be carried out only by truck, as it is the main means of transportation in Brazil (Wanke and Fleury, 2006; Araújo, Campos and Bandeira, 2013).

São Paulo state is described as “irregular plains”, with large homogeneous plain areas and a division between the east central and western part of the state by tableland (Manfré, de Albuquerque Nóbrega and Quintanilha, 2015). The homogeneity of the land and absence of mountains ensure a reasonably constant fuel consumption throughout the state. Additionally, the majority of the roads in SP are considered in “optimal” or “good condition” (CNT Confederacao Nacional do Transporte, 2017), hence road quality is assumed to have no influence on fuel consumption.

The process used in our study considers the transportation of the material from the quarry to heaps at the edge of field and includes the return of the empty truck to the quarry (Weidema *et al.*, 2013).

2.3.5.4 Rock spreading

The delivered basalt is then loaded using a front loader into the field spreader, which was assumed to be a conventional lime spreader.

CHAPTER 2

We calculated the diesel consumption for the spreading of the material according to the agricultural machinery consumption from Goglio *et al.*, (2018). We used the average São Paulo tractor power from Felipe *et al.*, (2008) and working speed, width and application rate from Brazilian agricultural machinery retailers.

2.3.6 Impact assessment

We accounted for acidification and abiotic depletion according to the CML methodology (Guinee *et al.*, 2002), human toxicity, cancer and non-cancer, as well as freshwater ecotoxicity according to the USEtox model (Fantke *et al.*, 2017). Cumulative energy demand (CED) was considered according to the method published by EcoInvent version 2.0 (Frischknecht *et al.*, 2007). GHG balance accounting for the global warming potential (GWP) over a timeframe of 100 years is in accordance with the characterisation factors of the 5th assessment report of the intergovernmental panel on climate change (IPCC, 2014). Units CTUh and CTUe are defined according to Golsteijn (2014). The slope and constant for each linear equation were calculated using Excel and based on the trend line associated with the impact category.

2.3.7 Estimating São Paulo state potential

To approximate the capture potential of SP state, we first calculated the net theoretical sequestration potential of each buffer zone by multiplying the agricultural area of the considered buffer zone by the sequestration potential of EW and carbonation (i.e. 225 kg and 125 kg CO₂e /t of rock applied, respectively). We then subtracted the emissions associated with the setup of the practice for the considered buffer zone. The quarry to field distances used for the calculation of the emissions were the straight-line distances to each buffer zone multiplied by the road tortuosity ratio of 1.3. Finally, we summed the net sequestration potential of each buffer zone inside SP.

2.3.8 Energy for comminution

The energy requirements to reach a 5 mm diameter size particle is based on the machinery recommendation from Metso, (2015), and the available data from Rosado *et al.* (2017). Still according to Metso (2015), the energy needed to specifically grind particles finer than 5 mm is based on Bond's equation:

$$W = 10Wi \left(\frac{1}{\sqrt{P_{80}}} - \frac{1}{\sqrt{F_{80}}} \right) \quad (2-6)$$

With W: Energy [kWh/t]; Wi: Empirical Work Index of the Material [kWh/t]; and P_{80} & F_{80} : Size of particles for which 80% of the product or feed respectively passes through the mesh [μm]

According to this equation, the energy required for grinding increases exponentially as the product maximum particle size gets smaller.

To calculate the energy demand for further grinding from a particle size of 5mm, we used the work index of basalt for grinding of 17.10 (Kanda and Kotake, 2007; Metso, 2015), and added the energy of three conveyor belts, one vibrating feeder, and one vibrating sieve to account for the additional machinery needed.

2.3.9 Uncertainty analysis

Uncertainty analysis was conducted using a numerical approach based on Monte Carlo simulations in R software (Core Team, 2018). For each simulation, 10,000 values were randomly selected from each process's distribution. Computation of the uncertainty related to the sequestration potential of basalt included the observed correlation between CaO and MgO contents when randomly selecting values in the distributions. Additional information about the uncertainty analysis can be found in the Appendix A.

2.4 Results

2.4.1 Life cycle impact assessment

We calculated the values of each impact category using the average quarry to field distance of 65 km accounting for road tortuosity (Table 2.3). Results show that the impacts per hectare increase proportionally with the rate of application while the results per tonne of CO_{2e} removed occur independently of application rate and are inversely proportional to the sequestration potential of the path (EW & carbonation).

Table 2.3: LCIA results for three application rates. Results are shown per hectare and per tonne of CO₂e removed according to the two paths (EW & Carbonation).

Impact Category	Unit	Application Rate [t/ha]	Total [per hectare]	Total Carbonation [per t CO ₂ removed]	Total EW [per t CO ₂ removed]
GHG balance	kg CO ₂ e	5	84.1	135	75.1
		20	337		
		50	842		
CED	MJ	5	1647	2635	1471
		20	6588		
		50	16470		
Abiotic Depletion	kg Sb eq	5	1.74E-04	2.79E-04	1.56E-04
		20	6.97E-04		
		50	1.74E-03		
Acidification	kg SO ₂ eq	5	0.618	0.990	0.552
		20	2.47		
		50	6.18		
Freshwater ecotoxicity	CTUe	5	323	517	289
		20	1293		
		50	3233		
Human Toxicity, Cancer	CTUh	5	2.23E-06	3.57E-06	1.99E-06
		20	8.92E-06		
		50	2.23E-05		
Human Toxicity, Non Cancer	CTUh	5	1.81E-05	2.89E-05	1.61E-05
		20	7.23E-05		
		50	1.81E-04		

CTUh: “estimated increase in morbidity (the number of disease cases) in the total human population per unit of mass of the chemicals emitted.” (Laura Golsteijn, 2014)

CTUe: “estimated potentially affected fraction of species integrated over time and the volume of the freshwater compartment, per unit of mass of the chemicals emitted.” (Laura Golsteijn, 2014)

Our contribution analysis (Figure 2.5) shows the share of each process stage for the impact categories assessed using the average quarry to field distance (i.e. 65 km). The

results show that transportation accounts for the majority of the impact categories, ranging from 35% for acidification to 88% for abiotic depletion.

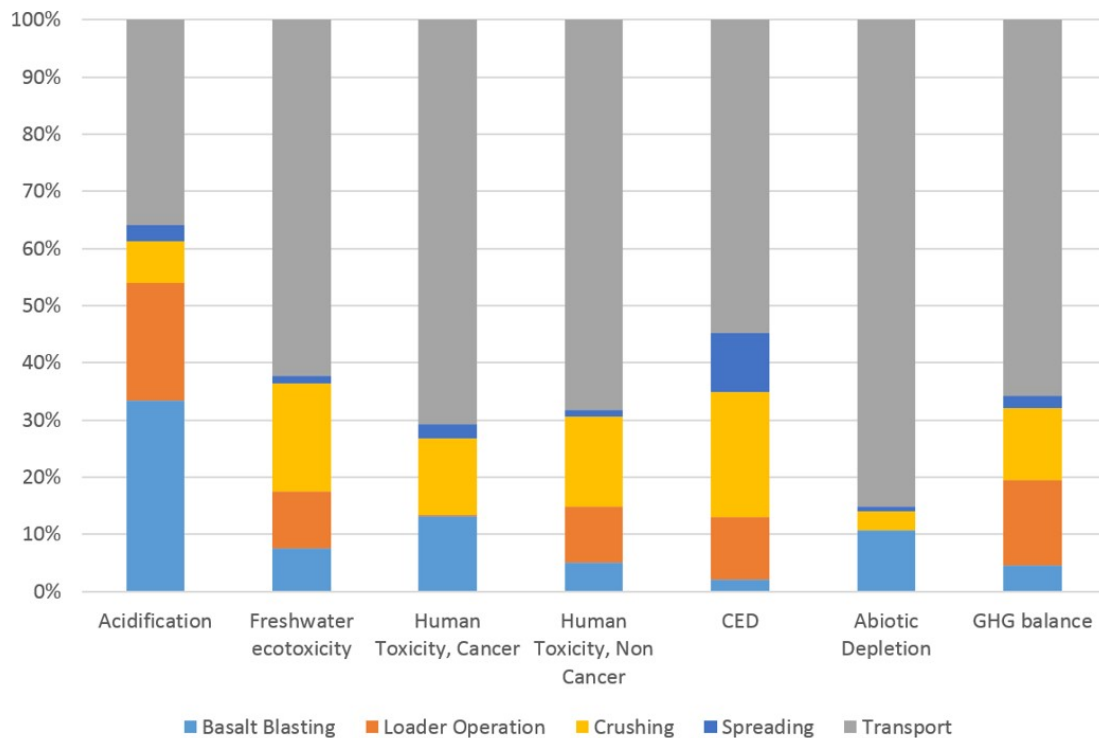


Figure 2.5: Impact categories contribution analysis considering the quarry to field distance of 65km.

Figure 2.6 represents the effect of an increasing quarry to field distance on the GHG balance impact category with a fixed application rate of 5 t/ha. Results show that transportation impact increases as distance increases while crushing, loading, spreading, and blasting process stages remain unchanged, and thus can be considered as “*spatially static*” (Moosdorf, Renforth and Hartmann, 2014).

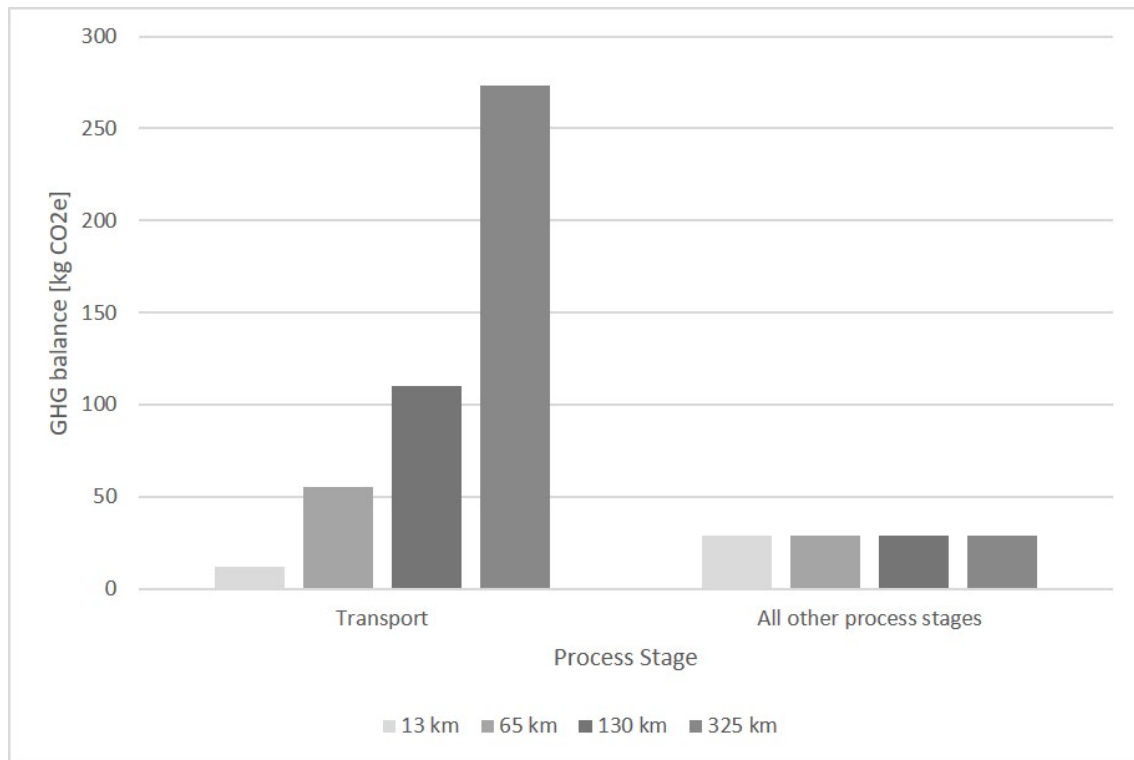


Figure 2.6: Effect of an increasing quarry to field distance on the process stages considering GHG balance [kg CO₂e] impact category and an application rate of 5 t/ha.

Figure 2.7 illustrates the increase of the GHG emissions associated with the transportation stage as the quarry to field distance increases (grey line). The blue line represents the same trend with the added GHG balance impacts of the spatially static process stages and the orange line, the proportion of transportation within the value of total GHG balance. Figure 2.7 shows that the trend is linear and thus that the impact value increases proportionally with the quarry to field distance.

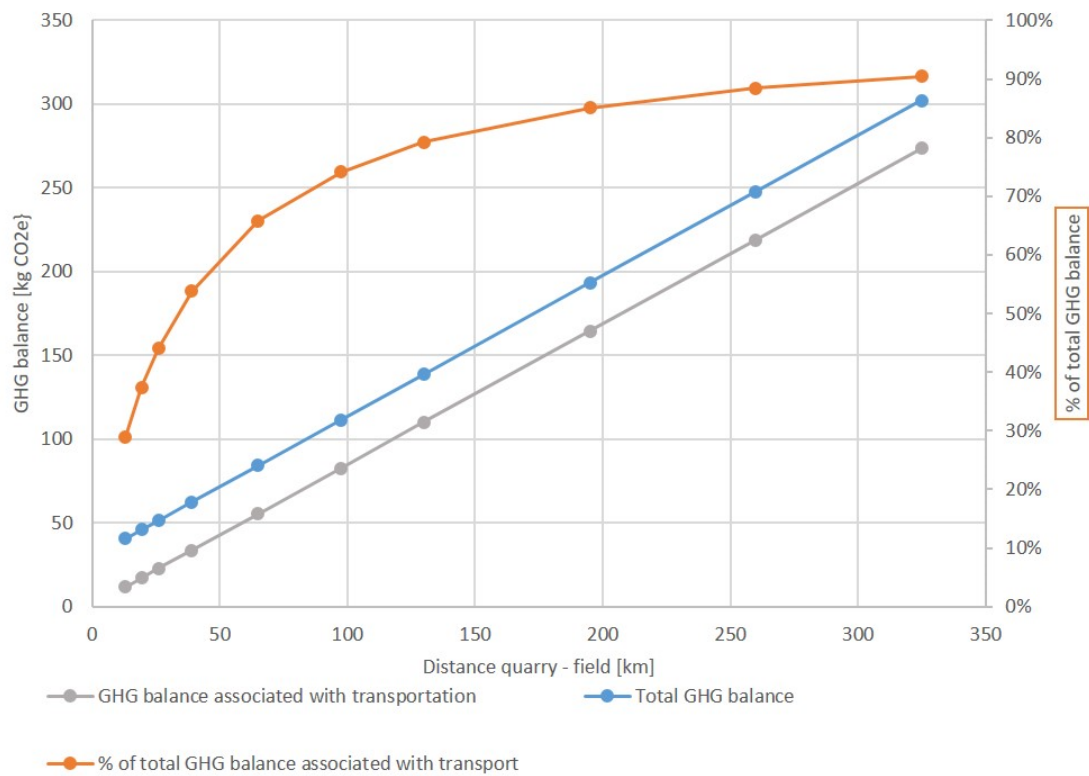


Figure 2.7: Share increase of transport linked with increasing quarry to field distance [km] considering total GHG balance impact category and an application rate of 5t/ha.

These results show that not only is truck transportation the most impacting process of the practice (when considering the average quarry to field distance) (Figure 2.5), but also that the values for each impact category can be readily derived from linear equations because of the proportional trend between quarry to field distance and transportation impact (Figure 2.7).

Table 2.4 gives the slope and constant values for the impact category assessed. Values are shown for the baseline application rate of 1 t/ha.

Table 2.4: Slope and constant of each impact category for linear prediction.

Impact Category	Unit	Application Rate [t/ha]	Slope	Constant
GHG balance	kg CO ₂ e	1	0.218	5.92
CED	MJ	1	3.56	151
Freshwater ecotoxicity	CTUe	1	0.610	25.0

Impact Category	Unit	Application Rate [t/ha]	Slope	Constant
Abiotic Depletion	kg Sb eq	1	5.85E-07	5.60E-06
Acidification	kg SO ₂ eq	1	8.74E-04	8.00E-02
Human Toxicity, Cancer	CTUh	1	6.22E-09	1.35E-07
Human Toxicity, Non-Cancer	CTUh	1	4.86E-08	1.19E-06

The linear equation $y = mx + b$ with y: the value of the impact category; x: the quarry to field distance in km; m: the slope; and b: the constant.
The constant and slope should be multiplied beforehand by the application rate considered

2.4.2 CO₂e capture potential of São Paulo State

Table 2.5 shows how the net sequestration potential of São Paulo state was calculated for an application rate of 5 t/ha. The column “distance quarry field” represents the concentric buffer zone distances of Figure 2.2 updated with the tortuosity ratio of 1.3 discussed in 2.3.4. The “agricultural area covered” column shows the number of hectares included in each of the buffer zone, considering all quarries in SP. The emissions associated with the setup of the practice at the different distance range are reported in the “total GHG balance” column and the values consider the application rate of 5 t/ha of ground basalt on all the hectares within the buffer zone considered. The “net sequestration” columns result from the subtraction of the emission associated with the practice from the potential sequestration (125 and 225 kg CO₂e per tonne of rock applied for carbonation and EW respectively), within a buffer zone, and according to the number of hectares associated. The “loss ratio” represents the part of the sequestration potential that is lost throughout the setup of the practice. The loss ratio increases greatly with the quarry to field distance due to the impact of truck transportation.

Table 2.5: Emissions and net sequestration in kt CO₂e for the implementation of the practice considering the area included under each buffer zones (with application rate of 5t/ha)

Distance Quarry - Field [km]	Agricultural Area covered [Million ha]	Application Rate: 5 t/ha			
		GHG balance ^a	Net sequestration (Carbonation) _a	Net sequestration (EW) ^a	Loss Ratio (Carbona tion)
13	0.781	31.6	456	843	6.5%
19.5	0.835	38.4	483	897	7.4%
26	0.999	51.3	573	1,067	8.2%
39	2.17	135	1,223	2,298	10.0%
65	3.57	300	1,931	3,698	13.5%
97.5	2.19	244	1,123	2,206	17.8%
130	0.814	113	396	798	22.2%
195	0.222	42.8	95.8	206	30.9%
260	0.102	25.2	38.3	88.7	39.6%
325	0.000376	0.114	0.121	0.307	48.4%
TOTAL	11.7		6.32	12.10	

^a Emissions and sequestration values for the area covered under the considered buffer zone. In kt CO₂e

Table 2.6 shows the potential net sequestration of the state for the different rates of application considered (5, 20, 50 t/ha), and compares it with the 2016 emissions of SP state (SEEG, 2016a), Brazil (SEEG, 2016b), and the world (Olivier, Schure and Peters, 2017).

Table 2.6: São Paulo (SP) CO₂e sequestration potential through carbonation and EW as compared with 2016 SP, Brazilian, and global CO₂e emissions. Expressed in Mt CO₂e

Application Rate	SP net sequestration Potential [Mt CO ₂ e]	2016			
		SP Emission	Brazil Emission	World Emission	
		159 [Mt CO ₂ e]	2278 [Mt CO ₂ e]	46400 [Mt CO ₂ e]	
5 t/ha	Carbonation	6.32	3.98%	0.28%	0.01%
	EW	12.1	7.62%	0.53%	0.03%
20 t/ha	Carbonation	25.3	15.91%	1.11%	0.05%

Application Rate	SP net sequestration Potential [Mt CO ₂ e]		SP Emission 2016	Brazil Emission 2016	World Emission 2016
			159 [Mt CO ₂ e]	2278 [Mt CO ₂ e]	46400 [Mt CO ₂ e]
50 t/ha	EW	48.4	30.48%	2.13%	0.10%
	Carbonation	63.2	39.79%	2.77%	0.14%
	EW	121	76.19%	5.31%	0.26%

Mt = 10⁹ kg

2.5 Discussion

2.5.1 GHG removal potential

Several publications have attempted to estimate the sequestration potential of EW (EASAC, 2018). Although some relied on theoretical calculation to assess the energy cost to implement the practice, most provide overestimated sequestration values.

For instance, Kantola *et al.* (2017) suggest that 1.1 Pg CO₂e could be sequestered by the application of 50t/ha of basalt on the 70 x 10⁶ ha of the corn belt of North America. However, let us hypothesize that i) the North American production system of 5 mm basalt particles is equivalent to the one represented in this study; ii) that 50% of the material takes the EW path and the other 50% the carbonation path; iii) that all the considered croplands are situated at an average road distance of 65 km from the closest basalt mine (it may be much more than this in agricultural parts of North America); iv) and that the topography and road quality of the area does not affect the emissions of the truck transport. Then, when accounting for the burdens of our optimistic hypothetical system, only 50% (0.55 Pg CO₂e) of the potential postulated by Kantola *et al.* (2017) can be reached.

The difference between Kantola *et al.* (2017)'s prediction and ours results from the voluntary omission of the emissions associated with the setup of the practice by Kantola *et al.* (16.8 kg CO₂e/t of rock in our hypothetical system) and an estimated sequestration potential of basalt of 0.32 t CO₂e / tonne of rock as compared to our average 0.175 t CO₂e / tonne of rock ((225 [EW]+125 [carbonation])/2).

As distance from quarry to field increases, further reduction in the average sequestration potential of basalt is observed and declines to 147 and 125 kg CO₂e / tonne of rock when the quarry to field distance reaches 130 and 260 km of flat, good quality road, respectively.

2.5.2 Co-benefits/drawbacks to basalt dust addition

In addition to its carbon capture capacity, studies suggest that the weathering of basalt rock on soils improves crop growth and soil fertility (reviewed by Beerling *et al.*, 2018). According to these studies, the addition of basalt rock dust to soil provides a slow release of nutrients, increases yield and CEC, rebalances soil pH, increases plant resistance to insects, disease, frost and drought, and increases microorganism growth and earthworm activity, among other things (Mersi *et al.*, 1991; Leonardos *et al.*, 1998; Harley and Gilkes, 2000; Gillman *et al.*, 2002; Theodoro and Leonardos, 2006; Massey *et al.*, 2007; Manning, 2010; Melo *et al.*, 2012; Bergmann *et al.*, 2013; Nunes *et al.*, 2014; Campe *et al.*, 2015; Ramos *et al.*, 2015, 2017; Kantola *et al.*, 2017).

From an LCA perspective, the resulting increased biomass, reduced use of fertilizers and pesticides, and the potential removal of the need for lime application (Kantola *et al.*, 2017) (and decrease of linked soil CO₂ emissions (Beerling *et al.*, 2018)) would all contribute to the reduction of the carbon footprint per unit crop output and per ha associated with basalt addition.

The impacts of these benefits are soil, climate and plant specific hence they are impossible to determine in a coarse scale assessment. However, it is reasonable to expect further reductions in the carbon footprint of the overall practice.

Negative impacts related to the addition of basalt, such as possible increased turbidity in drainage to nearby streams or rivers due to unweathered particles (Kohler *et al.*, 2010; Edwards *et al.*, 2017) may be expected as well; the use of a <5mm fraction rather than a finely milled (e.g. <100µm) fraction may mitigate this. The net effects on other categories such as eco or human toxicity impact categories are unclear, but they do not affect net C sequestration.

2.5.3 Particle size and weathering rate

Several studies promote the use of fine dust particles (10 to 30 micron in diameter) for their rapid carbon sequestration potentials (Kohler *et al.*, 2010; Moosdorf *et al.*, 2014; Beerling *et al.*, 2018). However, although the particulate emissions (PM₁₀) impact category was not assessed in our LCA due to lack of data on the spreading process stage, transport and application of dust products may have adverse health effects for practitioners (Schenker *et al.*, 2009; Taylor *et al.*, 2016). In addition, fine grinding or milling demands major energy input (Orumwense and Forssberg, 1992) that could offset the potential sequestration of EW or carbonation. The case for further milling of the <5mm fraction typically produced in construction aggregate crushing operations would need to be supported by evidence of the associated costs and benefits.

According to the “catchment scale-based conversion” used in Streffler *et al.* (2018) to estimate the share of spread material that weathered within one year (depending on grain size), the application of 5mm basalt particles on soil at pH 7 and 25°C would result in the dissolution of 0.021% of the material after one year. However, this calculation is theoretical and idealized for it ignores the shrinking of the particles as they weather, and the particle size distribution (the <5mm fraction is a size range). Moreover, none of the factors such as the climatic condition, soil type, presence of vegetation, microorganisms, and fungi, and concentration of atmospheric CO₂ (all known to affect the weathering rate of silicate rocks) are taken into account (Harley and Gilkes, 2000; Beaulieu *et al.*, 2010; Manning and Renforth, 2013; Moosdorf, Renforth and Hartmann, 2014; Opolot and Finke, 2015; Li *et al.*, 2016; Kantola *et al.*, 2017; Manning, 2018; Streffler *et al.*, 2018).

In addition, as the grain size of a final construction aggregate product gets smaller, the co-production of quarry fines (material <5mm) increases. Although their production depends on various factors (e.g. mineral composition and texture of the rock, crusher type and number of stages) the production of 10 mm aggregate usually result in 35–40 % of fines <5mm by weight (Mitchell, Mitchell and Pascoe, 2008). A product with low amounts of use in the aggregate industry, these fines could be spread on the field for EW or carbonation where they will rapidly weather (Manning *et al.*, 2013).

Precisely predicting the weathering rates without measurements from local field experiments is, as yet, impossible because of the manifold parameters involved. However,

the tropical climate, acidic soils, and the presence of vegetation on the application area considered in our study as well as the substantial proportion of material <5mm should ensure a faster weathering rate than the 0.021% per year calculated here above.

Weathering rate, along with the vertical transportation of the applied material through soil by percolation and bioturbation (disturbance of soil by living organisms) (Taylor *et al.*, 2016), are crucial for they determine the delay between applications. This delay is essential to maintain the balance of the amended site and ensure durability of the potential benefits associated with the technique. As neither of those parameters were precisely considered in this study, the sequestered amount presented should not be considered as annual.

2.5.4 Maximum travel distance to offset potential capture

Basalt extraction, crushing and spreading induce fixed emissions (discussed in Section 2.4.1). However, as the quarry to field distance increases, the emissions associated with transport further reduce the sequestration potential of the practice. Based on the linear relationship between delivery distance and transport GHG emissions, the distance at which the carbon capture potential of basalt application is completely offset is about 540 and 990 km of road travel for carbonation and EW respectively. This distance is independent of the amount of basalt processed, as the emissions associated with the processes increase proportionally with the potential sequestration through carbonation and EW. These offsetting distances are proportionally related to the potential sequestration of each path due to the linear increase of GHG emissions and distance as discussed before.

In a global assessment, Moosdorf *et al.* (2014) suggest that “*The transportation CO₂ emissions from source to application areas average 0.007 (0.022) t CO₂ t⁻¹ [optimistic (pessimistic) scenarios; t⁻¹: per tonne of rock]*” considering transportation by railroad, road and waterways. From our hypothetical system and the average quarry to field distance of 65km, we reach an average emission of 0.011 t CO_{2e} per tonne of rock, which is within the range of Moosdorf *et al.* (2014). Yet, our hypothetical system is located on part of a large flood basalt outcrop where spatial density of basalt quarries can be assumed higher than in other parts of the world. The underestimation of transportation impact is seen further where Moosdorf *et al.* (2014) state that the maximum distance (at which the

CHAPTER 2

emission offset the capture potential) is 17,000 (5000) km by road when considering a potential sequestration of 1.1 (0.8) tCO₂e / t of rock (optimistic (pessimistic) scenario). Compared with our hypothetical system, Moosdorf *et al.* (2014)'s optimistic and pessimistic predictions are overestimated by 72 and 30%, respectively. This difference is due to the fact that Moosdorf *et al.* (2014) considered global routes from source rock location to each continuous area of arable land.

Considering other means of transportation, such a railroad or inland waterways, the offsetting distance is increased. According to SimaPro 8.3.0, the waterway and railroad processes emit around 3.5 times less CO₂/tkm than the truck considered for road transport.

Assuming that 50 km of road transportation is necessary from the quarry to the station/harbour and another 50 km is necessary to reach the field after railroad/waterway transportation, the offsetting distance considering carbonation would reach around 2200 and 2140 km through railroad and waterway, respectively. Regarding EW, these distances increase to around 4300 and 4200 km through railroad and waterway, respectively.

2.5.4.1 Impact of uncertainties on offsetting distance

Our uncertainty analysis (in the Supplementary Information, Appendix A) showed that the most uncertain parameters in our assessment are i) the fraction of the cations that will be transferred to the ocean (ω value); and ii) the type of truck used to set up the practice (emission factor). In Figure 2.8, we show the impacts of both these parameters on the offsetting distance of the practice. The error bars on the figure represent the propagation of the uncertainty associated with all the other processes. Based on our uncertainty analysis, the error in the ω value represents around 11% of the mean sequestration potential and errors in the different process stages represent around 5% of the emissions.

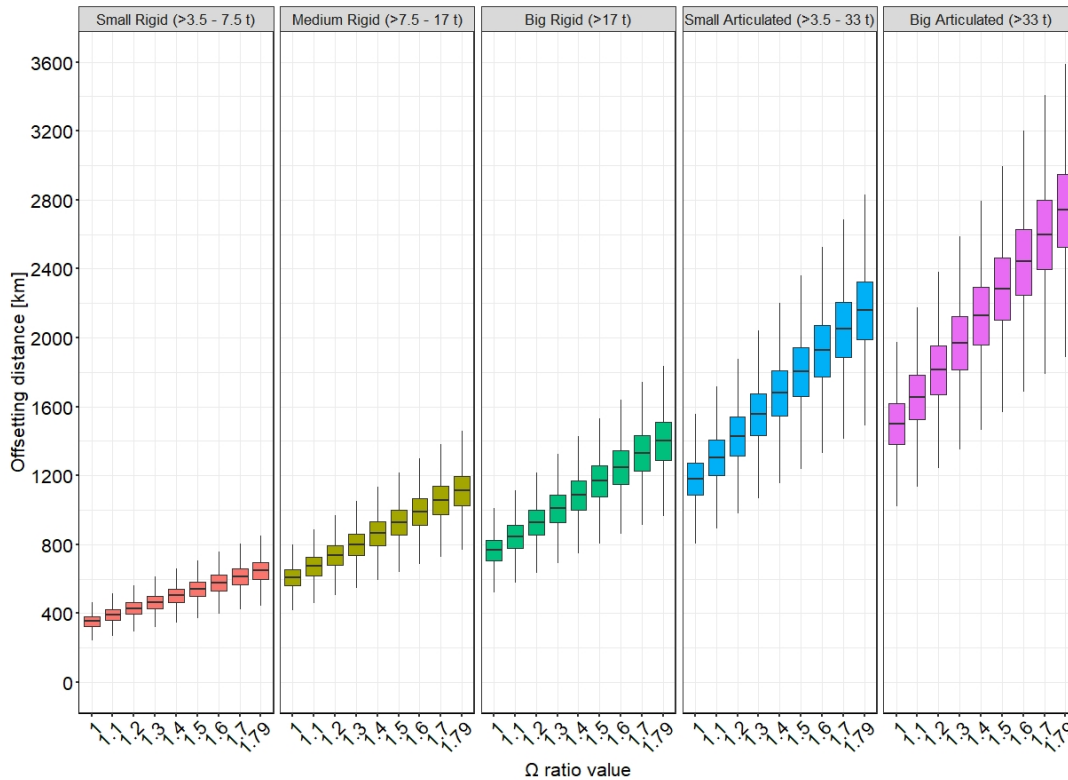


Figure 2.8: Effect of Emission Factor (truck type) and W value on the offsetting distance. The W value of 1 and 1.79 represent Carbonation and EW, respectively. The error bars represent the propagation of the uncertainty on emissions associated with all the other processes as well as the uncertainty on sequestration potential according to the W value.

Figure 2.8 highlights the influence of the emission factor (in kg CO₂e/tkm) of the truck selected for the transportation of the material (threefold difference between most and least efficient trucks) as well as the impact of the additional sequestration due to the transfer of the cations to the ocean (ω value) on the offsetting distance. Hauliers would usually use the largest vehicle possible, given the road conditions.

2.5.5 Comminution and energy requirement

Based on the conversion factor of SimaPro 8.3 from Brazilian electricity to kgCO₂e we calculated the energy required to offset the sequestration values, considering the average quarry to field distance of 65 km. The energy needed to offset the sequestration potential of the practices is around 276 and 529 kWh for carbonation and EW, respectively.

Therefore, considering Bond's equation, the energy required to reach 5 mm particles, the additional equipment needed to consider the grinding facility, and the emissions

CHAPTER 2

associated with the other process stages (average quarry to field distance of 65 km), the particle size to reach this offsetting energy demand would be 0.00062 mm and 0.00032 mm for carbonation and EW, respectively. Although further comminution induces a steep increase of energy, the particle size to offset the sequestration potential is, in practice, unreachable.

The distance at which the setup of the practice offsets the sequestration potential is based on the emissions associated with the implementation of the practice. As the particle size decreases, the emissions increase and so reduce the offsetting distance of the practice. When the final particle size is 5mm, the offsetting distances are 544 and 994 km by road for carbonation and EW, respectively. When the final particle size reaches 0.02 mm the offsetting distances decline to 526 and 977 km for carbonation and EW, respectively. The distance decreases exponentially with lower particle size due to the exponential increase in energy requirement as the product particle size decreases.

2.5.6 Basalt production in SP and relevance of the proposed application rate

According to the data extracted from UCLouvain, (2017), São Paulo state has a total of 11.7 million hectares of cropland and grassland. Therefore, 58.5, 234, and 585 million tonnes of basalt would be needed to amend the land considering our three application rates of 5, 20, and 50 tonnes per hectare, respectively.

The representative quarry used in this study has an annual production of 450,000 tonnes (Rosado *et al.*, 2017) and is considered as a medium size facility (DNPM, 2018). To reach the application rates of 5, 20, and 50 t/ha proposed annually, São Paulo state would need 130, 520, 1300 medium size facilities producing 450,000 t/year, respectively. However, according to DNPM, (2018), São Paulo state had a total of 47 basalt quarries in 2015: 1 big facility (> 1,000,000 t/year), 32 medium size ([100,000 – 1,000,000] t/year), 13 small ([10,000 – 100,000] t/year) and 1 micro facility (<10,000 t/year).

If we consider an average production of 450,000 t/year for each facility, and the allocation of 100% of the production to the practice, the conceivable application rate reaches 1.8 t/ha. Despite being strongly approximated, these numbers shed light on the mismatch of the application rate sometimes considered in publications. As shown above, transport distances play a key role in the sequestration potential of the practice. Considering an

application rate of 50 t/ha while staying in the geographical range of already existing basalt quarries overshoots their production capacity, which would need to increase by a factor of 3 (5 t/ha) -10 (20 t/ha) to have a reasonable impact. This could be achieved either by expanding existing quarries or by creating new quarries.

2.6 Conclusion

In this study, we have shown that an existing network of basalt quarries, already producing construction aggregates at low cost, has the potential to supply a region with crushed material suitable for enhanced weathering or carbonation. Our LCA distinguished fixed and transport-related burdens of extraction, delivery and application, and enabled us to assess the net CO₂ sequestration benefit from using ground basalt on agricultural soil.

We showed that transportation of the ground material greatly affects the potential sequestration of the technique. In our case study, the maximum delivery distances before basalt management emissions exceeded CO₂ sequestration were 544 ± 65 and 994 ± 116 km when considering carbonation and EW, respectively. Comparisons with previously published assessment show logistics and transport impacts can be underestimated, leading to overoptimistic sequestration values.

Considering São Paulo state sequestration potential, our study shows that the application of crushed basalt could capture 1.3 to 2.4 Mt CO_{2e} through the application rate of 1t/ha applied on all its 12 million hectares through carbonation and enhanced weathering respectively, including the emissions associated with the setup of the practice.

We considered the likely positive soil and crop response to basalt addition that could increase the sequestration potential of the practice as well as potential negative effects that warrant further experimental investigation.

Finally, as various techniques are increasingly promoted as efficient solutions to remove or sequester CO₂, this study emphasizes the need to consider the practice in its entirety to show the limitations and potential caveats. The holistic approach of LCA helps in identifying the processes with the largest impacts and their significance. However, its precision and reliability are only as high as the amount, quality and representability of the

input data. Therefore, fundamental studies on each process stage considered should be conducted, focusing on local practices to increase accuracy.

2.7 Acknowledgments

We acknowledge funding through the UP-Green-LCA (NE/P019668/1) and SOILS-R-GGREAT (NE/P019498/1) projects of the greenhouse gas removal (GGR) programme. The GGR programme is financed by the UK Natural Environment Research Council (NERC), Engineering and Physical Science Research Council (EPSRC), Economic and Social Science Research Council (ESRC) and the UK department for Business, Energy and Industrial Strategy (BEIS). The authors wish to acknowledge the Royal Society for providing precious insights at the Sackler Forum in 2017.

2.8 References

- Araújo, M. da P. S., Campos, V. B. G. and Bandeira, R. A. M. (2013) *An overview of road cargo transport in Brazil, International Journal of Industrial Engineering and Management*. Available at: http://ijiemjournal.org/images/journal/volume4/ijiem_vol4_no3_7.pdf.
- Baggio, S. B., Hartmann, L. A. and Bello, R. M. S. (2016) 'Paralavas in the cretaceous paraná volcanic province, Brazil - a genetic interpretation of the volcanic rocks containing phenocrysts and glass', *Anais da Academia Brasileira de Ciencias*, 88(4), pp. 2167–2193. doi: 10.1590/0001-3765201620150088.
- Beaulieu, E. *et al.* (2010) 'Impact of atmospheric CO₂ levels on continental silicate weathering', *Geochemistry, Geophysics, Geosystems*, 11(7). doi: 10.1029/2010GC003078.
- Beerling, D. J. (2017) 'Enhanced rock weathering: Biological climate change mitigation with co-benefits for food security?', *Biology Letters*, 13(4), pp. 4–7. doi: 10.1098/rsbl.2017.0149.
- Beerling, D. J. *et al.* (2018) 'Farming with crops and rocks to address global climate, food and soil security', *Nature Plants*, 4(3), pp. 138–147. doi: 10.1038/s41477-018-0108-y.
- ten Berge, H. F. M. *et al.* (2012) 'Olivine weathering in soil, and its effects on growth and

nutrient uptake in ryegrass (*Lolium perenne* L.): A pot experiment', *PLoS ONE*, 7(8). doi: 10.1371/journal.pone.0042098.

Bergmann, M. *et al.* (2013) *Amygdaloidal Basalts to Zeolites of the "Serra Geral" Formation of Paraná Basin : Potential Agronomic Use, Brazilian Stonemeal Conference*. Available at: <https://remineralize.org>.

Berner, R. A. and Kothavala, Z. (2001) 'GEOCARB III; A Revised Model of Atmospheric CO₂ Over Phanerozoic Time', *American Journal of Science*, 301(1), pp. 182–204. doi: 10.2475/ajs.294.1.56.

Braz Machado, F. *et al.* (2018) 'Geochemistry of the Northern Paraná Continental Flood Basalt (PCFB) Province: implications for regional chemostratigraphy', *Brazilian Journal of Geology*, 48(2), pp. 177–199. doi: 10.1371/journal.pone.0018728.

Bryan, S. E. and Ernst, R. E. (2008) 'Revised definition of Large Igneous Provinces (LIPs)', *Earth-Science Reviews*, 86(1–4), pp. 175–202. doi: 10.1016/j.earscirev.2007.08.008.

Campe, J., Kittredge, D. and Klinger, L. (2015) *The Potential of Remineralization With Rock Mineral Fines To Transform Agriculture, Forests, Sustainable Biofuels Production, Sequester Carbon, and Stabilize the Climate*. Available at: <http://remineralize.org/wp-content/uploads/2015/10/ODB1.pdf>.

CNT Confederacao National do Transporte (2017) *Pesquisa de Rodovias Sudeste [MAP]*. Available at: <http://pesquisarodovias.cnt.org.br/Paginas/mapa-por-regiao-uf>.

Core Team, R. (2018) *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. Vienna. Available at: <https://www.r-project.org/>.

DNPM (2018) *Cadastro Nacional de Produtores de Brita - Ano Base 2015*. Available at: <http://www.dnpm.gov.br>.

EASAC (2018) *Negative emission technologies: What role in meeting Paris Agreement targets?* Halle - Germany. Available at: <https://easac.eu>.

Edwards, D. P. *et al.* (2017) 'Climate change mitigation : potential benefits and pitfalls of enhanced rock weathering in tropical agriculture', *Biology Letters*, (13). doi:

CHAPTER 2

10.1098/rsbl.2016.0715.

ESRI (2011) *ArcGIS Desktop*. Redlands, CA: Environmental System Research Institute. Available at: <http://desktop.arcgis.com>.

Fantke, P. (Ed.). *et al.* (2017) *USEtox® 2.0, Documentation (version 1)*. Edited by P. Fantke. doi: 10.11581/DTU:00000011.

Felipe, F. I., Lima, R. A. D. S. and Rodrigues, S. M. (2008) *Evolução da estrutura da indústria de tratores de rodas, no Brasil, no período de 1999 a 2008, 47 Congresso Sociedade Brasileira de Economia, Administração e Sociologia Rural*. Available at: <https://www.cepea.esalq.usp.br>.

Frischknecht, R. *et al.* (2007) *Implementation of Life Cycle Impact Assessment Methods, American Midland Naturalist*. Available at: http://www.ecoinvent.org/fileadmin/documents/en/03_LCIA-Implementation.pdf.

Fuss, S. *et al.* (2014) ‘Betting on negative emissions’, *Nature Climate Change*. Nature Publishing Group, 4(10), pp. 850–853. doi: 10.1038/nclimate2392.

Fuss, S. *et al.* (2018) ‘Negative emissions — Part 2: Costs , potentials and side effects’, *Environmental Research Letters*, 13 063002(063002). doi: 10.1088/1748-9326/aabf9f.

Gillman, G. P., Burkett, D. C. and Coventry, R. J. (2002) ‘Amending highly weathered soils with finely ground basalt rock’, *Applied Geochemistry*, 17(8), pp. 987–1001. doi: 10.1016/S0883-2927(02)00078-1.

Goglio, P. *et al.* (2018) ‘Development of Crop.LCA, an adaptable screening life cycle assessment tool for agricultural systems: A Canadian scenario assessment’, *Journal of Cleaner Production*. Elsevier Ltd, 172, pp. 3770–3780. doi: 10.1016/j.jclepro.2017.06.175.

Goldich, S. S. (1938) ‘A study in rock-weathering’, *The Journal of Geology*, 46(1), pp. 17–58. doi: 10.1086/624619.

Google (2018) *Google Earth Pro*. Available at: <https://www.google.com/earth/download/gep/agree.html>.

Guinee, J.B. *et al.* (2002) *Handbook on Life Cycle Assessment, Operational guide to the*

- ISO standards*. Edited by Jeroen B. Guinee *et al.* Dordrecht: Springer Netherlands (Eco-Efficiency in Industry and Science). doi: 10.1007/0-306-48055-7.
- Harley, A. D. and Gilkes, R. J. (2000) 'Factors influencing the release of plant nutrient elements from silicate rock powders: A geochemical overview', *Nutrient Cycling in Agroecosystems*, 56(1), pp. 11–36. doi: 10.1023/A:1009859309453.
- Hartmann, J. *et al.* (2009) 'Global CO₂-consumption by chemical weathering: What is the contribution of highly active weathering regions?', *Global and Planetary Change*. Elsevier B.V., 69(4), pp. 185–194. doi: 10.1016/j.gloplacha.2009.07.007.
- Hartmann, J. and Moosdorf, N. (2012) 'The new global lithological map database GLiM: A representation of rock properties at the Earth surface', *Geochemistry, Geophysics, Geosystems*, 13(12), pp. 1–37. doi: 10.1029/2012GC004370.
- Harvey, L. D. D. (2008) 'Mitigating the atmospheric CO₂ increase and ocean acidification by adding limestone powder to upwelling regions', *Journal of Geophysical Research: Oceans*, 113(4), pp. 1–21. doi: 10.1029/2007JC004373.
- IBGE (2017) *Resultados do Censo Agro 2017, Censo Agro 2017*. Available at: https://censoagro2017.ibge.gov.br/templates/censo_agro/resultadosagro/estabelecimentos.html?localidade=35 (Accessed: 11 September 2018).
- IPCC (2014) *Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Edited by T. F. Stocker *et al.* Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press. Available at: <https://www.ipcc.ch/report/ar5/wg1/>.
- Kanda, Y. and Kotake, N. (2007) 'Comminution Energy and Evaluation in Fine Grinding', in *Handbook of Powder Technology*, pp. 529–550. doi: 10.1016/S0167-3785(07)12015-7.
- Kantola, I. B. *et al.* (2017) 'Potential of global croplands and bioenergy crops for climate change mitigation through deployment for enhanced weathering', *Biology Letters*, (13). doi: <http://dx.doi.org/10.1098/rsbl.2016.0714>.
- Kheshgi, H. S. (1995) 'Sequestering Atmospheric Carbon Dioxide by Increasing Ocean

CHAPTER 2

Alkalinity', *Energy*, 20(9), pp. 915–922. doi: 0360-5442(95)00035-6.

Khoo, H. H. *et al.* (2011) 'Carbon capture and utilization: Preliminary life cycle CO₂, energy, and cost results of potential mineral carbonation', *Energy Procedia*, 4, pp. 2494–2501. doi: 10.1016/j.egypro.2011.02.145.

Kohler, P., Hartmann, J. and Wolf-Gladrow, D. A. (2010) 'Geoengineering potential of artificially enhanced silicate weathering of olivine', *Proceedings of the National Academy of Sciences*, 107(47), pp. 20228–20233. doi: 10.1073/pnas.1000545107.

Kolosz, B. W., Sohi, S. p. and Manning, D. A. C. (2019) 'CASPER: A modelling framework to link mineral carbonation with the turnover of organic matter in soil', *Computers and Geosciences*. Elsevier Ltd, 124(January), pp. 58–71. doi: 10.1016/j.cageo.2018.12.012.

Landi, A., Mermut, A. R. and Anderson, D. W. (2003) 'Origin and rate of pedogenic carbonate accumulation in Saskatchewan soils, Canada', *Geoderma*, 117(1–2), pp. 143–156. doi: 10.1016/S0016-7061(03)00161-7.

Laura Golsteijn (2014) *How to Use USEtox® Characterisation Factors in SimaPro | PRé Sustainability*. Available at: <https://www.pre-sustainability.com/news/how-to-use-usetox-characterisation-factors-in-simapro> (Accessed: 16 October 2018).

Leonardos, O. H., Theodoro, S. H. and Assad, M. L. (1998) 'Remineralization for sustainable agriculture: A tropical perspective from a Brazilian viewpoint', *Nutrient Cycling in Agroecosystems*, 56(1), pp. 3–9. doi: 10.1023/A:1009855409700.

Li, G. G. *et al.* (2016) 'Temperature dependence of basalt weathering', *Earth and Planetary Science Letters*. Elsevier B.V., 443(June), pp. 59–69. doi: 10.1016/j.epsl.2016.03.015.

Manfré, L. A., de Albuquerque Nóbrega, R. A. and Quintanilha, J. A. (2015) 'Regional and local topography subdivision and landform mapping using SRTM-derived data: a case study in southeastern Brazil', *Environmental Earth Sciences*, 73(10), pp. 6457–6475. doi: 10.1007/s12665-014-3869-2.

Manning, D. A. C. (2010) 'Mineral sources of potassium for plant nutrition . A review',

Agronomy for Sustainable Development, 30, pp. 281–294. doi: 10.1051/agro/2009023>.

Manning, D. A. C. *et al.* (2013) ‘Carbonate precipitation in artificial soils produced from basaltic quarry fines and composts: An opportunity for passive carbon sequestration’, *International Journal of Greenhouse Gas Control*. Elsevier Ltd, 17, pp. 309–317. doi: 10.1016/j.ijggc.2013.05.012.

Manning, D. A. C. (2018) ‘Innovation in Resourcing Geological Materials as Crop Nutrients’, *Natural Resources Research*. Springer US, 27(2), pp. 217–227. doi: 10.1007/s11053-017-9347-2.

Manning, D. A. C. and Renforth, P. (2013) ‘Passive sequestration of atmospheric CO₂ through coupled plant-mineral reactions in urban soils’, *Environmental Science and Technology*, 47(1), pp. 135–141. doi: 10.1021/es301250j.

Manning, D. A. C. and Theodoro, S. H. (2018) ‘Enabling food security through use of local rocks and minerals’, *The Extractive Industries and Society*. Elsevier, 7(2), pp. 480–487. doi: 10.1016/j.exis.2018.11.002.

Martin, J. B. (2017) ‘Carbonate minerals in the global carbon cycle’, *Chemical Geology*. Elsevier B.V., 449, pp. 58–72. doi: 10.1016/j.chemgeo.2016.11.029.

Massey, F. P., Roland Ennos, A. and Hartley, S. E. (2007) ‘Herbivore specific induction of silica-based plant defences’, *Oecologia*, 152(4), pp. 677–683. doi: 10.1007/s00442-007-0703-5.

Melo, V. F. *et al.* (2012) ‘Doses de basalto moído nas propriedades químicas de um Latossolo Amarelo distrófico da savana de Roraima’, *Acta Amazonica*, 42(4), pp. 471–476. doi: 10.1590/S0044-59672012000400004.

Mersi, W. Von, Kuhnert-Finkernagel, R. and Schinner, F. (1991) ‘The influence of rock powders on microbial activity of three forest soils’, *Zeitschrift für Pflanzenernährung und Bodenkunde*, 155(1), pp. 29–33. doi: 10.1002/jpln.19921550107.

Metso (2015) *Basics in Minerals Processing*. 10th edn. Edited by M. Corporation. Available at: <https://www.metso.com>.

Meysman, F. J. R. and Montserrat, F. (2017) ‘Negative CO₂ emissions via enhanced

CHAPTER 2

silicate weathering in coastal environments’, *Biology Letters*, 13(4), p. 20160905. doi: 10.1098/rsbl.2016.0905.

Mitchell, C., Mitchell, P. and Pascoe, R. (2008) *Quarry fines minimisation: Can we really have 10mm aggregate with no fines?*, Walton, Geoffrey, (ed.) *Proceedings of the 14th Extractive industry geology conference*. Available at: <http://nora.nerc.ac.uk/4854/>.

Moosdorf, N., Renforth, P. and Hartmann, J. (2014) ‘Carbon Dioxide Efficiency of Terrestrial Enhanced Weathering’, *Environmental science & technology*, 48, pp. 4809–4816. doi: 10.1021/es4052022.

Moraes, L. C. de, Seer, H. J. and Marques, L. S. (2018) ‘Geology, geochemistry and petrology of basalts from Paraná Continental Magmatic Province in the Araguari, Uberlândia, Uberaba and Sacramento regions, Minas Gerais state, Brazil’, *Brazilian Journal of Geology*, 48(0), pp. 221–241. doi: 10.1590/2317-4889201820170091.

Nduagu, E., Bergerson, J. and Zevenhoven, R. (2012) ‘Life cycle assessment of CO₂ sequestration in magnesium silicate rock - A comparative study’, *Energy Conversion and Management*. Elsevier Ltd, 55, pp. 116–126. doi: 10.1016/j.enconman.2011.10.026.

Nunes, J. M. G., Kautzmann, R. M. and Oliveira, C. (2014) ‘Evaluation of the natural fertilizing potential of basalt dust wastes from the mining district of Nova Prata (Brazil)’, *Journal of Cleaner Production*. Elsevier Ltd, 84(1), pp. 649–656. doi: 10.1016/j.jclepro.2014.04.032.

O’Connor, W. K. *et al.* (2005) *Aqueous Mineral Carbonation: Mineral Availability, Pretreatment, Reaction Parametrics, And Process Studies*, DOE. doi: 10.13140/RG.2.2.23658.31684.

Olivier, J. G. J., Schure, K. M. and Peters, J. A. H. W. (2017) *Trends in Global CO₂ and Total Greenhouse Gas Emissions*, PBL Netherlands Environmental Assessment Agency. Available at: <http://www.pbl.nl>.

Opolot, E. and Finke, P. A. (2015) ‘Evaluating sensitivity of silicate mineral dissolution rates to physical weathering using a soil evolution model (SoilGen2.25)’, *Biogeosciences*, 12(22), pp. 6791–6808. doi: 10.5194/bg-12-6791-2015.

- Orumwense, O. A. and Forssberg, E. (1992) ‘Superfine and Ultrafine Grinding - A Literature Survey’, *Mineral Processing and Extractive Metallurgy Review: An International Journal*, 11, pp. 107–127. doi: 10.1080/08827509208914216.
- Perrotta, M. M. *et al.* (2006) *Geologia e recursos minerais do estado de São Paulo*. Programa Nacional Geologia do Brasil. Available at: <http://rigeo.cprm.gov.br/jspui/handle/doc/2966> (Accessed: 15 August 2018).
- Ramezani, A. *et al.* (2013) ‘Addition of a volcanic rockdust to soils has no observable effects on plant yield and nutrient status or on soil microbial activity’, *Plant and Soil*, 367(1–2), pp. 419–436. doi: 10.1007/s11104-012-1474-2.
- Ramos, C. G. *et al.* (2015) ‘A preliminary evaluation of volcanic rock powder for application in agriculture as soil a remineralizer’, *Science of the Total Environment*. Elsevier B.V., 512–513, pp. 371–380. doi: 10.1016/j.scitotenv.2014.12.070.
- Ramos, C. G. *et al.* (2017) ‘Evaluation of the potential of volcanic rock waste from southern Brazil as a natural soil fertilizer’, *Journal of Cleaner Production*, 142, pp. 2700–2706. doi: 10.1016/j.jclepro.2016.11.006.
- REMIN (no date) *Improve Soil Health with Rock Dust | REMIN Scotland Ltd*. Available at: <http://www.remainscotland.com/home/how-to-use/> (Accessed: 21 September 2018).
- Renforth, P. *et al.* (2011) ‘Silicate production and availability for mineral carbonation’, *Environmental Science and Technology*, 45(6), pp. 2035–2041. doi: 10.1021/es103241w.
- Renforth, P. (2012) ‘The potential of enhanced weathering in the UK’, *International Journal of Greenhouse Gas Control*. Elsevier Ltd, 10, pp. 229–243. doi: 10.1016/j.ijggc.2012.06.011.
- Renforth, P., Pogge von Strandmann, P. A. E. and Henderson, G. M. (2015) ‘The dissolution of olivine added to soil: Implications for enhanced weathering’, *Applied Geochemistry*. Elsevier Ltd, 61, pp. 109–118. doi: 10.1016/j.apgeochem.2015.05.016.
- Rosado, L. P. *et al.* (2017) ‘Life cycle assessment of natural and mixed recycled aggregate production in Brazil’, *Journal of Cleaner Production*. Elsevier Ltd, 151, pp. 634–642. doi: 10.1016/j.jclepro.2017.03.068.

CHAPTER 2

Schenker, M. B. *et al.* (2009) ‘Pneumoconiosis from agricultural dust exposure among young California farmworkers’, *Environmental Health Perspectives*, 117(6), pp. 988–994. doi: 10.1289/ehp.0800144.

SEEG (2016a) *São Paulo - Emissions, System for Greenhouse Gas Emissions and Removal Estimates*. Available at: <http://plataforma.seeg.eco.br/territories/sao-paulo/card?year=2016> (Accessed: 14 September 2018).

SEEG (2016b) *Total Emissions - Brazil, System for Greenhouse Gas Emissions and Removal Estimates*. Available at: http://plataforma.seeg.eco.br/total_emission (Accessed: 14 September 2018).

Smith, P. *et al.* (2016) ‘Biophysical and economic limits to negative CO₂ emissions’, *Nature Climate Change*. Nature Publishing Group, 6(1), pp. 42–50. doi: 10.1038/nclimate2870.

Streffer, J. *et al.* (2018) ‘Potential and costs of carbon dioxide removal by enhanced weathering of rocks’, *Environmental Research Letters*, 13(3), p. 034010. doi: 10.1088/1748-9326/aaa9c4.

Taylor, L. L. *et al.* (2016) ‘Enhanced weathering strategies for stabilizing climate and averting ocean acidification’, *Nature Climate Change*, 6(4), pp. 402–406. doi: 10.1038/nclimate2882.

Taylor, L. L. *et al.* (2017) ‘Simulating carbon capture by enhanced weathering with croplands: An overview of key processes highlighting areas of future model development’, *Biology Letters*, 13(4). doi: 10.1098/rsbl.2016.0868.

Theodoro, S. H. and Leonardos, O. H. (2006) ‘The use of rocks to improve family agriculture in Brazil’, *Anais da Academia Brasileira de Ciencias*, 78(4), pp. 721–730. doi: 10.1590/S0001-37652006000400008.

UCLouvain (2017) *Climate Research Data Package (CRDP), ESA Climate Change Initiative*. Available at: <http://maps.elie.ucl.ac.be/CCI/viewer/download.php> (Accessed: 23 August 2018).

Wanke, P. and Fleury, P. F. (2006) *Transporte de cargas no Brasil: estudo exploratório*

das principais variáveis relacionadas aos diferentes modais e às suas estruturas de custos, Estrutura e Dinâmica do Setor de Serviços no Brasil. Available at: <https://www.en.ipea.gov.br>.

Washbourne, C. L. *et al.* (2015) ‘Rapid removal of atmospheric CO₂ by urban soils’, *Environmental Science and Technology*, 49(9), pp. 5434–5440. doi: 10.1021/es505476d.

Weidema, B. P. *et al.* (2013) *Data quality guideline for the ecoinvent database version 3.* Available at: <http://www.ecoinvent.org/database/methodology-of-ecoinvent-3/methodology-of-ecoinvent-3.html>.

Williamson, P. (2016) ‘Scrutinize CO₂ removal methods’, *Nature*, pp. 5–7. doi: 10.1038/530153a.

Williamson, P. and Bodle, R. (2016) *Update on Climate Geoengineering in Relation to the Convention on Biological Diversity: Potential Impacts and Regulatory Framework.* Montreal. Available at: <https://www.cbd.int/doc/publications/cbd-ts-84-en.pdf>.

CHAPTER 3

Soil Carbon Sequestration of Sugarcane Derived Biochar

“All models are wrong, but some are useful.”

GEORGE BOX

MODELLING THE POTENTIAL FOR SOIL CARBON SEQUESTRATION USING BIOCHAR FROM SUGARCANE RESIDUES IN BRAZIL

David Lefebvre ¹, Adrian Williams ¹, Jeroen Meersmans ^{1,2}, Guy J.D. Kirk ¹, Saran Sohi ³, Pietro Goglio ^{1,4}, and Pete Smith ⁵

¹ School of Water, Energy and Environment, Cranfield University, College Road, Bedford, MK43 0AL, UK

² TERRA Teaching and Research Centre, Gembloux Agro-Bio Tech, University of Liège, Gembloux 5030, Belgium

³ UK Biochar Research Centre (UKBRC), School of GeoSciences, Crew Building, University of Edinburgh, EH9 3FF, United Kingdom

⁴ Wageningen Economic Research, Wageningen University & Research, Leeuwenborch, Hollandsweg 1, 6706KN Wageningen, The Netherlands

⁵ Institute of Biological and Environmental Sciences, University of Aberdeen, 23 St Machar Drive, Aberdeen AB24 3UU, UK

3.1 Abstract

Sugarcane (*Saccharum officinarum* L.) cultivation leaves behind around 20 t ha⁻¹ of biomass residue after harvest and processing. We investigated the potential for sequestering carbon (C) in soil with these residues by partially converting them into biochar (recalcitrant carbon-rich material). First, we modified the RothC model to allow changes in soil C arising from additions of sugarcane-derived biochar. Second, we evaluated the modified model against published field data, and found satisfactory agreement between observed and predicted soil C accumulation. Third, we used the model to explore the potential for soil C sequestration with sugarcane biochar in São Paulo State, Brazil. The results show a potential increase in soil C stocks by 2.35 ± 0.4 t C ha⁻¹ yr⁻¹ in sugarcane fields across the State at application rates of 4.2 t biochar ha⁻¹ yr⁻¹. Scaling to the total sugarcane area of the State, this would be 50 Mt of CO₂ equivalent yr⁻¹, which is 31% of the CO₂ equivalent emissions attributed to the State in 2016. Future research should (a) further validate the model with field experiments; (b) make a full life cycle assessment of the potential for greenhouse gas mitigation, including additional effects of biochar applications on greenhouse gas balances

3.2 Introduction

Sugarcane (*Saccharum officinarum* L.) is the world's largest crop by production quantity, with a total of 1.8 billion tonnes of cane produced globally per annum in more than 90

CHAPTER 3

countries (FAO, 2019). Sugarcane fields were traditionally burned to facilitate manual harvest (Hassuani, Leal and Macedo, 2005). However, to avoid air pollution, in many countries the fields are now mostly left unburned and harvested mechanically. This ‘green harvesting’ leaves large quantities of biomass (hereafter referred as ‘trash’) in the field (Romero *et al.*, 2007). Although trash provides a mulch that can benefit soil fertility and the growth of subsequent crops, it can also increase the risk of fire, pest proliferation, and reduced soil warming and drying in the spring (Hassuani, Leal and Macedo, 2005; Cardoso *et al.*, 2015). Currently it is typical for all the trash to be left on the field, although studies into sustainable rates of removal have been made (Cardoso *et al.*, 2015; Caldeira-Pires *et al.*, 2018). A potential alternative use of the trash is for energy generation, substituting for fossil fuels (Wang *et al.*, 2014; Peters, Iribarren and Dufour, 2015). Another option is to make biochar, which potentially provides greenhouse gas (GHG) removal as well as returning carbon (C) and nutrients to the soil (Smith, 2016; El-Naggar, Awad, *et al.*, 2018). It is also argued that biochar has additional GHG abatement potential through effects on crop production, including reduced requirement for manufactured fertilizer (Peters, Iribarren and Dufour, 2015).

Predicting the potential of biochar for these purposes requires allowance for the wide range of biochar types that can be created, and the variable effects of soil conditions on biochar decomposition, and vice versa. The properties of biochar vary according to pyrolysis conditions and other manufacturing parameters as well as the nature of the biomass ‘feedstock’ (Kambo and Dutta, 2015; El-Naggar *et al.*, 2019). The effects on existing soil organic carbon (SOC) may lead to increased SOC mineralization (‘positive priming’) or decreased mineralization (‘negative priming’) (Wang *et al.*, 2016; Ding *et al.*, 2018). Studies report negative, positive and no priming effect (Wang *et al.*, 2016; Ding *et al.*, 2018; El-Naggar, Lee, *et al.*, 2018), sometimes with a change in the direction of priming over time, typically from increased SOC mineralisation in the first year or so, to decreased mineralization thereafter (Ding *et al.*, 2018). In a recent meta-analysis, Wang *et al.* (2016) found a wide range of priming effects depending on biochar and soil characteristics; but they found consistently large priming effects in low-fertility sandy soils which are typical of many sugarcane areas.

Attempts to model long-term increases in SOC following biochar application have relied on data from short-term studies (Leng *et al.*, 2019). Dil and Oelbermann adapted the CENTURY model to evaluate the long-term effect of biochar by representing it as 95% lignin added to the ‘slow C pool’ in CENTURY (Dil and Oelbermann, 2014). However, this pool has a turnover time of 10 to 50 yr (Parton *et al.*, 2001) which is at least an order of magnitude faster than typical biochar turnover (Zimmerman, 2010). Archontoulis *et al.* developed a biochar sub-model for the Agricultural Production System Simulator (APSIM), and its simulations compared favourably with some experimental observations, but it lacked wider calibration and validation (Archontoulis *et al.*, 2016). Lychuk *et al.* developed a sub-model to integrate biochar into the Environmental Policy Integrated Climate (EPIC) model by allowing for the effect of biochar on the initial soil properties represented in the model, but not explicitly for biochar turnover (Lychuk *et al.*, 2014). Mondini *et al.* modified the RothC SOC model to better describe the decomposition of exogenous organic matter, such as biochar, but without specific modifications for biochar (Mondini *et al.*, 2017). Overall, none of the existing models of biochar turnover in soils is suitable for our purposes, as these models were not optimized for biochar-amended soils.

In this paper, we develop and test a biochar sub-model for the RothC model with which to assess the potential for excess sugarcane trash and the bagasse residues to produce biochar for in-field soil C sequestration. The sub-model divides the biochar C into fresh plant material, which is fed into RothC, and recalcitrant material, which slowly decomposes to CO₂; and the combined model predicts long-term changes in SOC as a function of biochar properties and soil, cropping and environmental conditions. We evaluate the model against published data and use it to make predictions for São Paulo State in Brazil using available data on sugarcane production and relevant soil and climate conditions. São Paulo is a suitable case study because Brazil accounts for 40% of global sugarcane production and São Paulo accounts for 55% of the national production (Cardoso *et al.*, 2019) with 96% of its sugarcane fields now mechanically harvested (Cardoso *et al.*, 2019; Gonzaga *et al.*, 2019). We compare three scenarios in which either 100%, 50% or 25% of the available sugarcane residues are used to produce biochar, with the remaining residues added to the fields as fresh material. Thereby we provide the first

assessment of the carbon capture potential of sugarcane biochar at a regional scale, accounting for different climatic and soil characteristics.

3.3 Results and Discussion

3.3.1 Model evaluation.

Comparison of simulated values against the experimental observations from Liu *et al.* (2020) is presented in Figure 3.1. Accounting for leaching of biochar particles to sub-soil at 5.2% of the biochar per yr (Section 3.4.4) improved the agreement with the experimental data. Liu *et al.* reported a loss rate of between 11.8 – 19.1% of the BC-C over the 5 years, estimated by mass balance (Liu *et al.*, 2020). Our model reaches a loss of 4% of BC-C after 5 years without leaching and 12% with leaching, with no modelled interaction with application rate.

Additional limited evaluation of the model was possible by comparison with the results of controlled-environment experiments (soil–biochar, plant free, incubation experiments) and the dataset used by Archontoulis *et al.* (2016) to validate their APSIM biochar sub-model (Supplementary Information, Appendix B). The evaluation results using that prior dataset (Archontoulis *et al.*, 2016) (Supplementary Figure B.4) fits with their biochar sub-model validation results. Comparison with incubation experiments showed that our modelled biochar mineralization rate reflects the general trend reported in the literature (Supplementary Figure B.5).

The extent of this evaluation exercise is limited but we consider it the best available at the current time, owing to the general paucity of long-term data. The intensive investigation of biochar is rather recent compared to the heritage of long-established field experiments (Li *et al.*, 2018). The good level of fit provides some confidence in the model predictions.

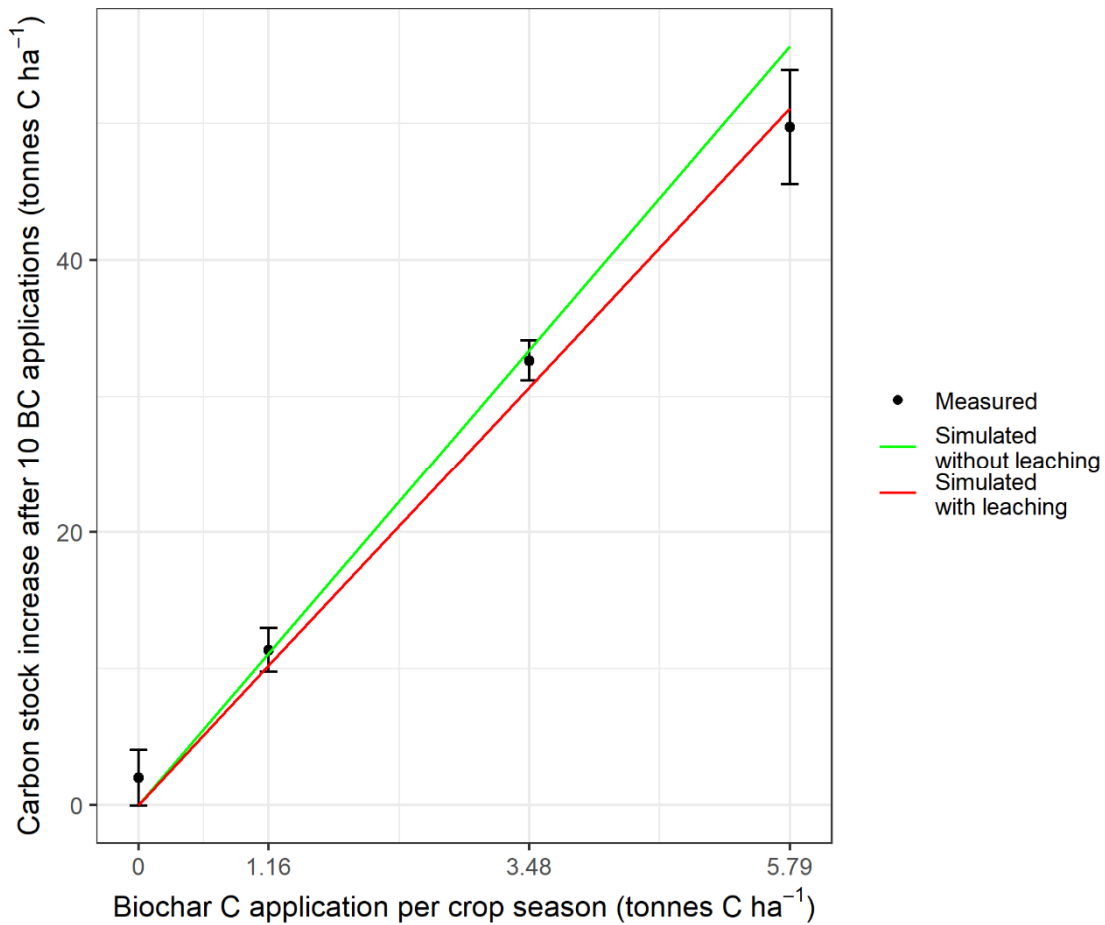


Figure 3.1: Comparison of the soil C stock increase after 5 years of biochar addition (10 applications of biochar) between measured values (black dots $\pm$ SD) and simulated (red and green lines) using the modified RothC model described in this manuscript. The red line accounts for the loss of 5.2% year⁻¹ of the biochar via leaching.

3.3.2 Potential C sequestration.

Figure 3.2 shows the predicted SOC stocks in sugarcane fields across São Paulo for the three biochar application scenarios (Section 3.4.3). There is a slightly higher C stock in the north-eastern region of São Paulo (Figure 3.2 A), consistent with field measurements (Maia *et al.*, 2014). This region is characterized by a clayey soil (Rhodic Ferralsols according to the FAO classification (Food and Agriculture Organization of the United Nations, 2014) or Ustox according to the USDA classification (USDA, 1999)) and mild climate (Supplementary Table B.2). Both characteristics are known to either increase the C storage capacity in soils (i.e. high clay contents) (Hartemink and McSweeney, 2014)

CHAPTER 3

or decrease the mineralization rate of C in soil (i.e. low temperatures) (Davidson and Janssens, 2006).

Under Scenario 1 (Figure 3.2 B-D), there is a steady increase in the SOC stock due to the incremental addition of biochar. Under Scenarios 2 (Figure 3.2 E-G) and 3 (Figure 3.2 H-J), there is a similar SOC increase in the first 5 yr after biochar application but a lower soil C stock increase after 10 and 20 yr as compared to Scenario 1. The reduction at 10 and 20 yr indicates that the conventional soil C pools reach new equilibria, while the biochar added in Scenario 1, present in the recalcitrant (RBC) pool, is subject to much slower mineralization and progressively expands.

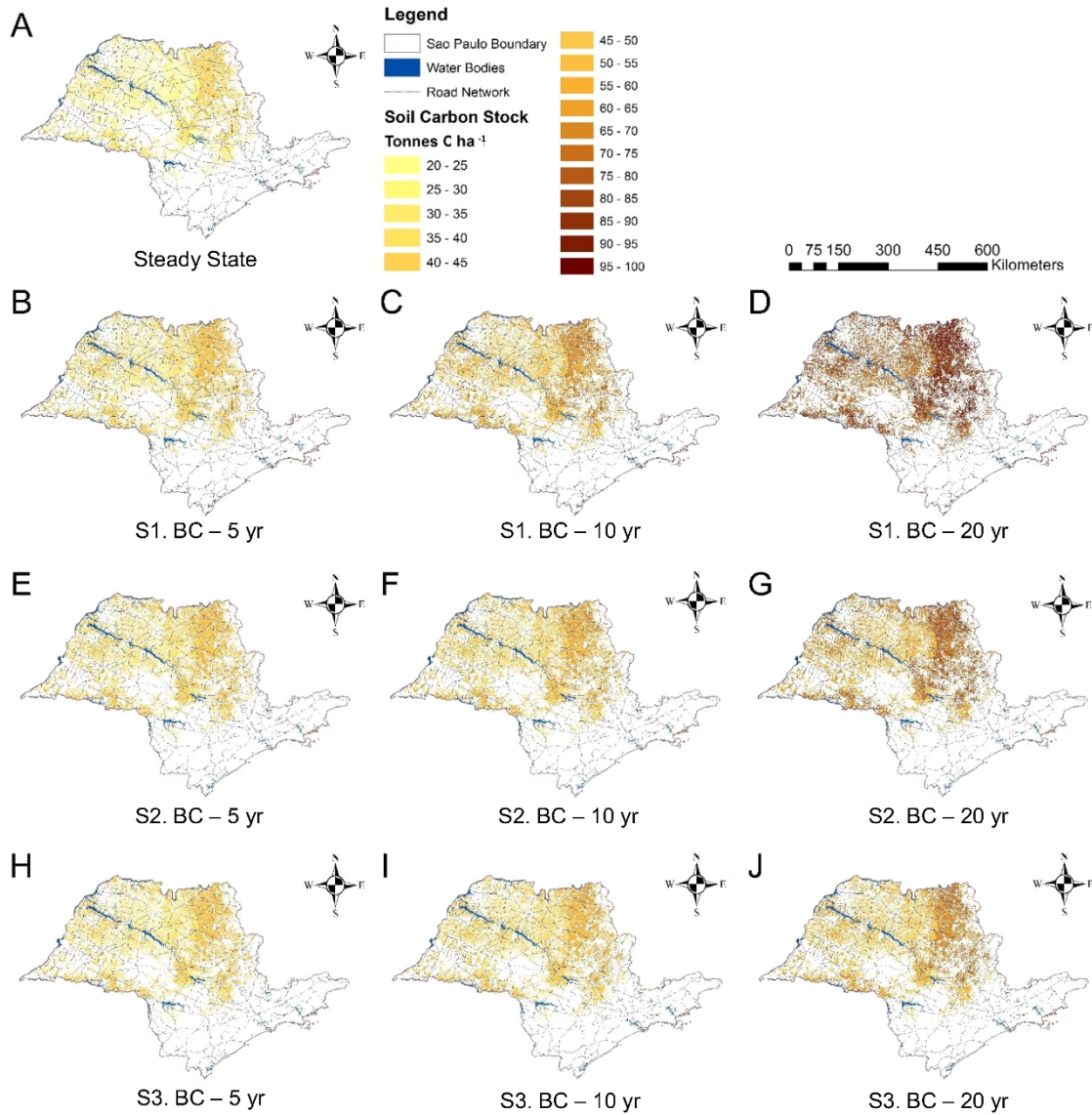


Figure 3.2: Predicted SOC stock in sugarcane fields across São Paulo. At steady state (A) and after 5, 10, and 20 year of biochar addition for Scenarios 1 (B, C, and D respectively), 2 (E, F, G), and 3 (H, I, J). For Scenarios 1, 2, and 3, the biochar additions are 2.46, 1.23, and 0.62 t C ha⁻¹ year⁻¹ respectively and the fresh C inputs are 6.57, 9.74, and 11.1 t C ha⁻¹ year⁻¹, respectively as trash, bagasse, roots, root exudates, and applications of vinasse and filter cake.

Figure 3.3 shows the C stock increase over the three considered periods for each soil type and biochar scenario. The consistently lower C stock at 10 and 20 yr for Scenarios 2 and 3 reflects the results shown in Figure 3.2. The C accumulation rate decreases over time for both Scenarios 2 and 3 as a new SOC content equilibrium is reached. The increased

CHAPTER 3

addition of fresh plant material in Scenario 2 and 3 leads to a sudden increase of decomposable carbon in the soil system, which will eventually supply all carbon pools and affect their transformation rate to reach a new equilibrium. This slow process explains the decreasing rate of soil C stock increase witnessed for Scenario 2 and 3 as, with time, the additional fresh organic matter modelled in these Scenarios will not affect the soil C stock anymore. The overall higher C stocks in Ferralsols is due to their higher clay contents (Supplementary Table B.1).

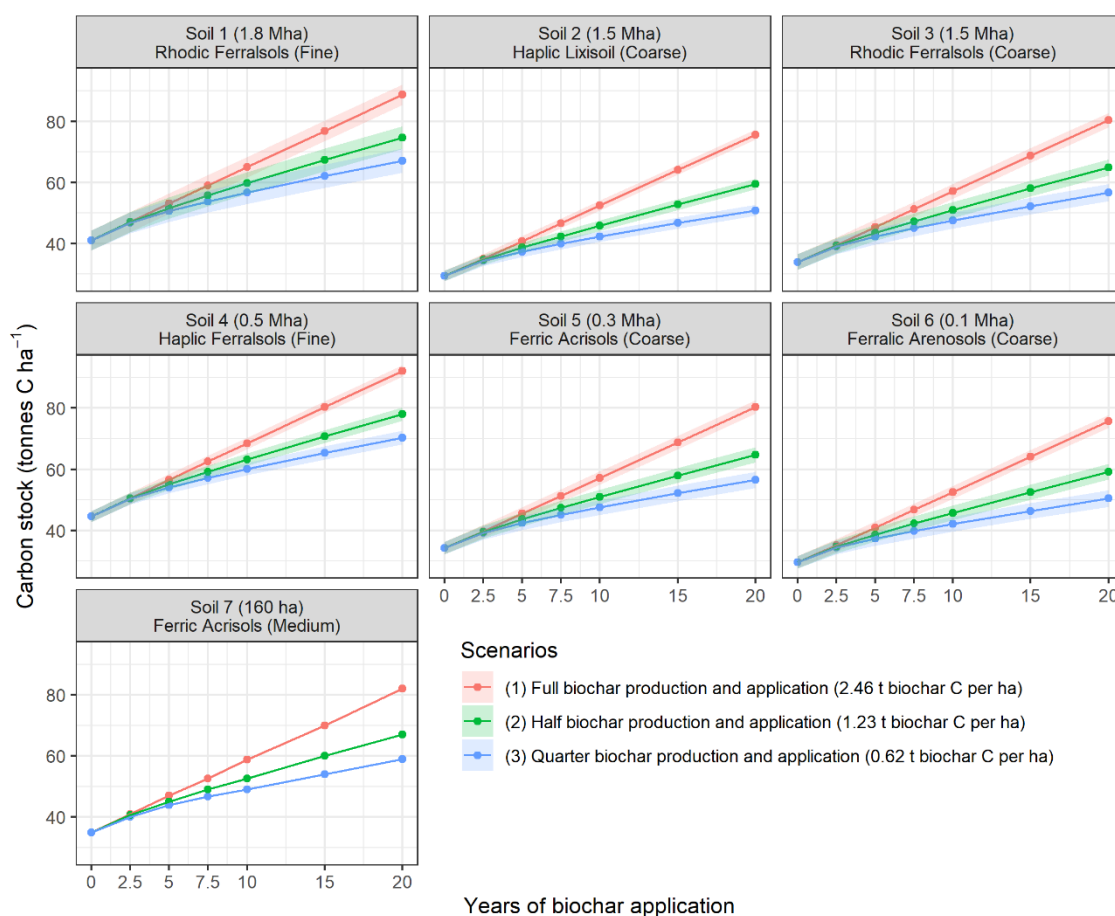


Figure 3.3: Effect of FAO soil type on predicted SOC stock up to 20 year. Dots are means across sugarcane fields at the indicated times (year 0 is the steady state value); ribbon width is 1 SD due to geographical climate variability; lines indicate general trends and not intermediate points. The SD is small for Soil 7 because they account for only a few sugarcane fields concentrated in one particular region (Supplementary Table B.3). The land area associated with each soil type is shown in parenthesis for each plot.

3.3.3 Priming effects.

Including biochar-induced priming reduces the predicted increases in SOC (Figure 3.4). Since priming was modelled independent of dose, the proportional effect on SOC is least when the addition of biochar is highest (Scenario 1). The SOC accumulation relative to baseline was -4.1% using the conservative assumption (SOC decomposition rate increased 21%) and 13.9% for the extreme assumption (SOC decomposition increased 91%). The corresponding effects for Scenario 2 were -6.1% and -20.6%. The effects for Scenario 3 were -7.5% and -25.3%. Overall, a biochar-induced positive priming effect could impair the C sequestration potential of the practice. However, even under the extreme degrees of priming effect modelled here, the net SOC balance with biochar addition remains positive, resulting in substantial carbon sequestration and CO₂ removal.

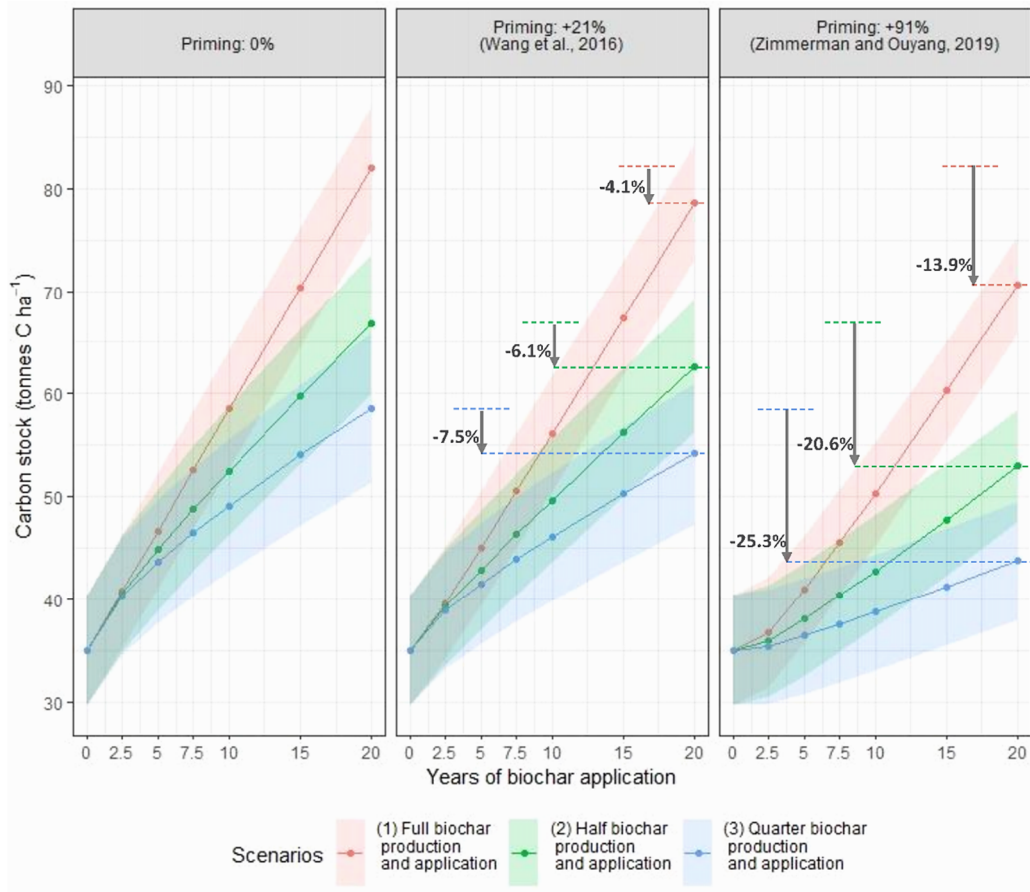


Figure 3.4: Priming effect of biochar on SOC under the three addition scenarios. Dots are mean values across sugarcane fields at the indicated times (year 0 is the steady state value); ribbon width is due to climate and soil variability (1 SD). The downward arrow indicates the % decrease in SOC stock after 20 year compared to no priming.

3.3.4 Sequestration potential of the whole state.

These three scenarios result in a wide range of potential C sequestration over all 5.77 Mha (Theodor Rudorff, 2014) of sugarcane fields in São Paulo State (Figure 3.5). Considering 20 yr of biochar application, the most effective scenario in term of C sequestration (Scenario 1, no priming of SOC) could sequester 13.5 Mt of C per year over the State (49.5 Mt of CO₂e), or 31% of the 159 Mt CO₂e emissions attributed to the State in 2016 (SEEG, 2016). On the other hand, the least promising scenario (Scenario 3, assumption of extreme priming) could sequester 2.5 Mt of C per year over the State (9.1 Mt of CO₂e), or 6% of the 159 Mt CO₂e attributed to the State in 2016 (SEEG, 2016). These numbers indicate the sequestration potential of biochar application on the sugarcane fields of São Paulo. As far as we are aware, there are no long-term field experiments on the C sequestration potential of sugarcane biochar in Brazil or elsewhere, with which to test our model predictions. This needs to be done in future.

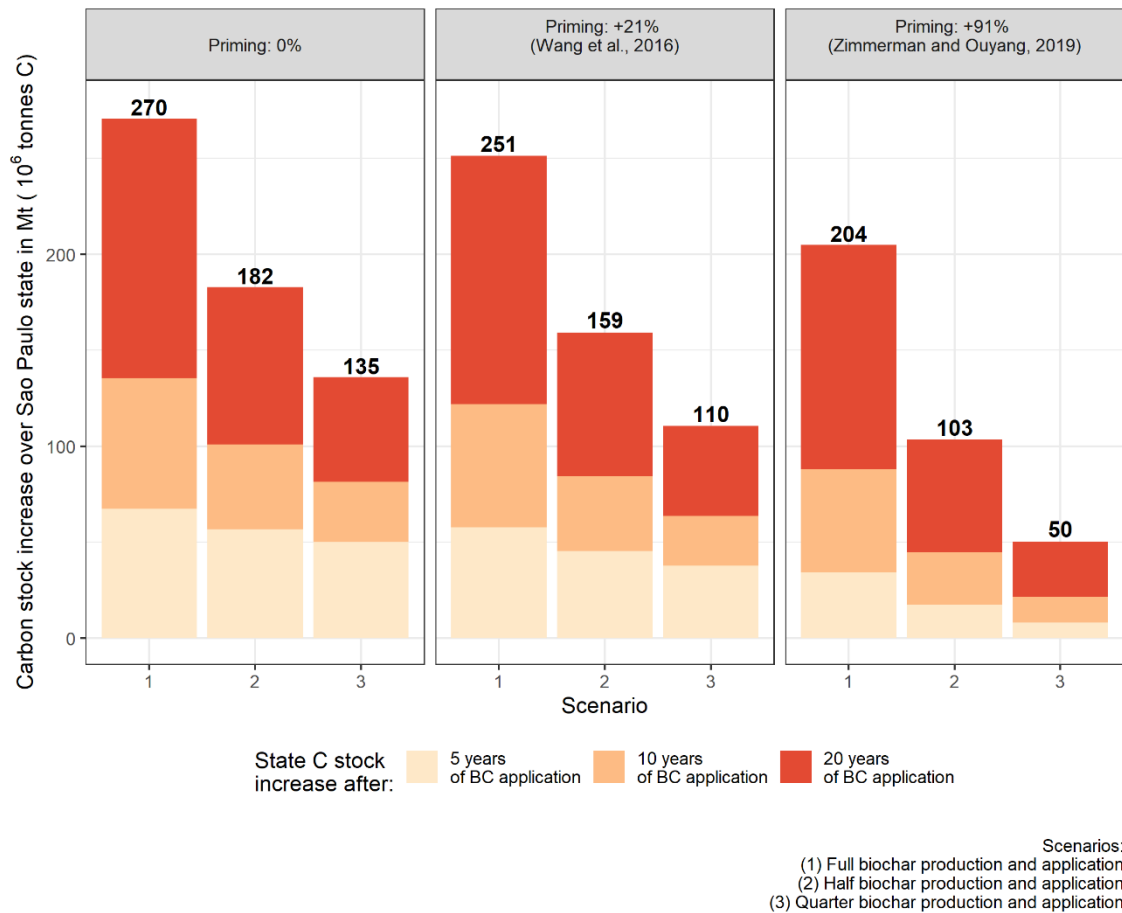


Figure 3.5: São Paulo State C sequestration potential. The number in bold above each bar represent the State total C sequestered after 20 years of biochar addition in Mt of C (10^6 t C).

We have not allowed for possible increases in sugarcane yield over time with biochar incorporation. The literature reports wide ranges in yield effects of biochar depending on crop type, soil conditions, climate, and biochar characteristics (Verheijen *et al.*, 2009; Jeffery *et al.*, 2011, 2017; Biederman and Harpole, 2013; Crane-Droesch *et al.*, 2013; Liu *et al.*, 2013, 2019). In general, yield responses are smaller for perennial crops, such as sugarcane, compared to annual crops (Biederman and Harpole, 2013; Liu *et al.*, 2013; Jeffery *et al.*, 2017). In any case, the management of sugarcane plantations in São Paulo is such that yields are already apparently optimized and there is little room left for improvement. According to the IBGE (Instituto Brasileiro de Geografia e Estatística), yields stabilized around 2007 (IBGE, 2019).

3.3.5 Potential additional greenhouse gas emissions

Biochar addition may affect N₂O emissions during nitrification and denitrification in soils, at least in the first year following application (Cowie *et al.*, 2015; Borchard *et al.*, 2018). Meta-analyses of field studies indicate 28% $\pm$ 16% lower N₂O emissions with various types of biochar application (Cayuela, Jeffery and van Zwieten, 2015), but the wide range of results indicate this is one of the most uncertain components of the GHG balance (Wang *et al.*, 2014).

Similar analysis suggest biochar may decrease CH₄ emissions from soils, particularly if flooded or acidic or both (Jeffery *et al.*, 2016). However, biochar applied to unflooded neutral or alkaline soils can increase CH₄ emissions. Taking landscape diversity and CH₄ uptake (oxidation) into account, it has also been suggested that biochar does not affect net CH₄ release (Ji *et al.*, 2018).

Other biochar aspects that potentially influence the GHG balance include the potential reduced need for irrigation due to improved soil water holding capacity with biochar (Cowie *et al.*, 2015), reduced fertilizer requirement due to the phosphorus and potassium available in the applied biochar (Cowie *et al.*, 2015; Glaser and Lehr, 2019) and reduced nitrogen leaching losses (Hagemann *et al.*, 2017).

While the model provides an efficient way to predict potential increases in SOC stocks in sugarcane fields following biochar addition, this constitutes only part of the GHG balance of the overall practice. Emissions during biochar production, transport and application and those discussed above should be considered in a full Life Cycle Assessment of the integration of biochar into sugarcane systems, so as to provide a more accurate figure for the carbon sequestration potential.

3.4 Method

3.4.1 Sugarcane production characteristics

In our scenarios, we have assumed that 7 t DM trash ha⁻¹ yr⁻¹ is left on the field to assist active cycling of organic matter (Ottoni *et al.*, 2018; Silva *et al.*, 2019; Popin *et al.*, 2020). We assumed that trash amounted to 140 kg DM per tonne of harvested cane (Waldheim, Monis and Verde Leal, 2001; Hassuani, Leal and Macedo, 2005), in agreement with

previous work (Caldeira-Pires *et al.*, 2018). Likewise, we assumed that the potentially available bagasse (pith and rind from cane progressing) amounted to 140 kg DM per tonne of harvest cane (Waldheim, Monis and Verde Leal, 2001; Khatiwada *et al.*, 2016). We used these data to determine the input of fresh C to the field for three different scenarios and a baseline (Section 3.4.3). The calculation draws on the estimated C content for trash and/or bagasse, adding estimates for the C input from root decay, root exudates, plus vinasse and filter cake (the aqueous and suspended solid fractions from the effluent of cane processing). The potential biochar C attributable to each hectare per year was obtained based on the biochar yield and C content of sugarcane trash and bagasse biochar.

We considered the seven soil types covering the sugarcane area of São Paulo and obtained meteorological data from 10 weather stations across and surrounding the area. Details of the soils and meteorological stations, a map of the study area, and details on the data used for soil C stock modelling are given in the Supplementary Table B.1 to Table B.4.

3.4.2 Soil carbon model

We combined RothC with a sub-model for biochar decomposition. In RothC, fresh plant material (influx I_0) is divided between two pools of differing decomposability (DPM and RPM) which decompose to two SOC pools of differing decomposability (BIO and HUM), which then inter-convert (Figure 3.6) (Coleman and Jenkinson, 2014). Each pool decomposes with first order kinetics, and in each inter-pool transformation, a proportion is lost as CO₂.

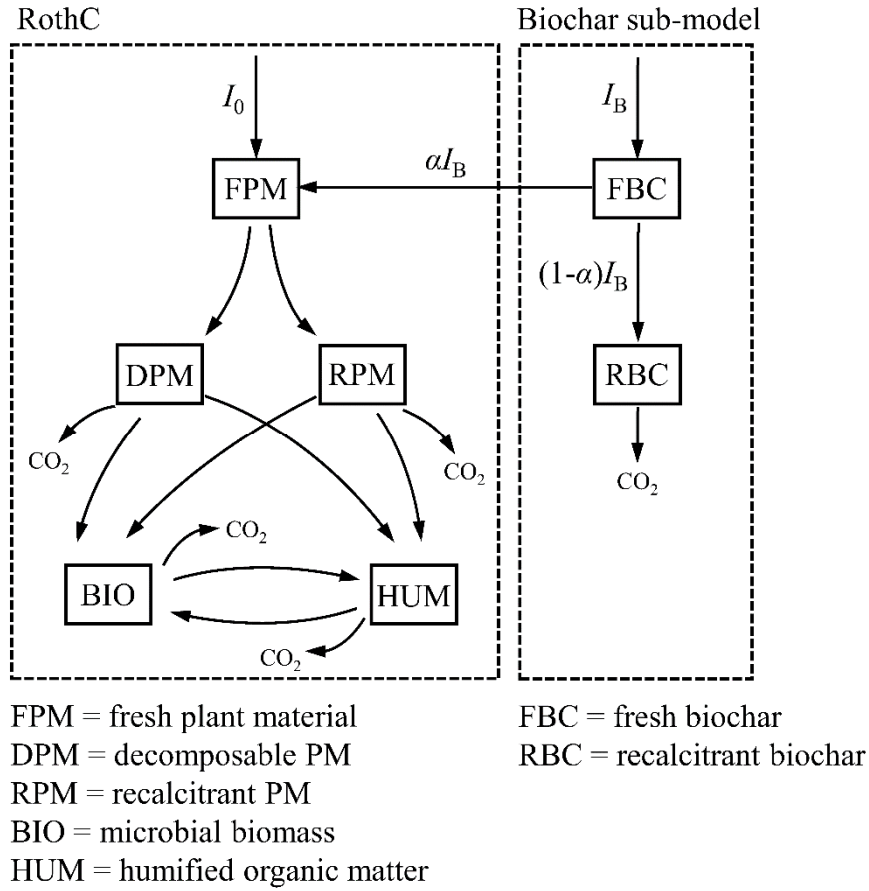


Figure 3.6: Soil carbon model. RothC is combined with a sub-model for the decomposition of fresh biochar. Details in text.

In the biochar sub-model, a proportion, α , of the C in fresh biochar (influx I_B) is treated as fresh plant material and added to FPM pool in RothC (flux $I_B = \alpha I_B$). The remaining, recalcitrant material (RBC) decomposes very slowly releasing CO_2 . The products of such slow decomposition will have a minor impact on soil C pools (BIO and HUM) and can be neglected for simplicity. Hence

$$\frac{dC_{\text{RBC}}}{dt} = (1 - \alpha)I_B - k_{\text{RBC}}C_{\text{RBC}} \quad (3-1)$$

where C_{RBC} is the concentration of RBC and k_{RBC} is its decomposition rate constant.

We nominally parameterised the coupled models for biochar produced by slow pyrolysis of bagasse and trash at 550°C. Lacking sufficient specific data we considered 3% of the applied biochar C as the portion reflecting the degradability of fresh plant material

(DPM/RPM ratio of 1.44, similar to sugarcane residues), based on a recent meta-analysis of all biochar types (Wang *et al.*, 2016) and which was consistent with previous assessments (Foereid, Lehmann and Major, 2011; Woolf and Lehmann, 2012). Based on a sugarcane biochar specific incubation study, we assumed that the remaining fraction would decompose at a rate of 11.9% over 100 yr (Zimmerman, 2010), i.e. mean residence time = 840 yr, consistent with 560 ± 480 yr calculated by Wang *et al.* (2016). Hence $k_{RBC} = 0.00119 \text{ yr}^{-1}$. This k value is consistent with recent IPCC guidelines for estimating the change in SOC stock in mineral soils from biochar amendment (IPCC, 2019).

We explored the sensitivity of the model to positive priming of SOC turnover by biochar by increasing the RothC rate constants of all pools except RBC by (a) 21% based on Wang *et al.*'s (2016) meta-analysis, which reported a mean 21% increase in SOC turnover with biochar applications to sandy soils; and (b) by 91% based on increased sucrose turnover with sugarcane bagasse biochar in an incubation experiment with a simulated soil (Zimmerman and Ouyang, 2019). We consider the latter to indicate the maximum possible positive priming effect of biochar, since most SOC is highly stabilised and unlikely to respond as the labile material. Since published meta-analyses encompass short-term study of large doses and single additions, our permanent increase of C turnover across all pools is highly conservative with respect to SOC.

We used the RothC model from the package 'SoilR' (Sierra, Müller and Trumbore, 2012), coded in R (Core Team, 2018). We amended the SoilR code to obtain output that better corresponded to the original RothC model for its own calibration data (see Supplementary Information, Appendix B). At steady state, before any biochar application, the 'inert' C stock for São Paulo soils was calculated according to Falloon *et al.* (1998). Model results are presented using ArcGIS 10.5.1. (ESRI, 2011). To smoothen the values between climate stations, we interpolated the meteorological data using inverse distance weighting, which uses the distance of known points to unknown points to estimate their values (Li and Heap, 2008). The graphical representations of the results were created using R package 'ggplot2' (Wickham, 2016) from R software version (3.5.1) (Core Team, 2018).

3.4.3 Modelled Scenarios

We considered three scenarios for biochar and organic matter management together with a baseline (Figure 3.7). We used the baseline scenario to obtain the steady-state C stock expected in the long-term under emergent practice and in the absence of climate change. It considers that 67% of sugarcane trash amounting to $3.17 \text{ t C ha}^{-1} \text{ yr}^{-1}$ ($7 \text{ t DM ha}^{-1} \text{ yr}^{-1}$) is left on the field. The remaining trash is supplied to a combined heat and power plant, along with 100% of the bagasse. The total fresh organic C input to the field is $6.57 \text{ t C ha}^{-1} \text{ yr}^{-1}$, since the C from trash is supplemented by below ground inputs (estimate for decaying roots and root exudates, after allowing for rhizome growth) as well as the application of 100% vinasse and filter cake (Supplementary Table B.4). We ran the model for 500 yr to determine the steady-state SOC content, allowing for the effects of intermittent replanting (Scarpore *et al.*, 2016). Scenarios 1, 2 and 3 represent different managements relative to this baseline.

In Scenario 1, 1.29 and $4.62 \text{ t C ha}^{-1} \text{ yr}^{-1}$ of the available trash and bagasse respectively (i.e. 100% of each) are supplied to the pyrolysis plant, which yields $2.46 \text{ t biochar C ha}^{-1} \text{ yr}^{-1}$ to be returned to the sugarcane fields (calculation details in Supplementary Information, Appendix B). These are repeated annual applications. The other inputs are unchanged. In Scenario 2, only half of the potential biochar production from trash and bagasse is realised (i.e. $1.23 \text{ t biochar C ha}^{-1} \text{ yr}^{-1}$ – from 60% of the available bagasse). The remainder is left in (or returned to) the field, increasing fresh organic C input from 6.57 in Baseline and Scenario 1 to $9.73 \text{ t C ha}^{-1} \text{ yr}^{-1}$. In Scenario 3, the maximum potential biochar production is further diminished to one-quarter of the potential, amounting to $0.62 \text{ t biochar C ha}^{-1} \text{ yr}^{-1}$ (from 30% of the available bagasse). The remainder is left in (or returned to) the field, increasing fresh organic C input to $11.1 \text{ t C ha}^{-1} \text{ yr}^{-1}$.

All the scenarios include a loss of 5% of the trash dry matter during the stalk collection and transport (Sampaio *et al.*, 2019). Scenarios 1, 2 and 3 are run for 5, 10, and 20 yr, starting at the steady-state (baseline) C stock. A maximum of 20 yr was chosen to match the higher experimental biochar additions tested in any cropping system (Jeffery *et al.*, 2017; Alotaibi and Schoenau, 2019).

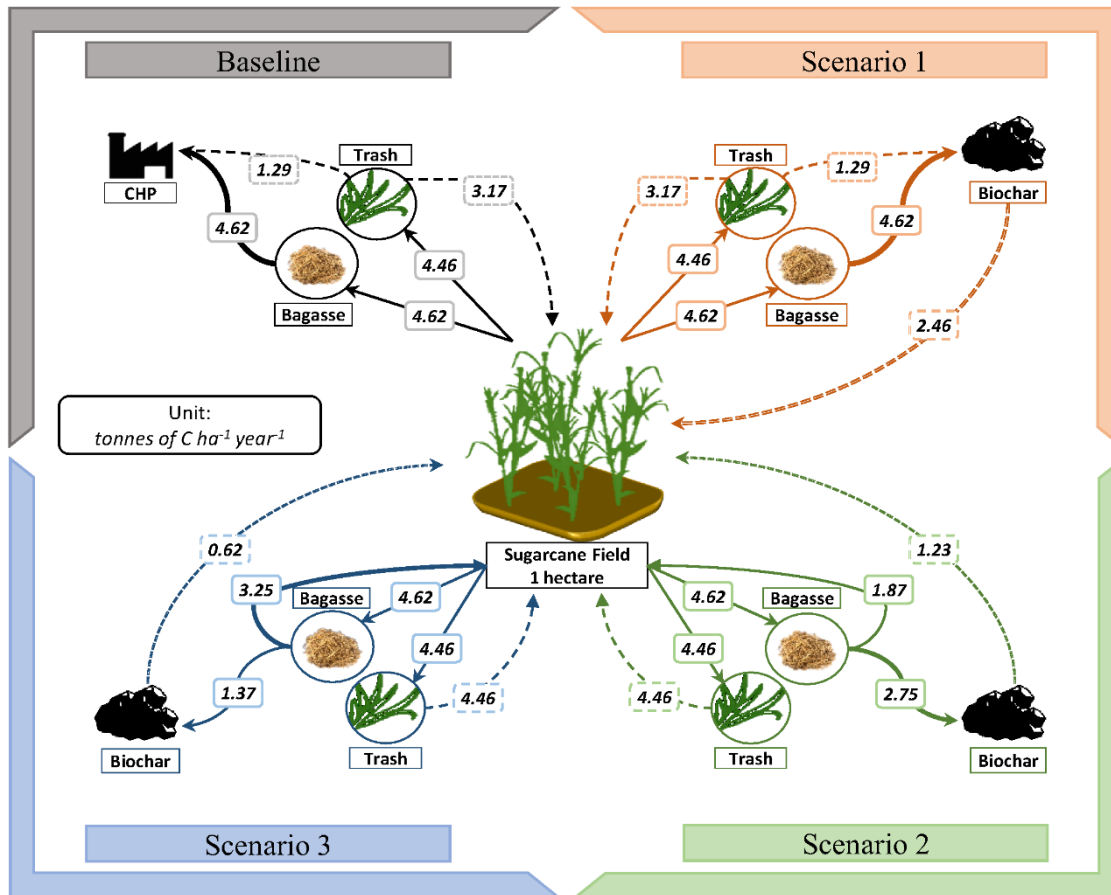


Figure 3.7: Scenario flowchart representing the fate of the bagasse and trash C produced per hectare per year on a sugarcane field in São Paulo. ‘CHP’ stands for ‘combined heat and power plant’.

3.4.4 Model evaluation

To attach confidence to our analysis, we decided to test the biochar sub-model that we coupled to RothC. The established RothC model has successfully described SOC turnover in many parts of the world including Brazil (Cerri *et al.*, 2007; Falloon *et al.*, 2007). The relevance of the coupled biochar sub-model is not diminished relative to RothC, as it contains parameters derived from meta-analysis and reflecting all published biochar research, so although it would have been ideal to calibrate against sugarcane crop data, it was not essential. Since there are to our knowledge no published data on long-term field experiments on biochar in sugarcane, we instead used data from a wheat-maize study in China (Liu *et al.*, 2020). In this study 0, 1.16, 3.48 or 5.79 t of rice straw biochar C ha⁻¹ were applied to both the wheat and following rice crop in each year. This study is useful as it is both long-term and involves repeated additions of biochar to all land,

representative of our modelled scenarios. Data required for the model evaluation are reported in the Supplementary Information - Table B.5).

The biochar literature suggests that some biochar is transported after topsoil application to sub-soil layers by rainfall (infiltration) and/or bioturbation (Haefele *et al.*, 2011; Obia *et al.*, 2016). Between 0.5% and 14.75% of applied biochar is found below the application depth (ranging between 5 and 20 cm) after one year (Major *et al.*, 2010; Haefele *et al.*, 2011; Singh *et al.*, 2015; Obia *et al.*, 2016; Ventura *et al.*, 2019), depending on the biochar particle size (Obia *et al.*, 2016), rainfall amount (Obia *et al.*, 2016) and soil texture and porosity (Haefele *et al.*, 2011; Obia *et al.*, 2016). Biochar movement to subsoil layers or beyond will not reduce net C sequestration and may even increase it compared with more-transient storage in the topsoil (Masiello and Druffel, 1998; Kögel-Knabner *et al.*, 2010), but could introduce errors in field experiments with limited depth of sampling. For the purpose of model evaluation by deriving a relation between percentage downward loss and soil texture using data reported in the literature (Major *et al.*, 2010; Haefele *et al.*, 2011; Singh *et al.*, 2015; Obia *et al.*, 2016; Ventura *et al.*, 2019), a probable loss could be derived for sandy loam soil of the wheat–maize experiment used in calibration data (Liu *et al.*, 2020). The value was 5.18% of the applied biochar per yr.

3.5 References

- Alotaibi, K. D. and Schoenau, J. J. (2019) ‘Addition of Biochar to a Sandy Desert Soil: Effect on Crop Growth, Water Retention and Selected Properties’, *Agronomy*, 9(6), p. 327. doi: 10.3390/agronomy9060327.
- Archontoulis, S. V. *et al.* (2016) ‘A model for mechanistic and system assessments of biochar effects on soils and crops and trade-offs’, *GCB Bioenergy*, 8(6), pp. 1028–1045. doi: 10.1111/gcbb.12314.
- Biederman, L. A. and Harpole, W. S. (2013) ‘Biochar and its effects on plant productivity and nutrient cycling: a meta-analysis’, *GCB Bioenergy*, 5(2), pp. 202–214. doi: 10.1111/gcbb.12037.
- Borchard, N. *et al.* (2018) ‘Biochar, soil and land-use interactions that reduce nitrate leaching and N₂O emissions: A meta-analysis’, *Science of The Total Environment*. Elsevier B.V., 651, pp. 2354–2364. doi: 10.1016/j.scitotenv.2018.10.060.

- Caldeira-Pires, A. *et al.* (2018) 'Implications of removing straw from soil for bioenergy: An LCA of ethanol production using total sugarcane biomass', *Journal of Cleaner Production*. Elsevier Ltd, 181, pp. 249–259. doi: 10.1016/j.jclepro.2018.01.119.
- Cardoso, T. F. *et al.* (2015) 'A vertical integration simplified model for straw recovery as feedstock in sugarcane biorefineries', *Biomass and Bioenergy*, 81, pp. 216–223. doi: 10.1016/j.biombioe.2015.07.003.
- Cardoso, T. F. *et al.* (2019) 'A regional approach to determine economic, environmental and social impacts of different sugarcane production systems in Brazil', *Biomass and Bioenergy*. Elsevier Ltd, 120(April 2018), pp. 9–20. doi: 10.1016/j.biombioe.2018.10.018.
- Cayuela, M. L., Jeffery, S. and van Zwieten, L. (2015) 'The molar H: C_{Org} ratio of biochar is a key factor in mitigating N₂O emissions from soil', *Agriculture, Ecosystems and Environment*. Elsevier B.V., 202, pp. 135–138. doi: 10.1016/j.agee.2014.12.015.
- Cerri, C. E. P. *et al.* (2007) 'Simulating SOC changes in 11 land use change chronosequences from the Brazilian Amazon with RothC and Century models', *Agriculture, Ecosystems and Environment*, 122(1), pp. 46–57. doi: 10.1016/j.agee.2007.01.007.
- Coleman, K. and Jenkinson, D. S. (2014) *RothC - A model for the turnover of carbon in soil Model - Model description and users guide*. Harpenden: Routledge. Available at: https://www.rothamsted.ac.uk/sites/default/files/RothC_guide_WIN.pdf.
- Core Team, R. (2018) *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. Vienna. Available at: <https://www.r-project.org/>.
- Cowie, Annette *et al.* (2015) 'Biochar, carbon accounting and climate change', in Lehmann, J. and Joseph, S. (eds) *Biochar for environmental management*. Second Edi. Routledge, pp. 763–794.
- Crane-Droesch, A. *et al.* (2013) 'Heterogeneous global crop yield response to biochar: a meta-regression analysis', *Environmental Research Letters*, 8(4), p. 044049. doi: 10.1088/1748-9326/8/4/044049.

CHAPTER 3

Davidson, E. A. and Janssens, I. A. (2006) 'Temperature sensitivity of soil carbon decomposition and feedbacks to climate change', *Nature*, 440(7081), pp. 165–173. doi: 10.1038/nature04514.

Dil, M. and Oelbermann, M. (2014) 'Evaluating the long-term effects of pre-conditioned biochar on soil organic carbon in two southern Ontario soils using the century model.', in Oelbermann, M. (ed.) *Sustainable agroecosystems in climate change mitigation*. The Netherlands: Wageningen Academic Publishers, pp. 251–270. Available at: <https://www.wageningenacademic.com/doi/book/10.3920/978-90-8686-788-2>.

Ding, F. *et al.* (2018) 'A meta-analysis and critical evaluation of influencing factors on soil carbon priming following biochar amendment', *Journal of Soils and Sediments*. *Journal of Soils and Sediments*, 18(4), pp. 1507–1517. doi: 10.1007/s11368-017-1899-6.

El-Naggar, A., Awad, Y. M., *et al.* (2018) 'Biochar influences soil carbon pools and facilitates interactions with soil: A field investigation', *Land Degradation & Development*, 29(7), pp. 2162–2171. doi: 10.1002/ldr.2896.

El-Naggar, A., Lee, S. S., *et al.* (2018) 'Influence of soil properties and feedstocks on biochar potential for carbon mineralization and improvement of infertile soils', *Geoderma*. Elsevier, 332(May), pp. 100–108. doi: 10.1016/j.geoderma.2018.06.017.

El-Naggar, A. *et al.* (2019) 'Biochar composition-dependent impacts on soil nutrient release, carbon mineralization, and potential environmental risk: A review', *Journal of Environmental Management*. Elsevier, 241(September 2018), pp. 458–467. doi: 10.1016/j.jenvman.2019.02.044.

ESRI (2011) *ArcGIS Desktop*. Redlands, CA: Environmental System Research Institute. Available at: <http://desktop.arcgis.com>.

Falloon, P. *et al.* (1998) 'Estimating the Size of the Inert Organic Matter Pool From Total Organic Carbon Content for Use in the Rothamsted Carbon Model', *Soil Biology & Biochemistry*, 30(8), pp. 1207–1211. doi: 10.1016/S0038-0717(97)00256-3.

Falloon, P. *et al.* (2007) 'Climate change and its impact on soil and vegetation carbon storage in Kenya, Jordan, India and Brazil', *Agriculture, Ecosystems and Environment*, 122(1), pp. 114–124. doi: 10.1016/j.agee.2007.01.013.

FAO (2019) *FAOSTAT, Food and Agriculture Organization of the United Nations - Statistic Division*. Available at: <http://www.fao.org/faostat/en/#data/QC> (Accessed: 21 October 2019).

Foereid, B., Lehmann, J. and Major, J. (2011) 'Modeling black carbon degradation and movement in soil', *Plant and Soil*, 345(1), pp. 223–236. doi: 10.1007/s11104-011-0773-3.

Food and Agriculture Organization of the United Nations (2014) *World Reference Base for Soil Resources 2014: International soil classification systems for naming soils and creating legends for soil maps (Update 2015)*, *World Soil Resources Reports No. 106*. Available at: <http://www.fao.org/soils-portal/soil-survey/soil-classification/world-reference-base/en/>.

Glaser, B. and Lehr, V.-I. (2019) 'Biochar effects on phosphorus availability in agricultural soils: A meta-analysis', *Scientific Reports*, 9(1), p. 9338. doi: 10.1038/s41598-019-45693-z.

Gonzaga, L. C. *et al.* (2019) 'Implications of Sugarcane Straw Removal for Soil Greenhouse Gas Emissions in São Paulo State, Brazil', *BioEnergy Research*. *BioEnergy Research*, 12(4), pp. 843–857. doi: 10.1007/s12155-019-10006-9.

Haefele, S. M. *et al.* (2011) 'Effects and fate of biochar from rice residues in rice-based systems', *Field Crops Research*. Elsevier B.V., 121(3), pp. 430–440. doi: 10.1016/j.fcr.2011.01.014.

Hagemann, N. *et al.* (2017) 'Organic coating on biochar explains its nutrient retention and stimulation of soil fertility', *Nature Communications*. Springer US, 8(1), p. 1089. doi: 10.1038/s41467-017-01123-0.

Hartemink, A. E. and McSweeney, K. (2014) *Soil Carbon*. Progress i. Edited by A. E. Hartemink and K. McSweeney. Cham: Springer International Publishing. Available at: <https://www.springer.com/gp/book/9783319040837>.

Hassuani, S. J., Leal, M. R. L. V. and Macedo, I. de C. (2005) *Biomass Power Generation - Sugar cane bagasse and trash*. First. Edited by S. J. Hassuani, M. R. L. V. Leal, and I. de C. Macedo. Piracicaba Brasil: PNUD - Programa das Nações Unidas para o

CHAPTER 3

Desenvolvimento And CTC - Centro de Tecnologia Canavieira.

IBGE (2019) *Censo Agropecuario - 2017*. Rio de Janeiro. Available at: https://biblioteca.ibge.gov.br/visualizacao/periodicos/3096/agro_2017_resultados_definitivos.pdf.

IPCC (2019) 'Appendix 4: Method for Estimating the Change in Mineral Soil Organic Carbon Stocks from Biochar Amendments', in *2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories*, pp. 2–6.

Jeffery, S. *et al.* (2011) 'A quantitative review of the effects of biochar application to soils on crop productivity using meta-analysis', *Agriculture, Ecosystems & Environment*. Elsevier B.V., 144(1), pp. 175–187. doi: 10.1016/j.agee.2011.08.015.

Jeffery, S. *et al.* (2016) 'Biochar effects on methane emissions from soils: A meta-analysis', *Soil Biology and Biochemistry*. Elsevier Ltd, 101, pp. 251–258. doi: 10.1016/j.soilbio.2016.07.021.

Jeffery, S. *et al.* (2017) 'Biochar boosts tropical but not temperate crop yields', *Environmental Research Letters*, 12(5), p. 053001. doi: 10.1088/1748-9326/aa67bd.

Ji, C. *et al.* (2018) 'Variation in Soil Methane Release or Uptake Responses to Biochar Amendment: A Separate Meta-analysis', *Ecosystems*. Springer US, 21(8), pp. 1692–1705. doi: 10.1007/s10021-018-0248-y.

Kambo, H. S. and Dutta, A. (2015) 'A comparative review of biochar and hydrochar in terms of production, physico-chemical properties and applications', *Renewable and Sustainable Energy Reviews*. Elsevier, 45, pp. 359–378. doi: 10.1016/j.rser.2015.01.050.

Khatriwada, D. *et al.* (2016) 'Optimizing ethanol and bioelectricity production in sugarcane biorefineries in Brazil', *Renewable Energy*. Elsevier Ltd, 85, pp. 371–386. doi: 10.1016/j.renene.2015.06.009.

Kögel-Knabner, I. *et al.* (2010) 'Biogeochemistry of paddy soils', *Geoderma*. Elsevier B.V., 157(1–2), pp. 1–14. doi: 10.1016/j.geoderma.2010.03.009.

Leng, L. *et al.* (2019) 'Biochar stability assessment by incubation and modelling: Methods, drawbacks and recommendations', *Science of the Total Environment*. Elsevier

B.V., 664, pp. 11–23. doi: 10.1016/j.scitotenv.2019.01.298.

Li, J. and Heap, A. D. (2008) *A Review of Spatial Interpolation Methods for Environmental Scientists*, Geoscience Australia. Canberra. Available at: <https://data.gov.au/data/dataset/a-review-of-spatial-interpolation-methods-for-environmental-scientists>.

Li, Y. *et al.* (2018) ‘Research on biochar via a comprehensive scientometric approach’, *RSC Advances*. Royal Society of Chemistry, 8(50), pp. 28700–28709. doi: 10.1039/c8ra05689g.

Liu, X. *et al.* (2013) ‘Biochar’s effect on crop productivity and the dependence on experimental conditions-a meta-analysis of literature data’, *Plant and Soil*, 373(1–2), pp. 583–594. doi: 10.1007/s11104-013-1806-x.

Liu, X. *et al.* (2019) ‘Impact of biochar application on yield-scaled greenhouse gas intensity: A meta-analysis’, *Science of the Total Environment*. Elsevier B.V., 656(127), pp. 969–976. doi: 10.1016/j.scitotenv.2018.11.396.

Liu, Y. *et al.* (2020) ‘Successive straw biochar amendments reduce nitrous oxide emissions but do not improve the net ecosystem economic benefit in an alkaline sandy loam under a wheat–maize cropping system’, *Land Degradation & Development*, 31(7), pp. 868–883. doi: 10.1002/ldr.3495.

Lychuk, T. E. *et al.* (2014) ‘Biochar as a global change adaptation: predicting biochar impacts on crop productivity and soil quality for a tropical soil with the Environmental Policy Integrated Climate (EPIC) model’, *Mitigation and Adaptation Strategies for Global Change*, 20(8), pp. 1437–1458. doi: 10.1007/s11027-014-9554-7.

Maia, S. M. F. *et al.* (2014) ‘Payback time for soil carbon and sugar-cane ethanol’, *Nature Climate Change*, 4(7), pp. 605–609. doi: 10.1038/nclimate2239.

Major, J. *et al.* (2010) ‘Fate of soil-applied black carbon: Downward migration, leaching and soil respiration’, *Global Change Biology*, 16(4), pp. 1366–1379. doi: 10.1111/j.1365-2486.2009.02044.x.

Masiello, C. A. and Druffel, E. R. M. (1998) ‘Black Carbon in Deep-Sea Sediments’,

CHAPTER 3

Science, 280(5371), pp. 1911–1913. doi: 10.1126/science.280.5371.1911.

Mondini, C. *et al.* (2017) ‘Modification of the RothC model to simulate soil C mineralization of exogenous organic matter’, *Biogeosciences*, 14(13), pp. 3253–3274. doi: 10.5194/bg-14-3253-2017.

Obia, A. *et al.* (2016) ‘Vertical and lateral transport of biochar in light-textured tropical soils’, *Soil and Tillage Research*. Elsevier B.V., 165, pp. 34–40. doi: 10.1016/j.still.2016.07.016.

Ottoni, M. V. *et al.* (2018) ‘Hydrophysical Database for Brazilian Soils (HYBRAS) and Pedotransfer Functions for Water Retention’, *Vadose Zone Journal*, 17(1), p. 17. doi: 10.2136/vzj2017.05.0095.

Parton, W. *et al.* (2001) *CENTURY tutorial*. Available at: http://pdf-release.net/external/2357835/pdf-release-dot-net-century_tutorial.pdf.

Peters, J. F., Iribarren, D. and Dufour, J. (2015) ‘Biomass Pyrolysis for Biochar or Energy Applications? A Life Cycle Assessment’, *Environmental Science & Technology*, 49(8), pp. 5195–5202. doi: 10.1021/es5060786.

Popin, G. V. *et al.* (2020) ‘Sugarcane straw management for bioenergy: effects of global warming on greenhouse gas emissions and soil carbon storage’, *Mitigation and Adaptation Strategies for Global Change*. Mitigation and Adaptation Strategies for Global Change, 25(4), pp. 559–577. doi: 10.1007/s11027-019-09880-7.

Romero, E. R. *et al.* (2007) ‘Sugarcane Potential Trash Estimation: Variety and Cane Yield Effect’, in *Int. Soc. Sugar Cane Technol*, pp. 9–13.

Sampaio, I. L. M. *et al.* (2019) ‘Electricity Production from Sugarcane Straw Recovered Through Bale System: Assessment of Retrofit Projects’, *BioEnergy Research*. BioEnergy Research, 12(4), pp. 865–877. doi: 10.1007/s12155-019-10014-9.

Scarpore, F. V. *et al.* (2016) ‘Sugarcane land use and water resources assessment in the expansion area in Brazil’, *Journal of Cleaner Production*, 133(June), pp. 1318–1327. doi: 10.1016/j.jclepro.2016.06.074.

SEEG (2016) *São Paulo - Emissions, System for Greenhouse Gas Emissions and Removal*

Estimates. Available at: <http://plataforma.seeg.eco.br/territories/sao-paulo/card?year=2016> (Accessed: 14 September 2018).

Sierra, C. A., Müller, M. and Trumbore, S. E. (2012) 'Models of soil organic matter decomposition: the SoilR package, version 1.0', *Geoscientific Model Development*, 5(4), pp. 1045–1060. doi: 10.5194/gmd-5-1045-2012.

Silva, A. G. B. *et al.* (2019) 'How Much Sugarcane Straw is Needed for Covering the Soil?', *BioEnergy Research*. *BioEnergy Research*, 12(4), pp. 858–864. doi: 10.1007/s12155-019-10008-7.

Singh, B. P. *et al.* (2015) 'In situ persistence and migration of biochar carbon and its impact on native carbon emission in contrasting soils under managed temperate pastures', *PLoS ONE*, 10(10), pp. 1–20. doi: 10.1371/journal.pone.0141560.

Smith, P. (2016) 'Soil carbon sequestration and biochar as negative emission technologies', *Global Change Biology*, 22(3), pp. 1315–1324. doi: 10.1111/gcb.13178.

Theodor Rudorff, B. F. (2014) *Canasat - INPE*. Available at: <http://www.dsr.inpe.br/laf/canasat/tabelas.html> (Accessed: 27 August 2019).

USDA (1999) *Soil Taxonomy. A Basic System of Soil Classification for Making and Interpreting Soil Surveys.*, USDA. Washington DC. Available at: https://www.nrcs.usda.gov/Internet/FSE_DOCUMENTS/nrcs142p2_051232.pdf.

Ventura, M. *et al.* (2019) 'Biochar mineralization and priming effect in a poplar short rotation coppice from a 3-year field experiment', *Biology and Fertility of Soils*. *Biology and Fertility of Soils*, 55(1), pp. 67–78. doi: 10.1007/s00374-018-1329-y.

Verheijen, F. *et al.* (2009) *Biochar application to soil - A Critical Scientific Review of Effects on Soil Properties, Processes and Functions*. Luxembourg,. Available at: <https://ec.europa.eu/jrc/en/publication/eur-scientific-and-technical-research-reports/biochar-application-soils-critical-scientific-review-effects-soil-properties-processes-and>.

Waldheim, L., Monis, M. and Verde Leal, M. R. (2001) 'Biomass Power Generation: Sugar Cane Bagasse and Trash', in Bridgwater, A. . (ed.) *Progress in Thermochemical*

Biomass Conversion. Oxford, UK: Blackwell Science Ltd, pp. 509–523. Available at: http://www.sucre-ethique.org/IMG/pdf/CTC_energy_-_biomass_1_.pdf.

Wang, J. *et al.* (2016) ‘Biochar stability in soil: Meta-analysis of decomposition and priming effects’, *GCB Bioenergy*, 8(3), pp. 512–523. doi: 10.1111/gcbb.12266.

Wang, Z. *et al.* (2014) ‘Effects of co-produced biochar in life cycle greenhouse gas emissions of pyrolysis-derived renewable fuels’, *Biofuels, Bioproducts and Biorefining*, 8, pp. 189–204. doi: 10.1002/bbb.

Wickham, H. (2016) *ggplot2: Elegant Graphics for Data Analysis*. Springer-V. New York. Available at: <https://ggplot2.tidyverse.org>.

Woolf, D. and Lehmann, J. (2012) ‘Modelling the long-term response to positive and negative priming of soil organic carbon by black carbon’, *Biogeochemistry*, 111(1–3), pp. 83–95. doi: 10.1007/s10533-012-9764-6.

Zimmerman, A. R. (2010) ‘Abiotic and Microbial Oxidation of Laboratory-Produced Black Carbon (Biochar)’, *Environmental science & technology*, 44(4), pp. 1295–1301. doi: 10.1021/es903140c.

Zimmerman, A. R. and Ouyang, L. (2019) ‘Priming of pyrogenic C (biochar) mineralization by dissolved organic matter and vice versa’, *Soil Biology and Biochemistry*. Elsevier, 130(November 2018), pp. 105–112. doi: 10.1016/j.soilbio.2018.12.011.

3.6 Acknowledgments

We acknowledge funding through the SOILS-R-GGREAT (NE/P019498/1) project of the greenhouse gas removal (GGR) program. The GGR program is financed by the UK Natural Environment Research Council (NERC), Engineering and Physical Science Research Council (EPSRC), Economic and Social Science Research Council (ESRC) and the UK department for Business, Energy and Industrial Strategy (BEIS).

CHAPTER 4

Biochar LCA

“truth [...] is much too complicated to allow anything but approximation”

JOHN VON NEUMANN

AN ANTICIPATORY LIFE CYCLE ASSESSMENT OF THE USE OF BIOCHAR FROM SUGARCANE RESIDUES AS A GREENHOUSE GAS REMOVAL TECHNOLOGY

David Lefebvre^a, Adrian Williams^a, Guy J.D. Kirk^a, Jeroen Meersmans^b, Saran Sohi^c, Pietro Goglio^{a,d}, and Pete Smith^e

^a School of Water, Energy and Environment, Cranfield University, College Road, Bedford, MK43 0AL, UK

^b TERRA Teaching and Research Centre, Gembloux Agro-Bio Tech, University of Liège, Gembloux 5030, Belgium

^c UK Biochar Research Centre (UKBRC), School of GeoSciences, Crew Building, University of Edinburgh, EH9 3FF, United Kingdom

^d Wageningen Economic Research, Wageningen University & Research, Atlas building, Droevendaalsesteeg 4, 6708 PB Wageningen, Netherlands

^e Institute of Biological and Environmental Sciences, University of Aberdeen, 23 St Machar Drive, Aberdeen AB24 3UU, UK

4.1 Abstract

Greenhouse gas removal technologies are needed to reach the targets of the UNFCCC Paris Agreement. Among existing technologies, the use of biochar is considered promising, particularly biochar derived from the large quantities of sugarcane residues available in South America and elsewhere. However, the net greenhouse gas removal potential of sugarcane biochar has not been assessed hitherto. We use a scenario-based anticipatory life cycle assessment to investigate the emissions associated with a change from the combustion of sugarcane residues in a combined heat and power plant to the pyrolysis of these residues for biochar production and field application in São Paulo State, Brazil. We define scenarios based on different mean marginal electricity production and biochar production share. The results indicate that emissions from covering the electricity deficit generated by partial combustion of biomass during biochar production is the main emitting process. Overall, the processes associated with biochar production lower the net greenhouse gas benefits of the biochar by around 25%. Our analysis suggests that allocating 100% of the available sugarcane residues to biochar production could sequester $6.3 \pm 0.5 \text{ t CO}_2\text{e ha}^{-1} \text{ yr}^{-1}$ of sugarcane in São Paulo State. Scaled up to the entire State, the practice could lead to the removal of 23% of the total amount of GHGs emitted by the State in 2016.

4.2 Introduction

A global decrease of greenhouse gas (GHG) emissions of around 6% yearly is needed to reach the targets of the UNFCCC Paris Agreement and limit global temperature increases to well below 2°C above pre-industrial levels (IPCC, 2018). Numerous greenhouse gas removal (GGR) technologies have been proposed to achieve this (Woolf *et al.*, 2010; Smith *et al.*, 2013; Haszeldine *et al.*, 2018; Nemet *et al.*, 2018). One of the promising technologies is based on biochar, the carbon-rich material produced in thermochemical conversion of biomass under restricted oxygen supply (Schmidt *et al.*, 2018; IPCC, 2019). Sugarcane (*Saccharum officinarum* L.) cultivated for ethanol as well as human consumption is one of the largest sources of residual biomass globally, with 1.9 Gt of fresh sugarcane harvested in 2018 (FAO, 2019). It is grown in more than 90 countries, but most current production is in Brazil (Cardoso *et al.*, 2019). Within Brazil, 55% of sugarcane is produced in the São Paulo State on more than 5.7 Mha (Theodor Rudorff, 2014). Sugarcane cultivation generates large quantities of residues, particularly with the increasingly widespread practise of “green harvesting” – mechanical harvest of cane without prior burning of leaves (Romero *et al.*, 2007).

The purpose of the current study is to refine a previous scoping of biochar GGR for sugarcane in São Paulo State (Lefebvre *et al.*, 2020) using life cycle assessment (LCA) to allow more accurate and transparent analysis of the full carbon burden (Goglio *et al.*, 2020). Published LCAs for biochar GGR have been site- and system-specific (Matušík, Hnátková and Kočí, 2020) and cannot be extended to the assessment of the high potential sugarcane system. In this study, we i) adjust the carbon (C) sequestration potential of sugarcane residue biochar for emissions associated with biochar production and use, and ii) compare the typical sugarcane processing chain (baseline) with a modified one in which biochar production from sugarcane bagasse and/or straw is integrated. The global warming potential of the alternate processes provides more accurate net greenhouse gas removal figures and, for the first time, an anticipatory LCA (Wender *et al.*, 2014) for biochar production and use in a high potential GGR system.

4.3 Method

4.3.1 Sequestration potential of sugarcane residue biochar in São Paulo

Our recent study assessed the carbon sequestration potential of transforming sugarcane production in São Paulo to include biochar (Lefebvre *et al.*, 2020). Available residues (dry basis) included 10.4 t ha⁻¹ yr⁻¹ of bagasse processing residues and 2.9 t ha⁻¹ yr⁻¹ of recovered straw (considering that 7 t ha⁻¹ yr⁻¹ of straw is reserved for optimal soil protection and agronomic sustainability (Silva *et al.*, 2019)). If 100% of these residues are used for biochar production, we calculated that conventional slow pyrolysis would produce up to 4.2 t of biochar (tBC) (Lefebvre *et al.*, 2020). Applied back to the field, we predicted this biochar would increase soil C stocks by 2.35 ± 0.4 t C ha⁻¹ yr⁻¹; the uncertainty represents the diversity of soil type and climate of the study area (Lefebvre *et al.*, 2020).

4.3.2 Objectives and system boundary

This study applies LCA methodology to assess the difference in net GHG emissions (as carbon dioxide equivalents; CO₂e) between use of available sugarcane residues in providing power (CHP combustion plant) versus producing biochar (in a slow pyrolysis plant) (Figure 4.1). This screening LCA does not take account of emissions associated with processes present in both baseline and biochar scenarios (e.g. emissions arising in common field operations or the emissions associated with sugarcane processing), where the emissions can be considered equivalent. This screening LCA uses 100-year global warming potentials (GWP) as prescribed in the IPCC Fifth Assessment Report (AR5) (IPCC, 2014). The functional units applied in our LCA are (i) per hectare of sugarcane crop for São Paulo state, and (ii) per tonne of CO₂e sequestered.

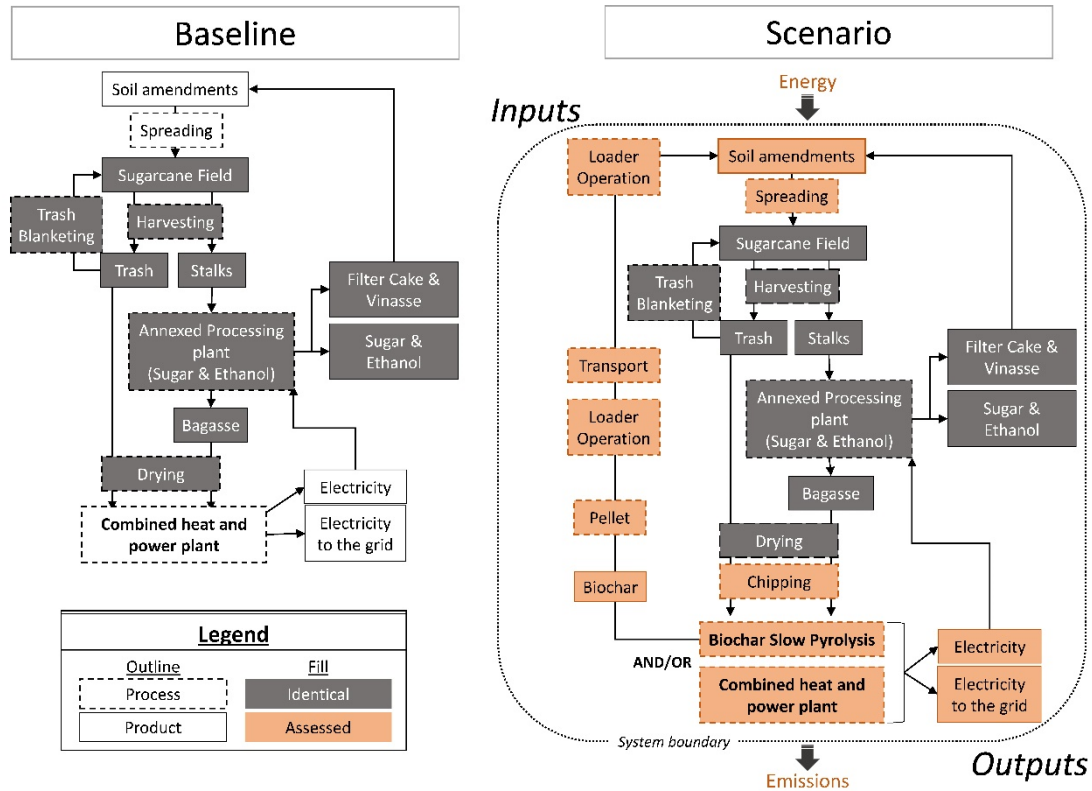


Figure 4.1: Comparison between the baseline system and the assessed scenario. The orange box represents the process or products susceptible to change from the baseline to the biochar producing scenario and, hence, are assessed for their carbon footprint.

4.3.3 Software, database & data processing

SimaPro 9 database (EcoInvent 3.6) were used to assess the emission factors of processes and fuels (PRé Consultants: Life Cycle consultancy and software solutions, 2019). The IPCC 2013 GWP 100a V1.03 method was used to assess the SimaPro processes' carbon footprint (Myhre *et al.*, 2013). Publicly available data were used when processes could not be found in the databases, when they better represented the process under study, or to provide a range of values used for the uncertainty analysis. The data and emission factors (EF) used in this study are available in the Supplementary Information - Section C.3. R software version (3.5.1) (Core Team, 2018) was used to compute the life cycle impact assessment and the uncertainty analysis. The R package ggplot2 (Wickham, 2016) was used to produce the figures.

4.3.4 Scenario description

Biochar contains a large proportion of the energy content of the original biomass (Bergman *et al.*, 2015), so pyrolysis logically produces less electricity than combustion in CHP. Our data collection found that, on average, the electricity generated by the CHP plant delivers 950 kWh t⁻¹ residue (electric) but only 114 kWh t⁻¹ residue for pyrolysis (see Supplementary Information – Section C.3). As a result, Brazil will need to cover the resulting deficit using other electricity sources that would require additional infrastructure. The national energy mix will consequently change by the increased input into the grid from the marginal supply, and hence lead to a new carbon footprint per unit of electricity produced nationwide. Therefore, the LCA of biochar also needs to encompass (i) the carbon footprint of the required additional infrastructure to allow for the production of this energy deficit, and (ii) the carbon footprint of the updated country energy mix to cover this electricity deficit.

4.3.4.1 Updating the national energy mix

To update the Brazilian energy mix we first approximated the additional energy needed at national scale, considering a change from CHP to BC production. The associated electricity deficit is 150 kWh from the residues associated with one ton of harvested sugarcane. Sugarcane yields on average 74 t ha⁻¹ (CONAB, 2018). In Brazil, there are 10,189,208 ha dedicated to sugarcane production (FAO, 2019). Therefore, supposing that available residues from all sugarcane field are used in a CHP, the national electricity deficit of changing from CHP to BC would be 113 TWh.

Table 4.1 shows the national electricity mix (in TWh) with their respective EF. By allocating the 113.2 TWh to the generation appropriate to the scenario (see section 4.3.4), as well as by adjusting the electrical output produced by the combustion of sugarcane biomass (“Biomass (Sugarcane)”) to 0 – now used for biochar production, we can define a new GWP (in kgCO₂e kWh⁻¹) for the updated country energy mix (Table 4.2).

Table 4.1. Brazilian energy mix in 2016 and predicted for 2021 (Barros, Piekarski and De Francisco, 2018) and emission factors (EF) (Wernet et al., 2016).

	2016	2021	EF
Unit	TWh	TWh	kgCO ₂ e kWh ⁻¹
Hydro	380	517	0.072
Wind	33	75	0.015
Solar	0	11	0.077
Biomass (Sugarcane)	38	51	0.029
Biomass (other)	11	15	0.057
Nuclear	16	15	0.011
Natural Gas	45	20	0.555
Hard Coal	16	10	1.223
Oil	38	34	0.725
TOTAL	579	749	/

4.3.4.2 Consequential electricity scenarios

The different scenarios of this anticipatory LCA are based on assumptions of the marginal energy supply of the country. The marginal energy production technology is the technology most likely to respond to changing demand (Lund *et al.*, 2010). Among the energy sources in Brazil, natural gas (NG – Scenario 1), wind (Scenario 2), and a mixture of both (75% NG, 25% wind – Scenario 3) were selected as being the most probable marginal energy in a near future (Seabra *et al.*, 2011; Khatiwada *et al.*, 2016; EPE, 2017; Santos *et al.*, 2017; Panorama Offshore, 2018; de Oliveira Noronha, Zanini and Souza, 2019; Diógenes, Claro and Rodrigues, 2019). An additional energy mix scenario (Scenario 4) was based on the recent work from Vandepaer *et al.* (2019) who, among other things, used consequential life cycle inventories to project the marginal electricity mix of Brazil for the year 2040 (Vandepaer *et al.*, 2019). Complete description of the scenarios is available in the Supplementary Information – Section C.1. Table 4.2 reports the updated national energy mixes and emissions factors for the scenarios assessed.

Table 4.2. Summary table of the electricity mixes and emission factors considered for each scenario.

	Unit	Baseline (Barros, Piekarski and De Francisco, 2018)	Scenario 1 (NG)	Scenario 2 (Wind)	Scenario 3 (NG [75%] & Wind [25%])	Scenario 4 (Vandepaer <i>et al.</i> , 2019)
Year	/	2021	2021	2021	2021	2040
Hydro	%	69	63.8	63.8	63.8	17.3
Wind	%	10	9.3	23.2	12.7	4.92
Solar	%	1.5	1.4	1.4	1.4	64.24
Sugarcane biomass	%	6.8	0.0	0.0	0.0	1.66*
Other biomass	%	2	1.9	1.9	1.9	
Nuclear	%	2	1.9	1.9	1.9	1.3
Natural gas	%	2.7	16.4	2.5	12.9	10
Hard coal	%	1.3	1.2	1.2	1.2	0
Oil	%	4.5	4.2	4.2	4.2	0.01
Updated GWP	kgCO ₂ e kWh ⁻¹	/	0.187	0.111	0.168	/
High to medium voltage ‡	kgCO ₂ e kWh ⁻¹	/	0.008	0.005	0.008	/
Infrastructure	kgCO ₂ e kWh ⁻¹	/	0.001	0.009	0.003	/
Total GWP	kgCO ₂ e kWh ⁻¹	0.153	0.196	0.125	0.178	0.136

‡ Transformation from high to medium voltage based on losses reported by the Ecoinvent 3.6 database (Wernet *et al.*, 2016; Severinghaus, 2019)

* Vandepaer *et al.* (2019) does not differentiate between sugarcane and other biomass in his electricity mix

4.3.4.3 Biochar scenarios

In addition to the marginal energy scenarios, we set up three sub-scenarios where 100%, 50%, and 25% of the maximum biochar production potential is reached. The 50% biochar production sub-scenario considers that respectively 0% and 60% of the available straw and bagasse, are allocated to biochar production while the 25% biochar production sub-scenario considers that respectively 0% and 30% of the available straw and bagasse are

allocated to biochar production. The residual biomass in the 50% and 25% sub-scenario is considered to be utilised in a CHP.

4.3.5 System description

4.3.5.1 Baseline

In the baseline system, it was assumed that the sugarcane processing plant is an annexed plant producing ethanol, sugar and electricity. This type of processing plant is the most widespread in SP (Cardoso *et al.*, 2019). The available residues are burned in a 65-bar pressure boiler with an efficiency of 88%. The electricity is produced by a CHP generator with condensing-extraction turbines (Cardoso *et al.*, 2019). The residual heat produced by the CHP (or pyrolyzer) was not considered in this analysis because there is function for surplus heat. The facility processes 3 Mt sugarcane during 190 days of operation per season (Cardoso *et al.*, 2019).

4.3.5.1.1 Sugarcane mills' surplus electricity

Sugarcane processing in Brazil use various technologies (Pippo and Luengo, 2013). The amount of electricity exported from the mills to the grid varies accordingly. Low technology facilities import electricity from the grid (Pippo and Luengo, 2013). The average electricity demand of a sugarcane processing site is around 30 kWh t⁻¹ of harvested cane (Seabra and Macedo, 2011; Pippo and Luengo, 2013; Klein *et al.*, 2018; Birru, Erlich and Martin, 2019; Cardoso *et al.*, 2019). In our assessment we assume that all facilities in São Paulo are 'modern' or 'state of the art' mills and export electricity to the grid. Based on the processing size of our baseline scenario in São Paulo (3 Mt sugarcane processed per season) (Cardoso *et al.*, 2019), with an average yield of 74 t ha⁻¹ (CONAB, 2018) and the sugarcane area of 5,768,172 ha (Theodor Rudorff, 2014), the state should have 143 processing sites. This number is consistent with the official number of 160 sites (Conab, 2019), meaning that our case study reflects the average situation for São Paulo state. Taking into account the cane yield and the available sugarcane residues and assuming that the available straw is combusted along with the bagasse, our representative site would generate about 170 kWh t⁻¹ of harvested sugarcane, use 30 kWh internally and export about 140 kWh to the grid (Olivério and Ferreira, 2010; Dias *et al.*, 2011; Seabra and Macedo, 2011; Alves *et al.*, 2015; Klein *et al.*, 2018; Birru, Erlich and Martin, 2019; Cardoso *et al.*, 2019; Sampaio *et al.*, 2019).

4.3.5.2 Biochar producing system

Our scenarios consider biochar to be produced by a conventional industrial slow pyrolyzer at around 550°C. Slow pyrolysis was selected because it favours biochar over other products when compared to other pyrolysis options (Basu, 2010). In our scenarios the bio-oil is not collected but rather combusted in the gas phase (Gabra *et al.*, 2001; Basu, 2010). This approach has been reported in previous studies (Park *et al.*, 2014; Wang, Camps-Arbestain and Hedley, 2014; Roy and Dias, 2017; Azzi, Karlton and Sundberg, 2019). The gas released during pyrolysis is used to generate electricity (Quirk *et al.*, 2012; de Oliveira Vilela *et al.*, 2014; You *et al.*, 2017; Oldfield *et al.*, 2018). In addition to the biochar plant, the biochar producing system also includes grinding of the biomass, the production of biochar pellets, and their transportation and application on the field. Description of each process is available in the Supplementary Information – Section C.2.

4.3.6 Additional effect of biochar application

4.3.6.1 Reduced nitrous oxide emissions from soils

Quirk *et al.* (2012) evaluated the effect of biochar from either sugarcane bagasse or sugarcane straw on the emission of nitrous oxide from soil under sugarcane land in Australia. They reported that “*In the fertilised soil bagasse biochar significantly reduced [by 14.3%] N₂O emissions compared to both control and straw biochar amended soils*” (Quirk *et al.*, 2012, p.324). The emission factor of nitrous oxide is 0.7% of applied N in São Paulo state under sugarcane cultivation when 5 tonnes of straw ha⁻¹ are removed (Gonzaga *et al.*, 2019). This agrees with the range offered by Filoso *et al.* (2015), and the control treatment of Quirk *et al.* (2012). The IPCC Tier 1 guidelines (IPCC, 2006) for direct and indirect N₂O emissions from organic fertilizer (vinasse, filter cake, straw blanket) are 1.22% of their N content. Combining the yearly addition of N containing amendments (i.e. urea, vinasse, filter cake, residue blanket) using the N₂O emissions factor from Gonzaga *et al.* (2019) for the urea and from the IPCC for the rest, the emissions reach 1.46 kg N₂O ha⁻¹ yr⁻¹, or 388 kg CO₂e ha⁻¹ yr⁻¹ (Hallegatte *et al.*, 2016). The findings of Quirk *et al.* (2012) are in the lower range reported by Abbruzzini *et al.* (2017) for sugarcane in São Paulo state and agree with the figure used in Thers *et al.* (2019) study using an application of 1 tBC ha⁻¹ on oilseed rape in Denmark.

4.3.6.2 Biochar induced fertilizer reduction

We considered biochar to increase the nitrogen use efficiency by 11% (Liu *et al.*, 2018) and phosphorus use efficiency by 10% (Borges *et al.*, 2020), independently of the biochar application rate simulated. In addition, we accounted for the liming potential of biochar according to Quirk *et al.* (2012). Fertilizer and lime inputs were reduced based on the increased nutrient use efficiency and liming potential of biochar (Supplementary Information – Section C.3).

4.3.6.3 Accounting for the pyrolysis plant construction material

Yang *et al.* (2016) provide a list of the materials and energy needed to build a pyrolysis plant. Their study accounts for the emissions associated with the workshop and office construction, the emissions to build the equipment (shredding, drying, and pyrolysis system), and the electricity required for the auxiliary systems and domestic consumption. The carbon footprint of the pyrolysis plant and its maintenance reaches $117 \text{ kg CO}_2\text{e tBC}^{-1}$ (using the 2021 Brazilian electricity emission factor (Barros, Piekarski and De Francisco, 2018)). Roberts *et al.* (2010) also reports the amount of material for a pyrolysis plant working at 80% full capacity with a lifetime of 20 yr, processing 10 t h^{-1} of biomass, including onsite storage of the biomass. Assuming 200 days of production, at 7 h day^{-1} , and the averaged 32.5% sugarcane residue BC yield (i.e. 31.3% for bagasse and 33.6% for straw, Supplementary Information, Section C.3), the carbon footprint of constructing the pyrolysis plant reaches $133 \text{ kg CO}_2\text{e tBC}^{-1}$.

4.3.6.4 Non - CO₂ GHG emissions

Data on pyrolysis emissions are scarce, especially considering the post combustion of the syngas for energy production (Pennise *et al.*, 2001; Azzi, Karlun and Sundberg, 2019) and the plant scale considered in this study. We assumed that the industrial pyrolysis plant modelled here releases few to none non-CO₂ GHG emissions. Considering the scale of the simulated plant, if GHG are released from the facility, the emissions would be under strict state regulations. We assume these regulations to be similar to the emission regulations of a CHP plant. Since our scenarios substitute CHP for pyrolysis, in its entirety or in part depending on the scenarios, we assumed that emissions from the pyrolysis plant (if any) would be equivalent to the emissions of a CHP and hence cancel each other.

4.3.7 Uncertainty analysis

The uncertainty analysis was conducted using the range of values obtained through the literature search. A statistical distribution was applied to each range of values from where a random selection of 10,000 values were made for the Monte Carlo simulations. The simulations were made in R software (3.5.1) (Core Team, 2018). The distributions applied were: emission factors and electricity productions/demands – pert, transport distance – uniform, diesel use – lognormal. Additional information on the uncertainty analysis can be found in the Supplementary Information – Section C.4.

4.4 Results

Figure 4.2 shows the contribution analysis of our LCA using NG as the marginal energy and considering 100% of the available residues for BC production. This result shows that the biochar soil application accounts for the majority of its sequestration potential while the reduced nitrous oxide emission from the soil, the electricity production during pyrolysis and the reduced fertilizer need due to biochar application accounts for less than 5%. On the emissions side, covering the electricity deficit is responsible for most of the emissions associated with the practice, while grinding the biomass, pelletizing, transporting, loading, and spreading the biochar accounts for less than 10% of the total emissions of the practice. These observations remain valid regardless of the scenario assessed. The contribution analyses for each of the scenarios are available in the Supplementary Information – Section C.5.

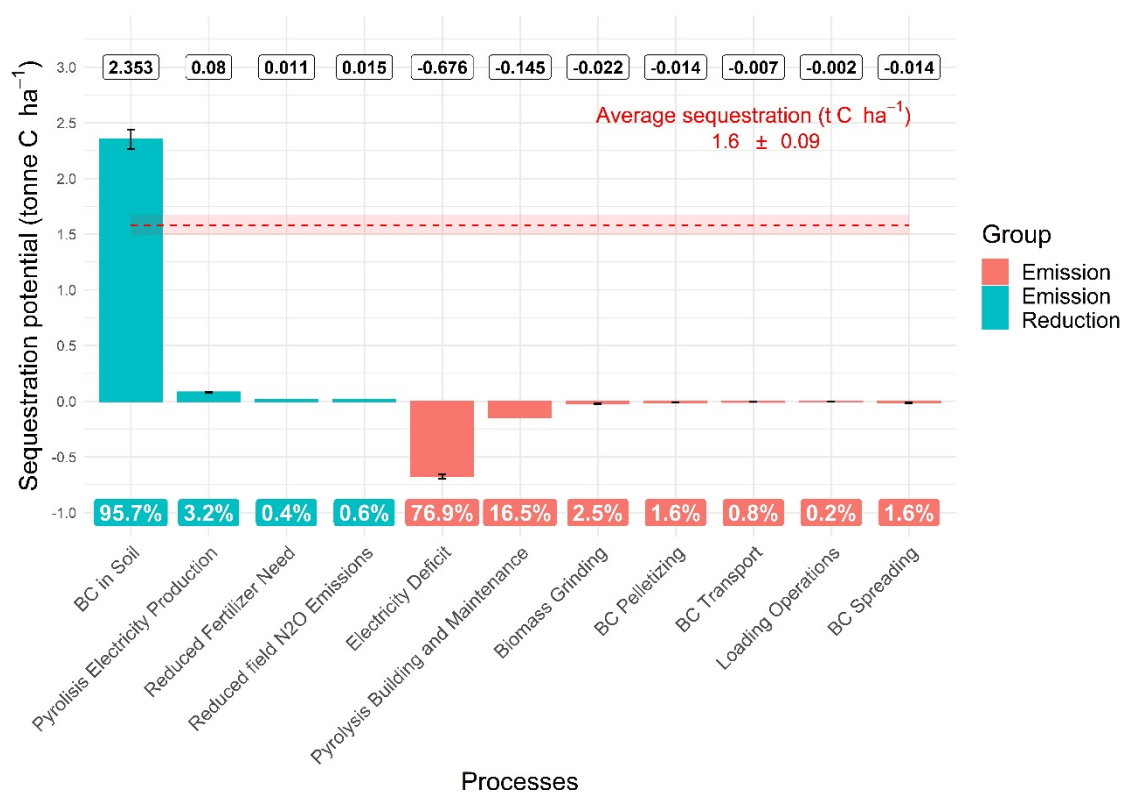


Figure 4.2: Contribution analysis considering NG as marginal energy and 100% allocation of available sugarcane residues to BC production. The upper outlined numbers report the exact figures in t C ha⁻¹ for each process. The under outlined numbers report the share of each process within each group (emission or emission reduction). The ribbon and error bars are ± 1 SD. The dotted line represents the net sequestration value.

Figure 4.3 shows the combined results for all the marginal energy mixes and the three sub-scenarios (biochar production share) assessed. On average, production of 100%, 50% and 25% of the maximum biochar production potential can sequester a net of 6.3 ± 0.5 , 3.6 ± 0.3 and 1.8 ± 0.3 t CO₂e ha⁻¹ (mean ± SD), respectively. In addition, these results show that the carbon sequestration of the practice increases as the EF of the marginal electricity mix decreases.

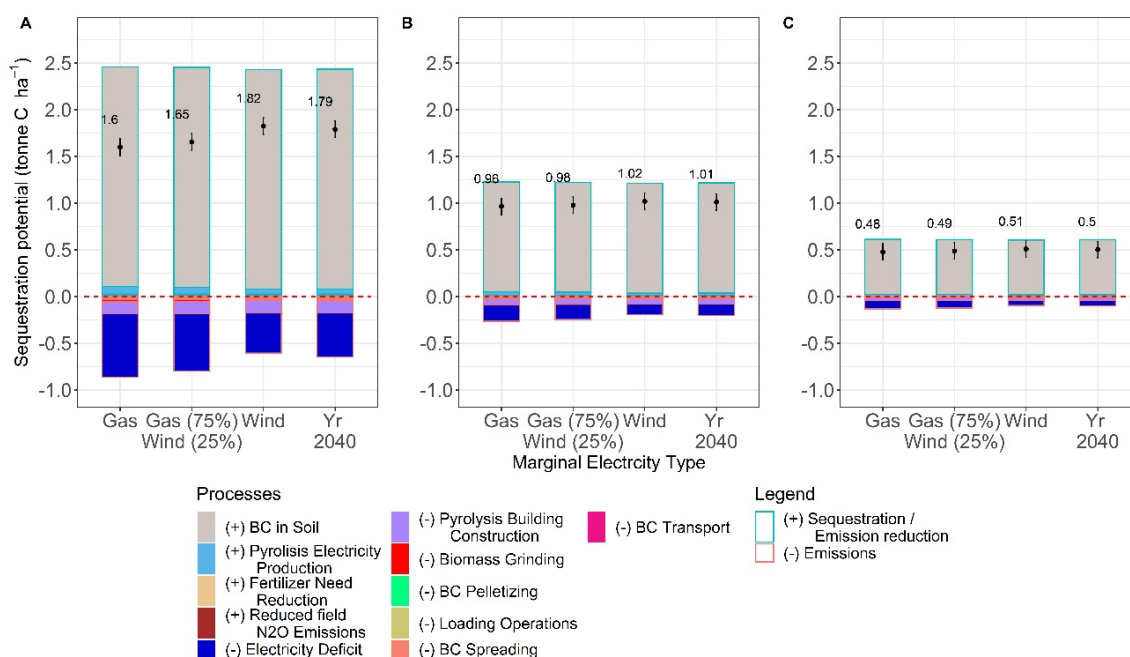


Figure 4.3: Combined results of all scenarios. Section A, B and C represent the production of 100%, 50% and 25% of the maximum biochar production. The remaining residues in section B and C are treated in a CHP similarly to the baseline. The black dots and labels are the net sequestration with associated error bars representing ± 1 SD.

Figure 4.4 shows the remaining fraction of the carbon sequestration potential of one tonne of biochar once we have accounted for the all the additional processes (biochar soil carbon sequestration efficiency). This value is calculated by comparing the actual carbon sequestration potential of biochar addition once accounting for all the processes included in this LCA with the original carbon sequestration potential of biochar alone (i.e. 2.35 t C ha⁻¹, Section 4.3.1). It shows that allocating part of the residues to the CHP plant (section B & C – Figure 4.4) increases the biochar soil carbon sequestration efficiency (residual sequestration potential of BC in soil after subtracting the emissions).

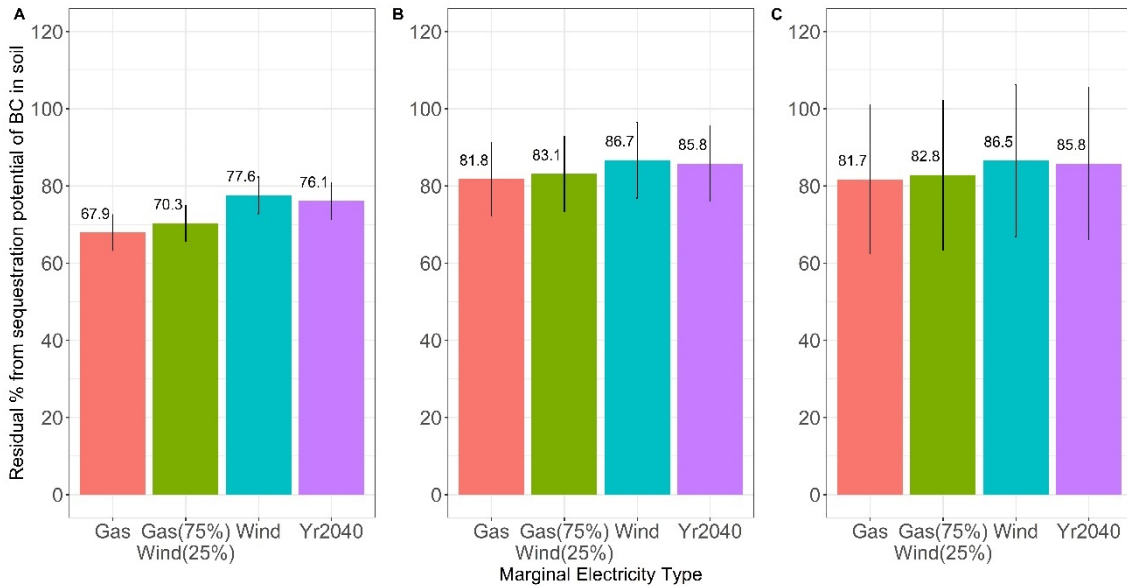


Figure 4.4: Biochar soil carbon sequestration efficiency: the remaining carbon capture potential of biochar in soils of after accounting for all the processes, in percent. Section A, B and C represent the production of 100%, 50% and 25% of the maximum biochar production. The remaining residues in section B and C are treated in a CHP similarly to the baseline. The error bars are ± 1 SD.

4.5 Discussion

4.5.1 Biochar carbon sequestration

Considering the averaged value of all marginal electricity mixes and BC production share, our LCA suggests that each tonne of BC that might be produced from sugarcane residues in São Paulo and applied back to fields should sequester 1.64 ± 0.11 t CO₂e tBC⁻¹. Comparisons with other biochar systems is complicated, because their abatement potentials are highly site- and scenario- specific (Matušík, Hnátková and Kočí, 2020). Our estimate is lower than that from the LCA of Hamedani *et al.* (2019) for willow wood biochar on Belgian willow plantation (~ 2 t CO₂e tBC⁻¹) and slightly higher than that of Thers *et al.* (2019) for biochar from Danish oilseed rape (~ 1.44 t CO₂e tBC⁻¹). Peters *et al.* (2015) explored the potential carbon benefit of producing and applying biochar produced from chipped poplar wood on Spanish poplar plantation, where the heat produced during pyrolysis was used as substitute for natural gas, with direct burning of the woodchip for heat as the reference scenario. They calculated a carbon abatement potential for their biochar scenario that is more than twice our average result (~ 3.9 t CO₂e

tBC⁻¹). Brassard *et al.* (2018) found on average a carbon sequestration potential of almost half that using switchgrass as feedstock and applying the biochar to Canadian wheat (~2.3 t CO₂e tBC⁻¹). Our results reflect our choice of biochar produced at around 550°C. This could change with factors including soil management, the biomass streams utilised, how biochar is integrated into the cropping system, and the national electricity mix.

On average, allocating 100% of the available residues to biochar production led to emissions of 0.30 ± 0.07 t CO₂e per t CO₂e sequestered. This is reduced to 0.19 ± 0.05 and 0.19 ± 0.05 t CO₂e per t CO₂e sequestered when 50% and 25% of the maximum potential biochar production is reached (see Supplementary Information, Section C.5 for complete data). This is roughly twice the 0.135 t CO₂e emitted per t CO₂e sequestered for carbonation and four times the 0.075 t CO₂e emitted per t CO₂e sequestered for enhanced weathering of basalt rock in São Paulo State (Lefebvre *et al.*, 2019). Applied to the whole State, allocating 100% of the sugarcane residues to biochar production could lead to the sequestration of 36 Mt CO₂e yr⁻¹ or 23% of the 159 Mt CO₂e emitted by the State in 2016 (SEEG, 2016). Producing 50% and 25% of the maximum biochar production potential could lead to the sequestration of 21 and 10 Mt CO₂e yr⁻¹, respectively.

4.5.2 The marginal electricity mix

Determining the impact of energy use is a key component of LCA (Vélez-Henao and Garcia-Mazo, 2019). Our results show that the “greener” the energy mix of Brazil, the greater is the carbon sequestration benefit arising from biochar deployment. However, predicting future demand for electricity and the future energy mix in the face of increasing demand is highly uncertain (Vandepaer *et al.*, 2019).

Hydropower is currently the main source of electrical power in Brazil. Some argue that as the number of potential new dam sites is limited, hydropower’s share of the mix will not substantially increase (Dale *et al.*, 2013; Leal, Rego and de Oliveira Ribeiro, 2019). Others see 70 GW of untapped potential installed by 2035 (IEA, 2013), centred in the Amazon. Hydro projects require planning and consultation, and risk damage to local communities, biodiversity and the wider environment, potentially hampering their development (IEA, 2013; Santos *et al.*, 2017). In addition, massive investment would be required in the transmission grid from the Amazon region to the southeast region where electricity consumption is greatest. In our scenarios, the electricity used to offset

transition from biomass combustion to partial combustion for biochar has a major impact on the results, accounting for at least 50% of emissions across all scenarios assessed. If the anticipated increase in renewable energy in the Brazil energy mix (EPE, 2017; de Oliveira Noronha, Zanini and Souza, 2019) was realised, this would slightly alter our results. Nevertheless, as the EF of Brazilian electricity decreases, the net carbon sequestration potential of biochar will increase. Conversely, as the EF of electricity increases, the carbon sequestration decreases. Therefore, sugarcane producing countries mainly relying on coal for their electricity production will benefit less or even emit more CO₂e from such practice than countries with greener energy mixes. Under Brazilian conditions, the net zero carbon sequestration benefit of the practice is reached when the emission factor of electricity reaches 0.687 kg CO₂e kWh⁻¹; e.g. with an energy mix of 12% coal and 88% natural gas. Finally, seasonal and daily variations of energy supply and demand also need to be taken into account. We expect the wind powered scenario to require a greater installed capacity to cover the electricity deficit than the more versatile NG scenario. However, this difference is likely to be small.

4.5.3 Selecting the residue

The following section explores the results of our analysis if only the available bagasse or straw is used to produce biochar while the other residue is combusted in the CHP. For instance, the allocation of all the available straw (i.e. 2.85 t dry matter ha⁻¹) to biochar production and the combustion of the bagasse (i.e. 10.4 t dry matter ha⁻¹) in a CHP leads to an average (including all electricity mixes scenarios - Table 4.2) net sequestration potential of 0.57 t CO₂e ha⁻¹ (or 0.199 t CO₂e t of straw residue⁻¹) and a biochar soil carbon sequestration efficiency of 41%. Conversely, allocating all the available bagasse to biochar production and incinerating the available straw in a CHP leads to an average net sequestration potential of 6.11 t CO₂e ha⁻¹ (or 0.590 t CO₂e t of bagasse residue⁻¹) and a biochar soil carbon sequestration efficiency of 84%. Although sugarcane straw and bagasse have an equivalent low heating value (15.25 MJ kg⁻¹ and 15.13 MJ kg⁻¹ dry weigh, respectively), processing the bagasse requires drying it from 50% moisture content by weight, lowering the overall energy production; whereas the straw usually has a moisture content of 15% by weight when collected from the field (Dias *et al.*, 2011). Data collection on the energy production of straw and bagasse support this observation as

sugarcane straw produces slightly more electricity when treated in a CHP than bagasse (Supplementary Information – Section C.3). In addition, bagasse biochar has a higher carbon content than straw biochar (i.e. 1 kg of bagasse or straw produces 0.20 kg BC-C and 0.14 kg BC-C, respectively; Cross and Sohi, 2013; Quirk *et al.*, 2012), leading to a lower loss of carbon sequestration potential. Further investigations of the properties of sugarcane bagasse and straw biochar when produced in the modelled pyrolyzer, as well as on the electricity production of both biomass types, are needed before these findings can be generalized. In addition, the combustion of straw can be problematic if used in combination with some gas turbines without proper filtration systems, because straw's high silica content can lead to corrosion of the turbine blade and abrasion of the tubing while its high chlorides content can create fouling in the boilers (Gabra *et al.*, 2001; Olivério and Ferreira, 2010; Alves *et al.*, 2015; Anukam *et al.*, 2016). These observations show that the physicochemical properties of the biomass should be taken into account when selecting the most appropriate processing method either to minimize the production of elements reducing the overall efficacy of turbines and/or boilers, or to maximise biochar carbon content production by selecting a carbon rich feedstock.

4.5.4 Additional effects

Decreased albedo induced by biochar application to crop fields (Williamson, 2016; Zhang *et al.*, 2018) is thought to reduce its carbon abatement potential by up to 20% (at application rate of 30 tBC ha⁻¹) (Meyer *et al.*, 2012), as a result of increased soil temperature promoting oxidation of other soil carbon pools (EASAC, 2018). We did not account for such effect in this study because sugarcane is a perennial and fast-growing species (Scarpore *et al.*, 2016) covering the soil rapidly, thereby reducing the effect on albedo.

Likewise, although there is evidence of positive effects of BC on yield (Ye *et al.*, 2020), this study does not adjust sugarcane yield following biochar application. Yield response to biochar depends on biochar characteristics, soil and crop type, co-amendment and other factors (Ye *et al.*, 2020). We know of no published field studies on the effects of sugarcane biochar on sugarcane yield in São Paulo. Doses of 5 t ha⁻¹ and 10 t ha⁻¹ biochar from sugarcane straw had no impact on sugarcane yield in Indonesia (Hariyono *et al.*, 2020).

CHAPTER 4

Recent and future innovations in biomass pyrolysis could increase the LCA benefits of a biochar scenario; for example, alkali metal (K and Na) additions have been shown to increase biochar yield by around 15% as well as increasing the stability of biochar carbon in soil (Mašek *et al.*, 2019). Integration of biochar with fertiliser management has potential for synergistic effects on carbon abatement. Recent laboratory and glasshouse studies have shown the potential to integrate K and P into biochar as an effective compound product. Treatment of sugarcane-straw biochar with KOH followed by neutralisation with H₃PO₄ increased the P content of the biochar and led to higher sugarcane yield and P use efficiency (Borges *et al.*, 2020).

4.6 Conclusions

Changing the processing chain for sugarcane residues from the current typical CHP system to a system based on slow pyrolysis with biochar production could provide net carbon abatement of $6.3 \pm 0.5 \text{ t CO}_2\text{e ha}^{-1}$ or $1.64 \pm 0.11 \text{ t CO}_2\text{e t biochar}^{-1}$, taking into account the emissions associated with set up and running. The gain arises mainly from the C stored as biochar in the soil; losses are mostly due to the electricity deficit caused by the partial combustion of biomass during pyrolysis. Applied to São Paulo State, transformation of the available sugarcane residue to biochar could lead to the sequestration of 36 Mt CO₂e yr⁻¹ or 23% of the State's GHG emissions in 2016.

4.7 Acknowledgments

We thank Dr Ondřej Mašek from the University of Edinburgh and Dr Bernardo M.M.N. Borges from the University of São Paulo for their insights and advices on pyrolysis technologies and sugarcane management. We acknowledge funding through the UP-Green-LCA (NE/P019668/1) and Soils-R-GGREAT (NE/P019498/1) projects of the greenhouse gas removal (GGR) programme. The GGR programme is financed by the UK Natural Environment Research Council (NERC), Engineering and Physical Science Research Council (EPSRC), Economic and Social Science Research Council (ESRC) and the UK department for Business, Energy and Industrial Strategy (BEIS).

4.8 References

Abbruzzini, T. F. *et al.* (2017) 'Effects of Biochar on the Emissions of Greenhouse Gases from Sugarcane Residues Applied to Soils', *Agricultural Sciences*, 08(09), pp. 869–886.

doi: 10.4236/as.2017.89064.

Alves, M. *et al.* (2015) ‘Surplus electricity production in sugarcane mills using residual bagasse and straw as fuel’, *Energy*. Elsevier Ltd, 91, pp. 751–757. doi: 10.1016/j.energy.2015.08.101.

Anukam, A. *et al.* (2016) ‘Pre-processing of sugarcane bagasse for gasification in a downdraft biomass gasifier system: A comprehensive review’, *Renewable and Sustainable Energy Reviews*, 66, pp. 775–801. doi: 10.1016/j.rser.2016.08.046.

Azzi, E. S., Karlton, E. and Sundberg, C. (2019) ‘Prospective Life Cycle Assessment of Large-Scale Biochar Production and Use for Negative Emissions in Stockholm’, *Environmental Science & Technology*. American Chemical Society, 53(14), pp. 8466–8476. doi: 10.1021/acs.est.9b01615.

Barros, M. V., Piekarski, C. M. and De Francisco, A. C. (2018) ‘Carbon footprint of electricity generation in Brazil: An analysis of the 2016-2026 period’, *Energies*, 11(6). doi: 10.3390/en11061412.

Basu, P. (2010) *Biomass Gasification and Pyrolysis*. Elsevier. doi: 10.1016/C2009-0-20099-7.

Bergman, R. D. *et al.* (2015) ‘Life Cycle Analysis of Biochar’, in *Biochar: A Regional Supply Chain Approach in View of Climate Change Mitigation*. Cambridge University Press, pp. 46–69. doi: 10.1017/9781316337974.004.

Birru, E., Erlich, C. and Martin, A. (2019) ‘Energy performance comparisons and enhancements in the sugar cane industry’, *Biomass Conversion and Biorefinery*. Biomass Conversion and Biorefinery, 9(2), pp. 267–282. doi: 10.1007/s13399-018-0349-z.

Borges, B. M. M. N. *et al.* (2020) ‘Re-use of sugarcane residue as a novel biochar fertiliser - Increased phosphorus use efficiency and plant yield’, *Journal of Cleaner Production*. Elsevier Ltd, 262, p. 121406. doi: 10.1016/j.jclepro.2020.121406.

Brassard, P. *et al.* (2018) ‘Pyrolysis of switchgrass in an auger reactor for biochar production: A greenhouse gas and energy impacts assessment’, *Biomass and Bioenergy*. Elsevier Ltd, 116(June), pp. 99–105. doi: 10.1016/j.biombioe.2018.06.007.

CHAPTER 4

Cardoso, T. F. *et al.* (2019) 'A regional approach to determine economic, environmental and social impacts of different sugarcane production systems in Brazil', *Biomass and Bioenergy*. Elsevier Ltd, 120(April 2018), pp. 9–20. doi: 10.1016/j.biombioe.2018.10.018.

Conab (2019) *Perfil do setor do açúcar e etanol no Brasil - Edição para a safra 2015/16*. Brasília. Available at: <https://www.conab.gov.br/info-agro/safras/cana/perfil-do-setor-sucroalcooleiro>.

CONAB (2018) *Acompanhamento da Safra Brasileira - Cana-de-açúcar - Terceiro levantamento | Dezembro 2018*. Available at: <https://www.conab.gov.br/info-agro/safras/cana>.

Core Team, R. (2018) *R: A language and environment for statistical computing. R Foundation for Statistical Computing*. Vienna. Available at: <https://www.r-project.org/>.

Cross, A. and Sohi, S. P. (2013) 'A method for screening the relative long-term stability of biochar', *GCB Bioenergy*, 5(2), pp. 215–220. doi: 10.1111/gcbb.12035.

Dale, A. T. *et al.* (2013) 'Modeling future life-cycle greenhouse gas emissions and environmental impacts of electricity supplies in Brazil', *Energies*, 6(7), pp. 3182–3208. doi: 10.3390/en6073182.

Dias, M. O. S. *et al.* (2011) 'Second generation ethanol in Brazil: Can it compete with electricity production?', *Bioresource Technology*. Elsevier Ltd, 102(19), pp. 8964–8971. doi: 10.1016/j.biortech.2011.06.098.

Diógenes, J. R. F., Claro, J. and Rodrigues, J. C. (2019) 'Barriers to onshore wind farm implementation in Brazil', *Energy Policy*. Elsevier Ltd, 128(December 2018), pp. 253–266. doi: 10.1016/j.enpol.2018.12.062.

EASAC (2018) *Negative emission technologies: What role in meeting Paris Agreement targets?* Halle - Germany. Available at: <https://easac.eu>.

EPE (2017) *The Ten-Year Expansion Plan 2027*. Rio de Janeiro. Available at: http://www.epe.gov.br/sites-en/publicacoes-dados-abertos/publicacoes/PublicacoesArquivos/publicacao-195/PDE_2027_Executive_Summary.pdf.

FAO (2019) *FAOSTAT, Food and Agriculture Organization of the United Nations - Statistic Division*. Available at: <http://www.fao.org/faostat/en/#data/QC> (Accessed: 21 October 2019).

Filoso, S. *et al.* (2015) ‘Reassessing the environmental impacts of sugarcane ethanol production in Brazil to help meet sustainability goals’, *Renewable and Sustainable Energy Reviews*. Elsevier Ltd, 52, pp. 1847–1856. doi: 10.1016/j.rser.2015.08.012.

Gabra, M. *et al.* (2001) ‘Evaluation of cyclone gasifier performance for gasification of sugar cane residue - Part 2: Gasification of cane trash’, *Biomass and Bioenergy*, 21(5), pp. 371–380. doi: 10.1016/S0961-9534(01)00044-7.

Goglio, P. *et al.* (2020) ‘Advances and challenges of life cycle assessment (LCA) of greenhouse gas removal technologies to fight climate changes’, *Journal of Cleaner Production*. Elsevier Ltd, 244, p. 118896. doi: 10.1016/j.jclepro.2019.118896.

Gonzaga, L. C. *et al.* (2019) ‘Implications of Sugarcane Straw Removal for Soil Greenhouse Gas Emissions in São Paulo State, Brazil’, *BioEnergy Research*. BioEnergy Research, 12(4), pp. 843–857. doi: 10.1007/s12155-019-10006-9.

Hallegatte, S. *et al.* (2016) ‘Mapping the climate change challenge’, *Nature Climate Change*. Nature Publishing Group, 6(7), pp. 663–668. doi: 10.1038/nclimate3057.

Hariyono, B. *et al.* (2020) ‘Utilization of the trash biochar and waste of sugarcane to improve the quality of sandy soil and growth of sugarcane’, *IOP Conference Series: Earth and Environmental Science*, 418(1), p. 012067. doi: 10.1088/1755-1315/418/1/012067.

Haszeldine, R. S. *et al.* (2018) ‘Negative emissions technologies and carbon capture and storage to achieve the Paris Agreement commitments’, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 376(2119). doi: 10.1098/rsta.2016.0447.

IEA (2013) *World Energy Outlook - 2013, Economic Outlook*. Paris. doi: 10.1111/1468-0319.00088.

IPCC (2006) ‘Volume 4: Agriculture, forestry and other land use (AFOLU). Chapter 5: Cropland’, *IPCC Guidelines for National Greenhouse Gas Inventories*, p. 66.

CHAPTER 4

IPCC (2014) *Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Edited by T. F. Stocker et al. Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press. Available at: <https://www.ipcc.ch/report/ar5/wg1/>.

IPCC (2018) *Global Warming of 1.5°C - Special Report - Summary for Policymakers*. Edited by Intergovernmental Panel on Climate Change. Available at: https://www.ipcc.ch/site/assets/uploads/sites/2/2018/07/SR15_SPM_High_Res.pdf (Accessed: 2 January 2019).

IPCC (2019) 'Appendix 4: Method for Estimating the Change in Mineral Soil Organic Carbon Stocks from Biochar Amendments', in *2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories*, pp. 2–6.

Khatiwada, D. *et al.* (2016) 'Optimizing ethanol and bioelectricity production in sugarcane biorefineries in Brazil', *Renewable Energy*. Elsevier Ltd, 85, pp. 371–386. doi: 10.1016/j.renene.2015.06.009.

Klein, B. C. *et al.* (2018) 'Beyond ethanol, sugar, and electricity: a critical review of product diversification in Brazilian sugarcane mills', *Biofuels, Bioproducts and Biorefining*, pp. 1–13. doi: 10.1002/BBB.1969.

Leal, F. I., Rego, E. E. and de Oliveira Ribeiro, C. (2019) 'Natural gas regulation and policy in Brazil: Prospects for the market expansion and energy integration in Mercosul', *Energy Policy*. Elsevier Ltd, 128(February 2018), pp. 817–829. doi: 10.1016/j.enpol.2019.01.030.

Lefebvre, D. *et al.* (2019) 'Assessing the potential of soil carbonation and enhanced weathering through Life Cycle Assessment: A case study for Sao Paulo State, Brazil', *Journal of Cleaner Production*. Elsevier Ltd, 233, pp. 468–481. doi: 10.1016/j.jclepro.2019.06.099.

Lefebvre, D. *et al.* (2020) 'Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil', *Scientific Reports*. Nature Publishing Group UK, 10(1), p. 19479. doi: 10.1038/s41598-020-76470-y.

Liu, Q. *et al.* (2018) 'How does biochar influence soil N cycle? A meta-analysis', *Plant*

and Soil. Plant and Soil, 426(1–2), pp. 211–225. doi: 10.1007/s11104-018-3619-4.

Lund, H. *et al.* (2010) ‘Energy system analysis of marginal electricity supply in consequential LCA’, *International Journal of Life Cycle Assessment*, 15(3), pp. 260–271. doi: 10.1007/s11367-010-0164-7.

Mašek, O. *et al.* (2019) ‘Potassium doping increases biochar carbon sequestration potential by 45%, facilitating decoupling of carbon sequestration from soil improvement’, *Scientific Reports*, 9(1), pp. 1–8. doi: 10.1038/s41598-019-41953-0.

Matušík, J., Hnátková, T. and Kočí, V. (2020) ‘Life cycle assessment of biochar-to-soil systems: A review’, *Journal of Cleaner Production*, 259, p. 120998. doi: 10.1016/j.jclepro.2020.120998.

Meyer, S. *et al.* (2012) ‘Albedo Impact on the Suitability of Biochar Systems To Mitigate Global Warming’, *Environmental Science & Technology*, 46(22), pp. 12726–12734. doi: 10.1021/es302302g.

Myhre, G. *et al.* (2013) ‘Anthropogenic and Natural Radiative Forcing.’, in Jacob, D., Ravishankara, A. R., and Shine, K. (eds) *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. New York: Cambridge University Press. doi: 10.3390/jmse6040146.

Nemet, G. F. *et al.* (2018) ‘Negative emissions — Part 3 : Innovation and upscaling’, *Environmental Research Letters*.

Oldfield, T. L. *et al.* (2018) ‘Biochar, compost and biochar-compost blend as options to recover nutrients and sequester carbon’, *Journal of Environmental Management*, 218, pp. 465–476. doi: 10.1016/j.jenvman.2018.04.061.

de Oliveira Noronha, M., Zanini, R. R. and Souza, A. M. (2019) ‘The impact of electric generation capacity by renewable and non-renewable energy in Brazilian economic growth’, *Environmental Science and Pollution Research*. Environmental Science and Pollution Research, 26(32), pp. 33236–33259. doi: 10.1007/s11356-019-06241-4.

de Oliveira Vilela, A. *et al.* (2014) ‘A new technology for the combined production of charcoal and electricity through cogeneration’, *Biomass and Bioenergy*, 69, pp. 222–240.

doi: 10.1016/j.biombioe.2014.06.019.

Olivério, J. L. and Ferreira, F. M. (2010) 'Cogeneration - A new source of income for sugar and ethanol mills', in *Proc. Int. Soc. Sugar Cane Technol.*, pp. 1–13. Available at: <https://www.semanticscholar.org/paper/COGENERATION-A-NEW-SOURCE-OF-INCOME-FOR-SUGAR-AND-Olivério-Ferreira/7ed359fa22554680fe52445155c3d7fe9106f308>.

Panorama Offshore (2018) *Thermoelectric plant will be deployed in Sao Paulo until 2024*. Available at: <https://www.panoramaoffshore.com.br/en/plant-termoeletrica-sera-deployed-in-sao-paulo-until-2024/> (Accessed: 25 February 2020).

Park, J. *et al.* (2014) 'Slow pyrolysis of rice straw: Analysis of products properties, carbon and energy yields', *Bioresource Technology*. Elsevier Ltd, 155, pp. 63–70. doi: 10.1016/j.biortech.2013.12.084.

Pennise, D. M. *et al.* (2001) 'Emissions of greenhouse gases and other airborne pollutants from charcoal making in Kenya and Brazil', *Journal of Geophysical Research: Atmospheres*, 106(D20), pp. 24143–24155. doi: 10.1029/2000JD000041.

Peters, J. F., Iribarren, D. and Dufour, J. (2015) 'Biomass Pyrolysis for Biochar or Energy Applications? A Life Cycle Assessment', *Environmental Science & Technology*, 49(8), pp. 5195–5202. doi: 10.1021/es5060786.

Pippo, W. A. and Luengo, C. A. (2013) 'Sugarcane energy use: Accounting of feedstock energy considering current agro-industrial trends and their feasibility', *International Journal of Energy and Environmental Engineering*, 4(1), pp. 1–13. doi: 10.1186/2251-6832-4-10.

PRé Consultants: Life Cycle consultancy and software solutions (2019) *SimaPro 8.3*. Amersfoort, Netherlands: PRé Consultants: Life Cycle consultancy and software solutions. Available at: <https://www.pre-sustainability.com>.

Quirk, R. G. *et al.* (2012) 'Utilization of Biochar in Sugarcane and Sugar-Industry Management', *Sugar Tech*, 14(4), pp. 321–326. doi: 10.1007/s12355-012-0158-9.

Rajabi Hamedani, S. *et al.* (2019) 'Life Cycle Assessment and Environmental Valuation of Biochar Production: Two Case Studies in Belgium', *Energies*, 12(11), p. 2166. doi:

10.3390/en12112166.

Roberts, K. G. *et al.* (2010) ‘Life Cycle Assessment of Biochar Systems: Estimating the Energetic, Economic, and Climate Change Potential’, *Environmental Science and Technology*, 44, pp. 827–833. doi: 10.1021/es902266r.

Romero, E. R. *et al.* (2007) ‘Sugarcane Potential Trash Estimation: Variety and Cane Yield Effect’, in *Int. Soc. Sugar Cane Technol*, pp. 9–13.

Roy, P. and Dias, G. (2017) ‘Prospects for pyrolysis technologies in the bioenergy sector: A review’, *Renewable and Sustainable Energy Reviews*. Elsevier Ltd, 77(September), pp. 59–69. doi: 10.1016/j.rser.2017.03.136.

Sampaio, I. L. M. *et al.* (2019) ‘Electricity Production from Sugarcane Straw Recovered Through Bale System: Assessment of Retrofit Projects’, *BioEnergy Research*. BioEnergy Research, 12(4), pp. 865–877. doi: 10.1007/s12155-019-10014-9.

Santos, M. J. *et al.* (2017) ‘Scenarios for the future Brazilian power sector based on a multi-criteria assessment’, *Journal of Cleaner Production*. Elsevier Ltd, 167, pp. 938–950. doi: 10.1016/j.jclepro.2017.03.145.

Scarpore, F. V. *et al.* (2016) ‘Sugarcane land use and water resources assessment in the expansion area in Brazil’, *Journal of Cleaner Production*, 133(June), pp. 1318–1327. doi: 10.1016/j.jclepro.2016.06.074.

Schmidt, H.-P. *et al.* (2018) ‘Pyrogenic carbon capture and storage’, *GCB Bioenergy*, (March), pp. 1–19. doi: 10.1111/gcbb.12553.

Seabra, J. E. A. *et al.* (2011) ‘Life cycle assessment of Brazilian sugarcane products: GHG emissions and energy use’, *Biofuels, Bioproducts and Biorefining*, 5(5), pp. 519–532. doi: 10.1002/bbb.289.

Seabra, J. E. A. and Macedo, I. C. (2011) ‘Comparative analysis for power generation and ethanol production from sugarcane residual biomass in Brazil’, *Energy Policy*. Elsevier, 39(1), pp. 421–428. doi: 10.1016/j.enpol.2010.10.019.

SEEG (2016) *São Paulo - Emissions, System for Greenhouse Gas Emissions and Removal Estimates*. Available at: <http://plataforma.seeg.eco.br/territories/sao->

CHAPTER 4

paulo/card?year=2016 (Accessed: 14 September 2018).

Severinghaus, S. (2019) *Understanding Electricity in SimaPro*. Available at: <https://simapro.com/2019/understanding-electricity-in-simapro>.

Silva, A. G. B. *et al.* (2019) 'How Much Sugarcane Straw is Needed for Covering the Soil?', *BioEnergy Research*. *BioEnergy Research*, 12(4), pp. 858–864. doi: 10.1007/s12155-019-10008-7.

Smith, P. *et al.* (2013) 'How much land-based greenhouse gas mitigation can be achieved without compromising food security and environmental goals?', *Global Change Biology*, 19(8), pp. 2285–2302. doi: 10.1111/gcb.12160.

Theodor Rudorff, B. F. (2014) *Canasat - INPE*. Available at: <http://www.dsr.inpe.br/laf/canasat/tabelas.html> (Accessed: 27 August 2019).

Thers, H. *et al.* (2019) 'Biochar potentially mitigates greenhouse gas emissions from cultivation of oilseed rape for biodiesel', *Science of The Total Environment*. Elsevier B.V., 671, pp. 180–188. doi: 10.1016/j.scitotenv.2019.03.257.

Vandepaer, L. *et al.* (2019) 'The integration of long-term marginal electricity supply mixes in the ecoinvent consequential database version 3.4 and examination of modeling choices', *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 24(8), pp. 1409–1428. doi: 10.1007/s11367-018-1571-4.

Vélez-Henao, J. A. and Garcia-Mazo, C. M. (2019) 'Marginal technology based on consequential life cycle assessment. The case of Colombia', *Revista Facultad de Ingenieria*, (90), pp. 51–61. doi: 10.17533/UDEA.REDIN.N90A07.

Wang, T., Camps-Arbestain, M. and Hedley, M. (2014) 'The fate of phosphorus of ash-rich biochars in a soil-plant system', *Plant and Soil*, 375(1–2), pp. 61–74. doi: 10.1007/s11104-013-1938-z.

Wender, B. A. *et al.* (2014) 'Anticipatory life-cycle assessment for responsible research and innovation', *Journal of Responsible Innovation*. Taylor & Francis, 1(2), pp. 200–207. doi: 10.1080/23299460.2014.920121.

Wernet, G. *et al.* (2016) 'The ecoinvent database version 3 (part I): overview and

methodology’, *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 21(9), pp. 1218–1230. doi: 10.1007/s11367-016-1087-8.

Wickham, H. (2016) *ggplot2: Elegant Graphics for Data Analysis*. Springer-V. New York. Available at: <https://ggplot2.tidyverse.org>.

Williamson, P. (2016) ‘Scrutinize CO₂ removal methods’, *Nature*, pp. 5–7. doi: 10.1038/530153a.

Woolf, D. *et al.* (2010) ‘Sustainable Biochar to Mitigate Global Climate Change’, *Nature Communications*. Nature Publishing Group. doi: 10.1038/ncomms1053.

Yang, Q. *et al.* (2016) ‘Greenhouse gas emissions of a biomass-based pyrolysis plant in China’, *Renewable and Sustainable Energy Reviews*. Elsevier, 53, pp. 1580–1590. doi: 10.1016/j.rser.2015.09.049.

Ye, L. *et al.* (2020) ‘Biochar effects on crop yields with and without fertilizer: A meta-analysis of field studies using separate controls’, *Soil Use and Management*, 36(1), pp. 2–18. doi: 10.1111/sum.12546.

You, S. *et al.* (2017) ‘A critical review on sustainable biochar system through gasification: Energy and environmental applications’, *Bioresource Technology*. Elsevier Ltd, 246, pp. 242–253. doi: 10.1016/j.biortech.2017.06.177.

Zhang, Y. *et al.* (2018) ‘Response of surface albedo and soil carbon dioxide fluxes to biochar amendment in farmland’, *Journal of Soils and Sediments*. Journal of Soils and Sediments, 18(4), pp. 1590–1601. doi: 10.1007/s11368-017-1889-8.

CHAPTER 5

Reforestation LCA

“Planting a tree is an act of faith.”

ROBERT BOURDU

ASSESSING THE CARBON CAPTURE POTENTIAL OF REFORESTATION PROJECTS

David Lefebvre ^a, Adrian G. Williams ^a, Guy J.D. Kirk ^a, Paul, J. Burgess ^a, Jeroen Meersmans ^b, Miles R. Silman ^{c,d,e}, Francisco Román-Dañobeytia ^{c,d}, Jhon Farfan ^c, and Pete Smith ^f

^a School of Water, Energy and Environment, Cranfield University, Cranfield, Bedfordshire, MK43 0AL, UK

^b TERRA Teaching and Research Centre, Gembloux Agro-Bio Tech, University of Liège, Gembloux 5030, Belgium

^c Centro de Innovación Científica Amazónica – CINCIA, Madre de Dios 17001, Peru

^d Center for Energy, Environment and Sustainability, Wake Forest University, NC 27106, USA

^e Department of Biology, Wake Forest University, NC 27106, USA

^f Institute of Biological and Environmental Sciences, University of Aberdeen, 23 St Machar Drive, Aberdeen AB24 3UU, UK

5.1 Abstract

The number of reforestation projects worldwide is increasing. In many cases funding is obtained through the claimed carbon capture of the trees, presented as immediate and durable, whereas reforested plots need time and maintenance to realise their carbon capture potential. Further, claims usually overlook the environmental costs of natural or anthropogenic disturbances during the forest's lifetime, and greenhouse gas (GHG) emissions associated with the reforestation are not allowed for. This study uses life cycle assessment to quantify the carbon footprint of setting up a reforestation plot in the Peruvian Amazon. In parallel, we combine a soil carbon model with an above- and below-ground plant carbon model to predict the increase in carbon stocks after planting. We compare our results with the carbon capture claims made by a reforestation platform. Our results show major errors in carbon accounting in reforestation projects if they (1) ignore the time needed for trees to reach their carbon capture potential; (2) ignore the GHG emissions involved in setting up a plot; (3) report the carbon capture potential per tree planted, thereby ignoring limitations at the forest ecosystem level; or (4) under-estimate tree losses due to inevitable human and climatic disturbances. Further, we show that applications of biochar during reforestation can partially compensate for project emissions.

5.2 Introduction

Carbon (C) sequestration programs are necessary to reach the UNFCCC Paris Agreement targets and limit the global average temperature increase to well below 2°C (Smith, Davis, Creutzig, Fuss, Minx, Gabrielle, Kato, Jackson, Cowie, Kriegler, van Vuuren, *et al.*, 2016). Planting trees is an effective way to capture C (S. L. Lewis *et al.*, 2019). Compared with other greenhouse gas (GHG) capture practices, it is cheap and easy to set up using established technology (Griscom *et al.*, 2017). The number of tree planting projects globally has increased in the past decade (Holl and Pedro H. S. Brancalion, 2020), with the aim of both supporting livelihoods and sequestering carbon dioxide (CO₂) into long-term biomass (Smith *et al.*, 2016; EASAC, 2018). This year marks the beginning of the United Nation decade on ecosystem restoration where incentives will be put in place to restore degraded ecosystems, in part through reforestations (United Nation Decade on Ecosystem Restoration, 2021). However, the extent to which such projects can contribute to global GHG capture targets is debated (Anderson *et al.*, 2019) and it is important that claimed sequestration potentials are realistic.

So-called reforestation platforms distribute funding between different reforestation/afforestation projects. Reforestation platforms are responsible for planting large numbers of trees. *Ecosia*, for example, has planted over 100 million trees in more than 25 countries since its creation in 2009 (Ecosia, 2021). Reforestation platforms often cite a C or CO₂ capture figure per tree planted (Reforest'Action, 2020; Tree-Nation, 2020), rather than at the forest ecosystem level. Carbon sequestration claims are often calculated ahead of time based on expected wood density and maximum height of the planted trees (Tree-Nation, 2020), following carbon removal rates from published studies (e.g. Bernal *et al.*, 2018), or they are sometimes left to certification bodies, using similar techniques (e.g. Gold Standard (2017), Verra (2021)). This ignores the fact that reforestation is a long term undertaking and failure rates are often high, for example where a lack of soil care and seedling protection results in tree death during establishment (Mansourian, Vallauri and Dudley, 2006; Holl and Pedro H.S. Brancalion, 2020). In addition, many steps are involved in the setup and maintenance of a reforestation plot, all involving some type of energy consumption, leading to GHG emissions (Gaboury *et al.*, 2009). However, information from reforestation platforms about the time needed for

capture claims to be realised, or the environmental impact of the reforestation itself, are often over-looked (Ecosia, 2019; Reforest'Action, 2020; Tree-Nation, 2021).

In this study, we use life cycle assessment (LCA) to empirically derive the carbon footprint of setting up and maintaining a reforestation plot for one year in a tropical forest (Methods). We include a focus on biochar, a recently adopted soil amendment of widespread interest, used in the plot studied (Román-Dañobeytia *et al.*, 2021). In parallel, we combine above and below ground biomass models with a soil carbon model, RothC (Coleman and Jenkinson, 2014), to provide an evolving carbon capture profile tailored for the tropical plot under study. We then compare our results with claims made by a typical reforestation platform and discuss their relevance. Informative representation of the processes and stocks accounted for in this study is shown in Figure 5.1.

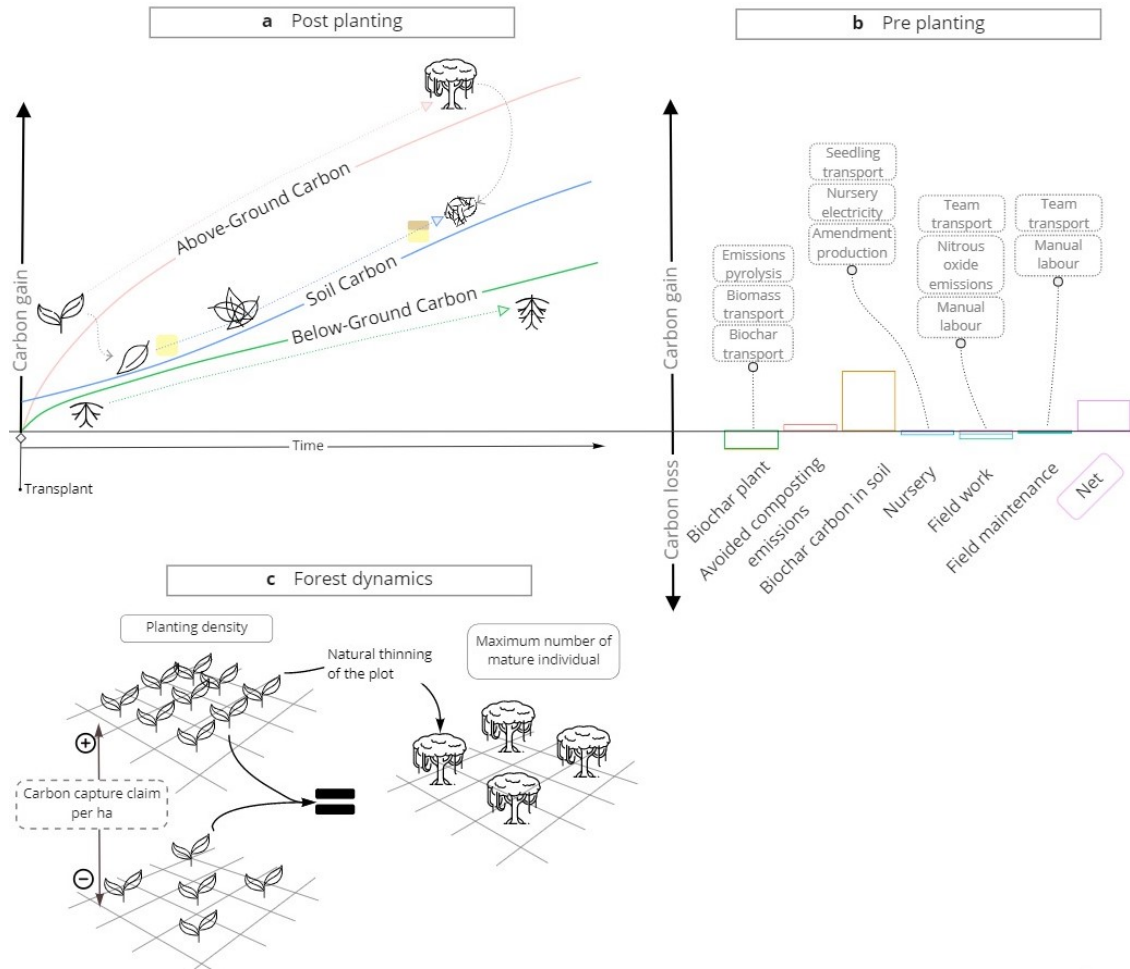


Figure 5.1: Descriptive schematic figure of the processes and stocks considered. a Vegetation and soil C stocks. b Processes included in the LCA and their relative impact. c effect of the planting density.

5.3 Results

5.3.1 Modelling soil and biomass pools

The C content of topsoil took around 50 years to reach steady state following transplanting the new forest, while both above- and below-ground vegetation pools increased over 100 years at a declining rate over time (Figure 5.2a) (Stephenson *et al.*, 2014; Jones *et al.*, 2019). Simulations of the below-ground biomass model matched a previous assessment (Vance and Nadkarni, 1992) and the evolution of total carbon stocks after transplant (Figure 5.2b) is consistent with previous assessments (Berenguer *et al.*, 2014; Jones *et al.*, 2019). About 80 years after transplanting, the additional carbon stocks comprise 45% above-ground carbon, 30% below-ground carbon and 25% soil carbon.

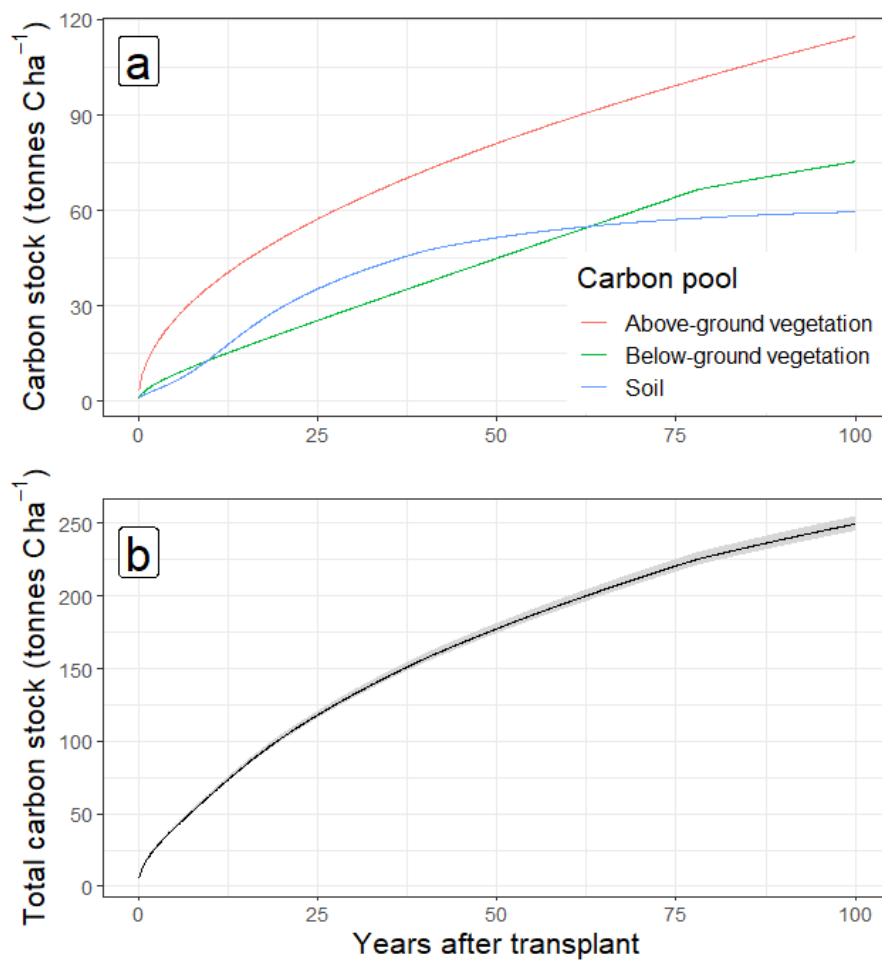


Figure 5.2: Changes in carbon stocks following transplanting. a Vegetation and soil. b Total, with ± 1 SD indicated by the ribbon.

5.3.2 Life cycle assessment of the case study plot

The LCA shows that using biochar as a reforestation amendment delivered a net capture of $0.51 \pm 0.18 \text{ t C ha}^{-1}$ ($1.87 \pm 0.66 \text{ t CO}_2\text{e ha}^{-1}$) within the first year after planting (Figure 5.3), but the biochar opportunity was site specific. Most of the establishment emissions arise from pyrolysis, fertilizer manufacture and use, and associated N_2O emissions, and 90% of the sequestration comes from the $1.11 \text{ t biochar ha}^{-1}$ application (capturing $3.45 \text{ t CO}_2\text{e ha}^{-1}$, i.e. 0.94 t C ha^{-1}). Excluding biochar production and use, establishing the case study plot emits $1.27 \pm 0.10 \text{ t CO}_2\text{e ha}^{-1}$ (0.35 t C ha^{-1}). The complete contribution analysis is given in the Supplementary Information, Appendix D.

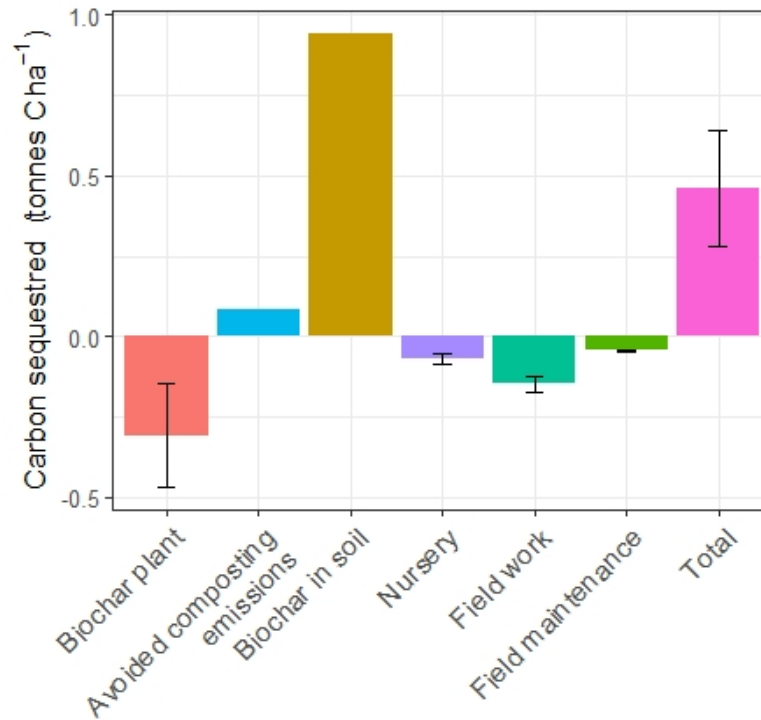


Figure 5.3: Grouped contribution analysis of the emission and emission reduction associated with reforesting one hectare from our case study plot including one year maintenance. The error bars represent ± 1 SD.

5.3.3 Comparison with a hypothetical reforestation hub

We calculate it would take 4.1 years after transplanting to capture 100 kg CO₂ per tree planted in our case study plot (including all C pools – Figure 5.4), which is a typical reforestation platform target. With a planting density of 1,111 seedlings per hectare (3 m spaced grid) 100 kg CO₂ (i.e., 27.3 kg of C) per tree equates to 111,100 kg CO₂ (i.e., 30,300 kg of C) captured per ha after 4.1 years. Without biochar, setting up the case study reforestation plot would emit 1.27 ± 0.10 t CO₂e ha⁻¹. This carbon debt is quickly covered by the growing seedlings (Figure 5.4). If we only account for aboveground biomass (i.e., excluding soil C stock and below-ground C) our model predicts 8.7 years to reach the 100 kg CO₂ carbon capture target per tree.

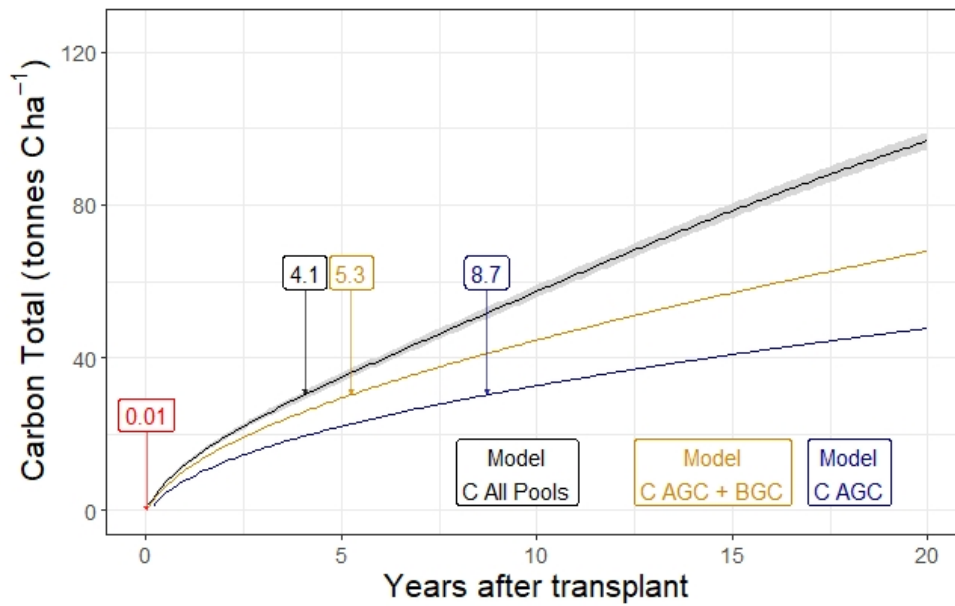


Figure 5.4: Time needed in years (labels) to reach estimated claim of 100 kg CO₂e captured per tree using our model considering all C pools (black), above and below-ground C pool only (yellow), and above-ground C pool only (blue). The red label represents the time in years necessary to offset the emissions associated to set up the plot. The ribbon around the model carbon capture rate is ± 1 SD.

Figure 5.5 shows the time needed to reach the 100 kg CO₂ tree⁻¹ as a function of seedling density. Following the carbon capture claim per tree, the more numerous the seedlings planted per hectare, the higher the total carbon claim per hectare. However, considering the non-linear increase in C stocks in a growing forest (Figure 5.2), the higher the total carbon capture claim per hectare, the longer it will take for the plot to achieve the claim.

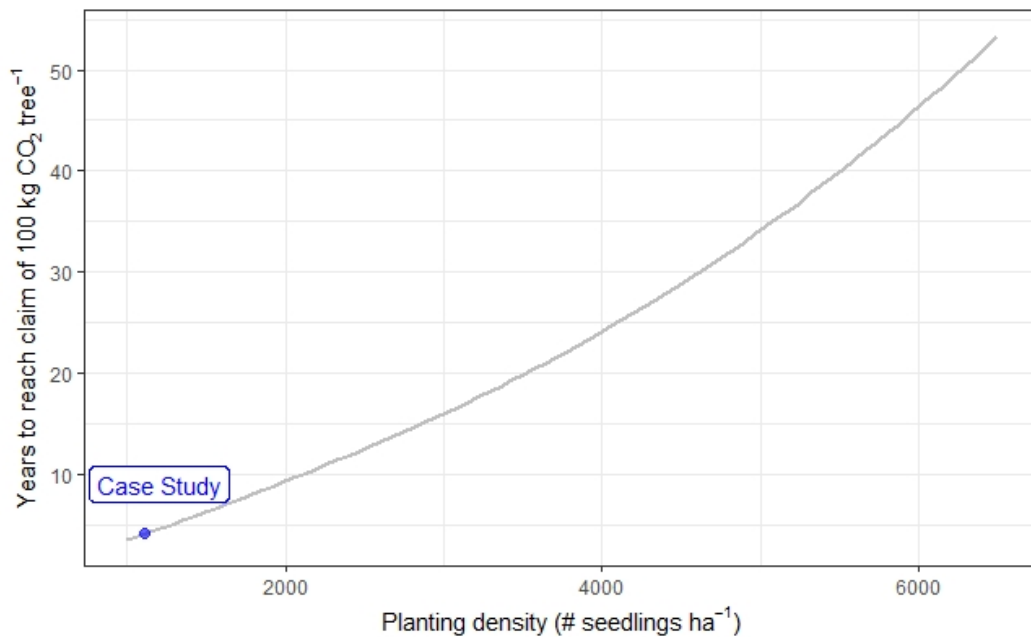


Figure 5.5: Effect of the planting density on the plot-scale carbon capture rate. No manual thinning and 100% seedling transplant survival are assumed. The blue dot represents the planting density of 1,111 seedling ha⁻¹ (where our model reach 100 kg CO₂ captured per tree 4.1 years after planting).

5.4 Discussion

Our study shows that the case-study plot would need to be cared for and maintained for 4.1 years before the 100 kg CO₂e captured per tree claimed could be reached (considering 1,111 seedling ha⁻¹). These modelled numbers are likely to be at the lower-end of actual values achieved in the field as they were derived on a soil that was highly degraded and the carbon stock consequently increased by around 40 t C ha⁻¹ in the first 50 years (Figure 5.2). In addition, the biomass growth is modelled for low altitude tropical rainforest, which is not representative of all reforestation projects. Whereas the carbon sequestration occurs in the future, some reforestation platforms assume the sequestration is immediate (Reforest'Action, 2020; Tree-Nation, 2021). This potentially misleads supporters and buyers into thinking that their carbon offsets are both instantaneous and permanent. Despite the importance of these reforestation platforms for climate change mitigation, their forward carbon capture claims can be misinterpreted due to i) the omission of forest growth trajectory, ii) by ignoring the emissions associated with the reforestation and monitoring processes, and iii) the risk that either the trees die or be destroyed or that they

are harvested for a use where the carbon is soon released as carbon dioxide (Ferreira *et al.*, 2020). These amplified and premature C capture values can allow companies to claim substantial carbon footprint reductions by massively investing in reforestation projects (e.g. Shell (2019)), hence misleading the public on their actual impact. This practice also risks devaluing CO₂ credits which, in turn, could undermine the attractiveness of other carbon capture techniques or platforms.

We show that GHG emissions associated with establishing the plot need to be factored into the carbon accounting. The amendments and substrates needed in the nursery and their associated nitrous oxide emissions represent a substantial share of the contribution analysis (Supplementary Information, Appendix D). As a result, planting density and seedling mortality should be monitored and minimised to reduce the overall carbon footprint of establishing the plot. Excluding biochar production and use, establishing the case study plot emitted 1.27 t CO₂ ha⁻¹, which is very similar to reforesting open woodland in a Canadian boreal forest, i.e. 1.25 t CO₂ ha⁻¹ (Gaboury *et al.*, 2009). Although low and being rapidly offset by the growing forest (Figure 5.4), projects relying on heavy machinery for land preparation, transporting and spreading fertile soil (the practice of “re-soiling”, sometimes used for land reclamation/restoration (ODNR, 2005)), eliminating competitive shrubs, mounding or subsoiling the plot to increase seedling growth and/or survival (Löf *et al.*, 2012) will have a higher establishment carbon footprint to offset before any carbon capture claim can be made. For instance, subsoiling would increase the carbon debt by an average of 41.4 ± 10.3 kg CO₂e ha⁻¹ (Lal, 2004) (apart from transporting the machinery to the site).

The use of biochar provides an opportunity to offset the GHG emissions associated with establishing the plot. Using another biochar feedstock that is not destined to be composted (Section 5.5.4), but could be used for other purposes, would reduce the overall capture of the practice by at least 8% (solely accounting for the avoided composting emissions – Supplementary Information, Appendix D). On the other hand, improving the pyrolysis process by improving equipment performance (at an economic cost) and reducing its emissions by half, would increase the carbon capture potential of using biochar by 30%. Similarly, applying the biochar at the hectare scale instead of locally to each seedling would vastly increase the carbon sequestration.

CHAPTER 5

We used an above-ground carbon model of secondary tropical forest growth. Natural forest regeneration models do not accurately represent the reforestation process where seedlings are deliberately transplanted to increase cover and forest regeneration. But they do account for the natural thinning of the plot as trees mature, which is not replicated by an individual seedling's growth trajectory on recently reforested land. In addition, studies show that natural regeneration can be more effective for increasing carbon stocks than reforestation, depending on the former land use and location of the site (Crouzeilles *et al.*, 2017; Meli *et al.*, 2017). The speed at which carbon stocks increase in the natural regeneration model is not altered by the planting density of the hypothetical reforestation project it represents. The asymptotic shape of the calculated change in above-ground C (Figure 5.2a) symbolises a rapid colonization of the plot by pioneer species followed by a slow reduction in rate over time as the number of individuals thins out and the respiration load of the woodland increases. Reforestation platforms' capture claims are based on individual trees, meaning that claims per hectare are highly dependent on the planting density. Figure 5.5 shows that planting density clearly affects the time needed to reach the carbon capture claim per tree. Nevertheless, no matter what the original planting density was, the maximum number of thriving individual trees per hectare is limited by environmental parameters (ter Steege *et al.*, 2013). Thinning is seen in the natural regeneration model used in this study (characterized by its asymptotic shape). Although advertising the carbon capture impact of each tree planted may be an effective communication strategy by reforestation platforms, these observations should motivate carbon capture verification bodies to require carbon capture claims per hectare using context-specific models accounting for ecosystem limitations, ideally supplemented by on-site surveys. A major issue when foreseeing carbon capture figures at the individual tree level is that it implies that all trees will prosper and reach maturity, which overlooks the competition between individual trees for light and other resources leading inevitably to a maximum number of individuals of a given size that any specific area can sustain (ter Steege *et al.*, 2013).

Growing forests are subject to numerous threats. The frequency of wildfire has increased in rainforests over the past decades (Silva *et al.*, 2020). While it is agreed that wildfires have a major negative impact on forest carbon stocks (Silva *et al.*, 2018), accounting for all potential effects of the fire on the regrowth of a forest plot is difficult (Silva *et al.*,

2020). Although wildfires happen predominantly on old growth forest, with more dead material in old growth forests compared to secondary forests (Withey *et al.*, 2018), tree mortality has increased in young secondary forests, particularly in the western and southern Amazon (Esquivel-Muelbert *et al.*, 2020). In addition, droughts have also become more frequent in Amazonia leading to potential lasting degradation of these ecosystems and their carbon sink ability (De Faria *et al.*, 2020; De Meira Junior *et al.*, 2020). The increase in atmospheric CO₂ concentration can increase tree growth in the absence of other constraints (Hubau *et al.*, 2020) but faster growth can lead to reduced tree lifespans with a negative impact on total carbon sequestration capacity (Brienen *et al.*, 2020). These observations add to the overall adverse effects of climate change on the C sink of the Amazon (De Meira Junior *et al.*, 2020; Hubau *et al.*, 2020). While the prospective impacts of wildfires, droughts or climate change are difficult to assess for a recently reforested plot in a remote location of the rainforest, their occurrence and impact have increased. Further, diseases can have a disastrous impact on the survival of some tree species (Hultberg *et al.*, 2020). Hence, there is a substantial risk in assuming that an unsupervised young reforested plot, particularly of a single species, will provide permanent carbon sequestration.

Brander *et al.* argue that businesses should only report CO₂ capture figures that have been already and permanently captured (Brander *et al.*, 2021). An attraction of carbon storage in the above-ground biomass of forests is that it can be readily assessed using remote sensing techniques supported by surveys (Li *et al.*, 2020). However, as discussed earlier, the above-ground C credits associated the future growth of reforestation projects come with associated risks, and the figures should be frequently monitored and evaluated to ensure appropriate accuracy (Gorte and Ramseur, 2008; Brander *et al.*, 2021). Some authors argue that the evolution of soil carbon stocks in reforested land can be a more durable form of carbon stock, less prone to disturbance (e.g. logging or fire) than the above-ground stock (Berenguer *et al.*, 2014) and reach equilibrium faster, at least in the tropics (Berenguer *et al.*, 2014; Jones *et al.*, 2019). Including soil C stock changes in the total C accounting scheme following reforestation would increase its accuracy (De Rosa *et al.*, 2017). Although, the calculation and verification of the effect of reforestation on soil carbon stocks remains complex and challenging to non-specialists (Smith *et al.*, 2019), validated methodologies could improve the attraction of afforestation projects on

degraded land where potential soil C gain is higher than, for example, that in grassland ecosystems with fertile soils (Guo and Gifford, 2002; Veldman *et al.*, 2015; Xiao *et al.*, 2020).

Overall, because the carbon dioxide released into the atmosphere will, on average, remain there for centuries (Archer *et al.*, 2009), the carbon credit sold as an emission permit must ensure that the carbon accounted for is also sequestered for centuries. Our modelling shows that the carbon sequestration claims of reforestation platforms are likely to be unreliable if they do not allow for the time dependency of the carbon capture by planted trees, the risks of tree failure and harvest, and potential changes in soil carbon. Reforestation/afforestation projects obtain massive investments worldwide and are a potential loophole for companies with historically high GHG emissions. Therefore, it is important to emphasise that the carbon capture of reforested/afforested sites relies on the permanence of the tree stand and the end-of-life use of any harvested trees. Accounting for actual carbon stocks in growing forests needs a transparent quantification of the risks, constant monitoring and relevant functional units. There should also be the possibility of withdrawal in case of losses. It is necessary to distinguish between predictions of future carbon sequestration and validated carbon sequestration. While selling future carbon sequestration units can provide reforestation projects with financial inputs to help development and growth, the credits should not be recognised as contributing to a company's greenhouse gas budget until the carbon has been sequestered and validated. Differentiation between “actual carbon stock increase” and “future carbon credits” should be clarified and marketed differently.

The global warming impact of planting seedlings, which is *per se* a GHG emitting process, can be lowered by using biochar. Reducing the global warming impact by planting trees is a useful first step, which is context-specific, but validation of the extent and permanence of the future growth is also required.

5.5 Methods

5.5.1 Study area

The present LCA is based on a reforestation project set up and maintained by the Centre for Amazonian Scientific Innovation (CINCIA, 2021), in the Peruvian Amazon. CINCIA

has, so far, reforested 42.5 ha of degraded forest using 74 tree species on 19 different plots (Cabanillas, Condori and B., 2019). The plot studied here is located in the south-eastern Peruvian Amazon region of Madre de Dios (12°41'3.15"S, 69°36'47.76"W) at an altitude of 266 meters above sea level (Figure 5.6). It corresponds to Site 1 in Román-Dañobeytia *et al.* (2021) and is on an open sandy area under an enriched biochar treatment including biochar and fertilizer (Supplementary Information, Appendix D) (Román-Dañobeytia *et al.*, 2021).



Figure 5.6: Case study location. Generated using Google Earth (Google, 2018)

5.5.2 System boundary and functional unit

The carbon footprint of this case study includes processes from the receipt of the seeds by the nursery, their development into seedlings and transport to the reforestation plot. In parallel, the system boundaries include the biomass collection, its transformation into biochar and the transportation of the amendments to the reforestation plot. They also include the field work necessary at the time of transplant and for the maintenance of the seedling up to one year after transplant (Figure 5.7), following which, no further care of the plot is made by the team. The reforestation takes place in a protected area and hence

the LCA excludes timber collection and assumes that the area is left untouched indefinitely. We use 100-year global warming potentials (GWP) as prescribed in the IPCC Fifth Assessment Report (AR5) (IPCC, 2014) and “one reforested hectare” as the functional unit.

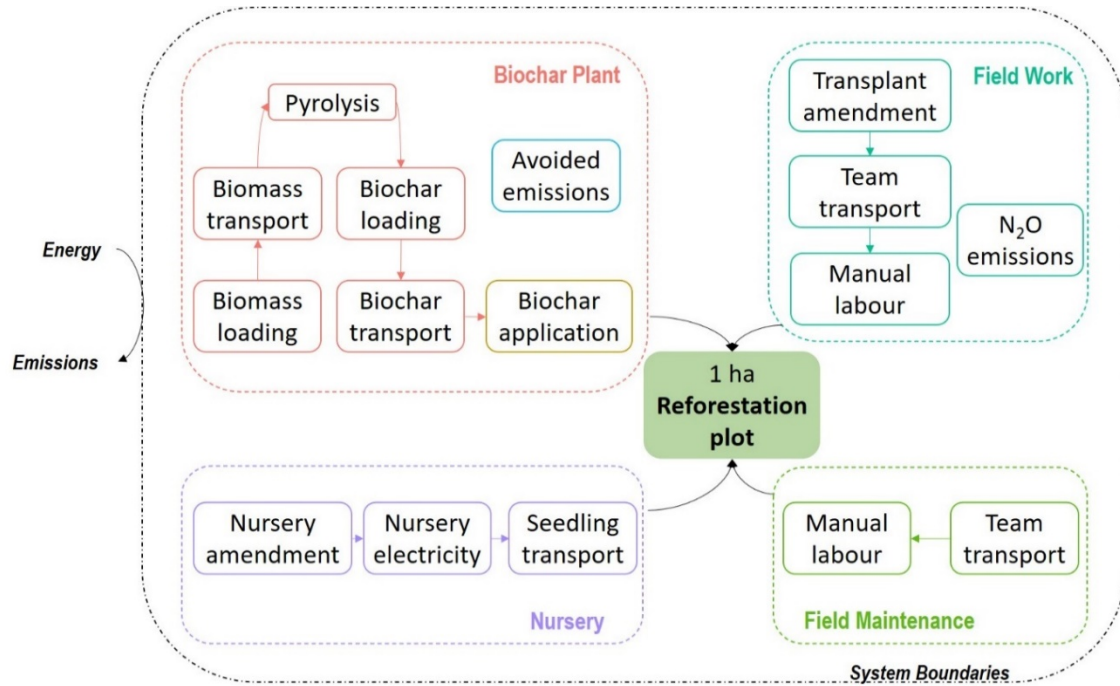


Figure 5.7: System boundaries of the study

5.5.3 Software, database and data processing

Emissions factors of processes and fuels were assessed using EcoInvent 3.7 and USLCI databases in SimaPro 8.3 (PRé Consultants: Life Cycle consultancy and software solutions, 2019) and their carbon footprint calculated using the IPCC 2013 GWP 100a V1.03 method (Myhre *et al.*, 2013). Data and emissions factors are available in the Supplementary Information, Appendix D. Life cycle impact assessment, uncertainty analysis (Monte Carlo method), modelling activities, and figures were produced using R software (version 3.5.1) (Core Team, 2018).

5.5.4 Biochar

Biochar is produced and added to the site at the time of seedlings transplant at a rate of one kg (dry mass) per seedling (Román-Dañobeytia *et al.*, 2021). The biochar is produced using Brazil nut husks, residues of the local Brazil nut production (Lefebvre *et al.*, 2018).

Pyrolysis of Brazil nut husks avoids the emissions associated with piling these residues in unmanaged heaps in the environment, as is it commonly done in the region (Jhon Farfan, *pers. com.*). First, the husks are loaded in a medium scale truck using a front loader. The truck then drives an average distance of 20 km from the city to the biochar producing area and unloads the biomass. The biomass is then sun-dried and manually loaded into a top-lit up-draft (TLUD) biochar pyrolyzer (Lefebvre *et al.*, 2018). It is then loaded into a truck and a boat, to be transported to the plot and applied alongside each seedling in the planting hole.

5.5.5 Nursery

CINCIA possesses its own technological tree nursery where the seeds develop into seedlings before transplant (Farfan, 2020). The seedlings are grown in reusable plastic nursery tubes in an in-house growing medium (Supplementary Information, Appendix D). CINCIA's seedling transplant rate is 1,111 seedlings ha⁻¹ (Román-Dañobeytia *et al.*, 2021). The nursery grows seedlings in batches and requires 20.5 kWh electricity per month (Supplementary Information, Appendix D). The seedlings need five months to reach a sufficient height (Jhon Farfan, *pers. com.*), hence the average electricity needed reaches 102 kWh per hectare. The seedlings are then transported from the nursery to the field by truck (128 km) and boat (9 km) to reach the plot.

5.5.6 Field work

At transplanting, each seedling is planted along with 1 kg biochar followed by a surface application of an additional 100 g granulated N-P₂O₅-K₂O 20-20-20 (Román-Dañobeytia *et al.*, 2021). All field work is done manually and requires eight people per hectare for around three days (Jhon Farfan, *pers. com.*). We accounted for the emissions associated with manual labour according to Rugani *et al.* (2012) considering the purchasing power parity of Peru (OECD, 2020) for increased accuracy. The field work process also accounts for the direct and indirect N₂O emissions associated with the nitrogen content of all amendments following the Tier 1 method (IPCC, 2006).

5.5.7 Field maintenance

Field maintenance (cleaning) is required three times in the first year (at three, six, and twelve months) (Farfan, 2020). We accounted for a team of eight people per hectare

working for eight hours for each maintenance round. Manual labour was accounted for similarly to field work.

5.5.8 Soil carbon modelling

We used the RothC soil carbon model (Coleman and Jenkinson, 2014) to simulate changes in the soil carbon stock over time after transplanting. We used RothC in reverse mode to get insights into the total carbon input needed to maintain the soil carbon stock at the level of the degraded areas and the adjacent forest. We then computed a sigmoid curve to describe the required carbon input over time, reaching maximum carbon input 40 years after transplanting (Becknell, Kissing Kucek and Powers, 2012; Jones *et al.*, 2019) and used this increasing carbon input over time in RothC to compute the soil carbon change following seedling transplant (as described in Cerri *et al.* (2003)). We accounted for biochar soil carbon stock impacts following the approach described in Lefebvre *et al.* (2020) and used their modified coding of RothC in R. Meteorological data, soil and additional data required for RothC are given in the Supplementary Information, Appendix D.

5.5.9 Above- and below-ground carbon modelling

For the above-ground carbon model we used the moist forest, non-plantation model reported in Busch *et al.* (2019), which simulates tropical secondary forests above-ground growth based on data from 829 tropical forest stands. We tested the model by comparing it with above-ground carbon value measured on plots near the case study plot (Asner *et al.*, 2010; Messinger, Asner and Silman, 2016; Csillik and Asner, 2020) (Figure 5.8). The model values are in the lower range of above-ground carbon stock increase for Peruvian forests (Cook-Patton *et al.*, 2020).

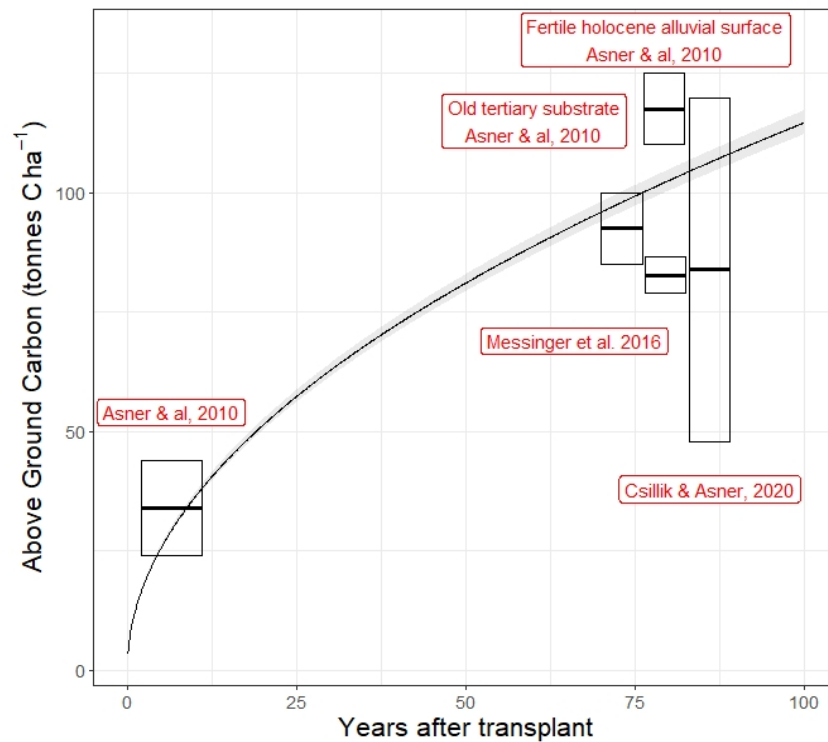


Figure 5.8: Above-ground carbon model (black line) and boxes representing above-ground carbon values from the literature of forest plot nearby our case study. Forest plots represented by the boxes on the right hand side were assumed to be a mature forest (≈ 80 years) (Becknell, Kissing Kucek and Powers, 2012; Requena Suarez et al., 2019; S. L. Lewis et al., 2019; T. Lewis et al., 2019)

The below-ground carbon model has an evolving root to shoot ratio over time, specific for tropical forests, with the root to shoot ratio of one tree increasing with tree size (ranging from 0.29 to 0.65 for young and old growth forests, respectively) (Waring and Powers, 2017).

5.5.10 Comparison with a reforestation platform

First, we used our three C pool model to foresee the time needed to reach a claim of 100 kg CO₂ captured tree⁻¹ planted (consistent with existing reforestation platforms (Ecosia, 2019; Reforest'Action, 2020; Tree-Nation, 2020)) considering the planting density similar to the one presented in the case study (1,111 seedling ha⁻¹). Similarly, we assumed the emissions associated with the nursery, field work and field maintenance processes equivalent to our case study. In addition, we considered that reforestation platform represented here does not use biochar and only reforests on highly degraded soils as the

one type in our case study. Then we modify the planting density and observe its impact on the time needed to reach the carbon capture claim. Data used for the modelling activities is available in the Supplementary Information, Appendix D.

5.6 Acknowledgements

We acknowledge funding through the UP-Green-LCA (NE/P019668/1) and Soils-R-GGREAT (NE/P019498/1) projects of the greenhouse gas removal (GGR) programme. The GGR programme is financed by the UK Natural Environment Research Council (NERC), Engineering and Physical Science Research Council (EPSRC), Economic and Social Science Research Council (ESRC) and the UK department for Business, Energy and Industrial Strategy (BEIS). We thank CINCIA and its funders (USAID and WWF) for their help and support during this project.

5.7 References

- Anderson, C. M. *et al.* (2019) ‘Natural climate solutions are not enough’, *Science*, 363(6430), pp. 933–934. doi: 10.1126/science.aaw2741.
- Archer, D. *et al.* (2009) ‘Atmospheric Lifetime of Fossil Fuel Carbon Dioxide’, *Annual Review of Earth and Planetary Sciences*, 37(1), pp. 117–134. doi: 10.1146/annurev.earth.031208.100206.
- Asner, G. P. *et al.* (2010) ‘High-resolution forest carbon stocks and emissions in the Amazon’, *Proceedings of the National Academy of Sciences of the United States of America*, 107(38), pp. 16738–16742. doi: 10.1073/pnas.1004875107.
- Becknell, J. M., Kissing Kucek, L. and Powers, J. S. (2012) ‘Aboveground biomass in mature and secondary seasonally dry tropical forests: A literature review and global synthesis’, *Forest Ecology and Management*. Elsevier B.V., 276, pp. 88–95. doi: 10.1016/j.foreco.2012.03.033.
- Berenguer, E. *et al.* (2014) ‘A large-scale field assessment of carbon stocks in human-modified tropical forests’, *Global Change Biology*, 20(12), pp. 3713–3726. doi: 10.1111/gcb.12627.
- Bernal, B., Murray, L. T. and Pearson, T. R. H. (2018) ‘Global carbon dioxide removal rates from forest landscape restoration activities’, *Carbon Balance and Management*.

- Springer International Publishing, 13(1). doi: 10.1186/s13021-018-0110-8.
- Brander, M. *et al.* (2021) ‘Carbon accounting for negative emissions technologies’, *Climate Policy*. Taylor & Francis, 0(0), pp. 1–19. doi: 10.1080/14693062.2021.1878009.
- Brienen, R. J. W. *et al.* (2020) ‘Forest carbon sink neutralized by pervasive growth-lifespan trade-offs’, *Nature Communications*. Springer US, 11(1), pp. 1–10. doi: 10.1038/s41467-020-17966-z.
- Busch, J. *et al.* (2019) ‘Potential for low-cost carbon dioxide removal through tropical reforestation’, *Nature Climate Change*. Springer US, 9(6), pp. 463–466. doi: 10.1038/s41558-019-0485-x.
- Cabanillas, F., Condori, E. and B., L. L. (2019) *Restauración de áreas degradadas por la extracción minera aurífera en Madre de Dios*. Available at: <https://cincia.wfu.edu/publicaciones/restauracion-de-areas-degradadas-por-mineria-aurifera-en-madre-de-dios/>.
- Cerri, C. E. P. *et al.* (2003) ‘Modeling Soil Carbon from Forest and Pasture Ecosystems of Amazon, Brazil’, *Soil Science Society of America Journal*, 67(6), p. 1879. doi: 10.2136/sssaj2003.1879.
- CINCIA (2021) *CINCIA – Centro de Innovación Científica Amazónica*. Available at: <http://cincia.wfu.edu/> (Accessed: 25 January 2021).
- Coleman, K. and Jenkinson, D. S. (2014) *RothC - A model for the turnover of carbon in soil Model - Model description and users guide*. Harpenden: Routledge. Available at: https://www.rothamsted.ac.uk/sites/default/files/RothC_guide_WIN.pdf.
- Cook-Patton, S. C. *et al.* (2020) ‘Mapping carbon accumulation potential from global natural forest regrowth’, *Nature*, 585(7826), pp. 545–550. doi: 10.1038/s41586-020-2686-x.
- Core Team, R. (2018) *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. Vienna. Available at: <https://www.r-project.org/>.
- Crouzeilles, R. *et al.* (2017) ‘Ecological restoration success is higher for natural regeneration than for active restoration in tropical forests’, *Science Advances*, 3(11), pp.

CHAPTER 5

1–8. doi: 10.1126/sciadv.1701345.

Csillik, O. and Asner, G. P. (2020) ‘Aboveground carbon emissions from gold mining in the Peruvian Amazon’, *Environmental Research Letters*. IOP Publishing, 15(1). doi: 10.1088/1748-9326/ab639c.

EASAC (2018) *Negative emission technologies: What role in meeting Paris Agreement targets?* Halle - Germany. Available at: <https://easac.eu>.

Ecosia (2019) *50! Million! Trees!* Available at: <https://blog.ecosia.org/ecosia-has-planted-50-million-trees/> (Accessed: 22 January 2021).

Ecosia (2021) *What is Ecosia? - The search engine that plants trees.* Available at: <https://info.ecosia.org/what> (Accessed: 22 April 2021).

Esquivel-Muelbert, A. *et al.* (2020) ‘Tree mode of death and mortality risk factors across Amazon forests’, *Nature Communications*, 11(1). doi: 10.1038/s41467-020-18996-3.

Farfan, J. F. (2020) *Produccion de Plantones en el Vivero Tecnificado de Mazuko Para Recuperar Suelos Degradado por Minería Aluvial en Madre de Dios.* Puerto Maldonado.

De Faria, B. L. *et al.* (2020) ‘Model-Based Estimation of Amazonian Forests Recovery Time after Drought and Fire Events’, *Forests*, 12(1), p. 8. doi: 10.3390/f12010008.

Ferreira, J. *et al.* (2020) ‘Life Cycle Assessment of Maritime Pine Wood: A Portuguese Case Study’, *Journal of Sustainable Forestry*. Taylor & Francis, 00(00), pp. 1–15. doi: 10.1080/10549811.2020.1768871.

Gaboury, S. *et al.* (2009) ‘Estimating the net carbon balance of boreal open woodland afforestation: A case-study in Québec’s closed-crown boreal forest’, *Forest Ecology and Management*, 257(2), pp. 483–494. doi: 10.1016/j.foreco.2008.09.037.

Gold Standard (2017) ‘Gold Standard Afforestation Reforestation (A/R) GHG Emissions Reduction & Sequestration Methodology. Version 1’, (July), p. 18. Available at: <https://globalgoals.goldstandard.org/wp-content/uploads/2017/07/401.13-AR-Methodology-V1-1.pdf>.

Google (2018) *Google Earth Pro.* Available at: <https://www.google.com/earth/download/gep/agree.html>.

- Gorte, R. W. and Ramseur, J. L. (2008) *Forest carbon markets: Potential and drawbacks, The Role of Forests in Carbon Capture and Climate Change*. Available at: www.crs.gov.
- Griscom, B. W. *et al.* (2017) 'Natural climate solutions', *Proceedings of the National Academy of Sciences of the United States of America*, 114(44), pp. 11645–11650. doi: 10.1073/pnas.1710465114.
- Guo, L. B. and Gifford, R. M. (2002) 'Soil carbon stocks and land use change: A meta analysis', *Global Change Biology*, 8(4), pp. 345–360. doi: 10.1046/j.1354-1013.2002.00486.x.
- Holl, K. D. and Brancalion, Pedro H.S. (2020) 'Supplementary Material - Tree planting is not a simple solution', *Science*, 368(6491), pp. 580–581. doi: 10.1126/science.aba8232.
- Holl, K. D. and Brancalion, Pedro H. S. (2020) 'Tree planting is not a simple solution', *Science*, 368(6491), pp. 580–581. doi: 10.1126/science.aba8232.
- Hubau, W. *et al.* (2020) 'Asynchronous carbon sink saturation in African and Amazonian tropical forests', *Nature*, 579(7797), pp. 80–87. doi: 10.1038/s41586-020-2035-0.
- Hultberg, T. *et al.* (2020) 'Ash dieback risks an extinction cascade', *Biological Conservation*. Elsevier, 244(March), p. 108516. doi: 10.1016/j.biocon.2020.108516.
- IPCC (2006) 'IPCC Guidelines for National Greenhouse Gas Inventories. Chapter 11: N₂O emissions from managed soils, and CO₂ emissions from lime and urea application', pp. 11.1-11.54. Available at: http://www.ipcc-nggip.iges.or.jp/public/2006gl/pdf/4_Volume4/V4_11_Ch11_N2O&CO2.pdf.
- IPCC (2014) *Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Edited by T. F. Stocker *et al.* Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press. Available at: <https://www.ipcc.ch/report/ar5/wg1/>.
- Jones, I. L. *et al.* (2019) 'Above- and belowground carbon stocks are decoupled in secondary tropical forests and are positively related to forest age and soil nutrients respectively', *Science of the Total Environment*. Elsevier B.V., 697, p. 133987. doi: 10.1016/j.scitotenv.2019.133987.

CHAPTER 5

Lal, R. (2004) 'Carbon emission from farm operations', *Environment International*, 30(7), pp. 981–990. doi: 10.1016/j.envint.2004.03.005.

Lefebvre, D. *et al.* (2018) *Producción y Utilización de Biocarbón*. Available at: http://cincia.wfu.edu/wp-content/uploads/Nota-Técnica_Biochar-1.pdf (Accessed: 11 June 2019).

Lefebvre, D. *et al.* (2020) 'Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil', *Scientific Reports*. Nature Publishing Group UK, 10(1), p. 19479. doi: 10.1038/s41598-020-76470-y.

Lewis, S. L. *et al.* (2019) 'Restoring natural forests is the best way to remove atmospheric carbon', *Nature*, 568(7750), pp. 25–28. doi: 10.1038/d41586-019-01026-8.

Lewis, T. *et al.* (2019) 'Reforestation of agricultural land in the tropics: The relative contribution of soil, living biomass and debris pools to carbon sequestration', *Science of The Total Environment*. Elsevier B.V., 649, pp. 1502–1513. doi: 10.1016/j.scitotenv.2018.08.351.

Li, Y. *et al.* (2020) 'Forest aboveground biomass estimation using Landsat 8 and Sentinel-1A data with machine learning algorithms', *Scientific Reports*. Springer US, 10(1), p. 9952. doi: 10.1038/s41598-020-67024-3.

Löf, M. *et al.* (2012) 'Mechanical site preparation for forest restoration', *New Forests*, 43(5–6), pp. 825–848. doi: 10.1007/s11056-012-9332-x.

Mansourian, S., Vallauri, D. and Dudley, N. (2006) 'Forest restoration in landscapes: beyond planting trees', *Landscape Ecology*, 22(3), pp. 477–479. doi: 10.1007/s10980-006-9029-7.

De Meira Junior, M. S. *et al.* (2020) 'The impact of long dry periods on the aboveground biomass in a tropical forests: 20 years of monitoring', *Carbon Balance and Management*. Springer International Publishing, 15(1), pp. 1–14. doi: 10.1186/s13021-020-00147-2.

Meli, P. *et al.* (2017) 'A global review of past land use, climate, and active vs. passive restoration effects on forest recovery', *PLoS ONE*, 12(2), pp. 1–17. doi: 10.1371/journal.pone.0171368.

Messinger, M., Asner, G. P. and Silman, M. (2016) ‘Rapid assessments of amazon forest structure and biomass using small unmanned aerial systems’, *Remote Sensing*, 8(8), pp. 1–15. doi: 10.3390/rs8080615.

Myhre, G. *et al.* (2013) ‘Anthropogenic and Natural Radiative Forcing.’, in Jacob, D., Ravishankara, A. R., and Shine, K. (eds) *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. New York: Cambridge University Press. doi: 10.3390/jmse6040146.

ODNR (2005) *Ohio Abandoned Mine Lands Reforestation Program*. Ohio. Available at: <http://www.ohiodnr.com/mineral/reforestation/tabid/17848/Default.aspx>.

OECD (2020) *Purchasing power parities (PPP)*. Available at: <https://data.oecd.org/conversion/purchasing-power-parities-ppp.htm> (Accessed: 19 October 2020).

PRé Consultants: Life Cycle consultancy and software solutions (2019) *SimaPro 8.3*. Amersfoort, Netherlands: PRé Consultants: Life Cycle consultancy and software solutions. Available at: <https://www.pre-sustainability.com>.

Reforest’Action (2020) *Reforest’Action | I plant my forest*. Available at: <https://www.reforestaction.com/en> (Accessed: 26 June 2020).

Requena Suarez, D. *et al.* (2019) ‘Estimating aboveground net biomass change for tropical and subtropical forests: Refinement of IPCC default rates using forest plot data’, *Global Change Biology*, 25(11), pp. 3609–3624. doi: 10.1111/gcb.14767.

Román-Dañobeytia, F. *et al.* (2021) ‘Survival and early growth of 51 tropical tree species in areas degraded by artisanal gold mining in the Peruvian Amazon’, *Ecological Engineering*. Elsevier B.V., 159(December 2020), p. 106097. doi: 10.1016/j.ecoleng.2020.106097.

De Rosa, M. *et al.* (2017) ‘A flexible parametric model for a balanced account of forest carbon fluxes in LCA’, *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 22(2), pp. 172–184. doi: 10.1007/s11367-016-1148-z.

Rugani, B., Panasiuk, D. and Benetto, E. (2012) ‘An input-output based framework to

evaluate human labour in life cycle assessment’, *International Journal of Life Cycle Assessment*, 17(6), pp. 795–812. doi: 10.1007/s11367-012-0403-1.

Shell (2019) *Shell invests in nature as part of broad drive to tackle CO2 emissions*. Available at: <https://www.shell.com/media/news-and-media-releases/2019/shell-invests-in-nature-to-tackle-co2-emissions.html> (Accessed: 22 October 2020).

Silva, C. V. J. *et al.* (2018) ‘Drought-induced Amazonian wildfires instigate a decadal-scale disruption of forest carbon dynamics’, *Philosophical Transactions of the Royal Society B: Biological Sciences*, 373(1760). doi: 10.1098/rstb.2018.0043.

Silva, C. V. J. *et al.* (2020) ‘Estimating the multi-decadal carbon deficit of burned Amazonian forests’, *Environmental Research Letters*, 15(11), p. 114023. doi: 10.1088/1748-9326/abb62c.

Smith, P., Davis, S. J., Creutzig, F., Fuss, S., Minx, J., Gabrielle, B., Kato, E., Jackson, R. B., Cowie, A., Kriegler, E., van Vuuren, D. P., *et al.* (2016) ‘Biophysical and economic limits to negative CO2 emissions’, *Nature Climate Change*. Nature Publishing Group, 6(1), pp. 42–50. doi: 10.1038/nclimate2870.

Smith, P., Davis, S. J., Creutzig, F., Fuss, S., Minx, J., Gabrielle, B., Kato, E., Jackson, R. B., Cowie, A., Kriegler, E., Van Vuuren, D. P., *et al.* (2016) ‘Biophysical and economic limits to negative CO2 emissions - Supplementary Information’, *Nature Climate Change*, 6(1), pp. 42–50. doi: 10.1038/nclimate2870.

Smith, P. *et al.* (2019) ‘How to measure, report and verify soil carbon change to realize the potential of soil carbon sequestration for atmospheric greenhouse gas removal’, *Global Change Biology*, 26(August), pp. 1–23. doi: 10.1111/gcb.14815.

ter Steege, H. *et al.* (2013) ‘Hyperdominance in the Amazonian Tree Flora’, *Science*, 342(6156), pp. 1243092–1243092. doi: 10.1126/science.1243092.

Stephenson, N. L. *et al.* (2014) ‘Rate of tree carbon accumulation increases continuously with tree size’, *Nature*, 507(7490), pp. 90–93. doi: 10.1038/nature12914.

Tree-Nation (2020) *Tree-Nation - La plateforme mondiale pour planter des arbres*. Available at: <https://tree-nation.com/> (Accessed: 26 June 2020).

- Tree-Nation (2021) *Tree-Nation - Project: Camino Verde*. Available at: 3/28/2021.
- United Nation Decade on Ecosystem Restoration (2021) *UN Decade on Restoration*. Available at: <https://www.decadeonrestoration.org/> (Accessed: 25 May 2021).
- Vance, E. D. and Nadkarni, N. M. (1992) 'Root biomass distribution in a moist tropical montane forest', *Plant and Soil*, 142(1), pp. 31–39. doi: 10.1007/BF00010172.
- Veldman, J. W. *et al.* (2015) 'Tyranny of trees in grassy biomes', *Science*, 347(6221), pp. 484–485. doi: 10.1126/science.347.6221.484-c.
- Verra (2021) *Methodologies - Verra*. Available at: <https://verra.org/methodologies/> (Accessed: 25 May 2021).
- Waring, B. G. and Powers, J. S. (2017) 'Overlooking what is underground: Root:shoot ratios and coarse root allometric equations for tropical forests', *Forest Ecology and Management*. Elsevier B.V., 385, pp. 10–15. doi: 10.1016/j.foreco.2016.11.007.
- Withy, K. *et al.* (2018) 'Quantifying immediate carbon emissions from El Niño-mediated wildfires in humid tropical forests', *Philosophical Transactions of the Royal Society B: Biological Sciences*, 373(1760). doi: 10.1098/rstb.2017.0312.
- Xiao, S. *et al.* (2020) 'Soil Organic Carbon Sequestration and Active Carbon Component Changes Following Different Vegetation Restoration Ages on Severely Eroded Red Soils in Subtropical China', *Forests*, 11(12), p. 1304. doi: 10.3390/f11121304.

CHAPTER 6

Discussion

“What is the use of having developed a science well enough to make predictions if, in the end, all we are willing to do is stand around and wait for them to come true”

SHERWOOD ROWLAND

DISCUSSION

6.1 Key results and testing of hypothesis

The following paragraphs summarise the key results against each Chapter's Hypothesis, described in the Introduction (Section 1.7).

In Chapter 2 on carbonation and enhanced weathering, I show that transporting ground basalt has an important environmental impact, and that there is a maximum distance from quarry to field at which the emissions offset the potential sequestration of the practice. It highlighted the influence of the emission factor of the truck used to transport the material, as well as the share of alkalinity from the dissolved basalt that is transferred to the ocean (ω value) on the offsetting distance, hence on the overall CO₂ sequestration potential of the practice. The assessment showed that accounting for the processes' emissions reduced the C capture potential of the practice claimed in previous studies (**Hypothesis 1** - *The environmental impacts associated with the setup of the practice reduces the sequestration estimates reported in the literature*).

Considering São Paulo State in Brazil, the assessment showed that the application of 1 t ha⁻¹ of crushed basalt on all its 12 Mha could capture 1.3 to 2.4 Mt CO₂e through carbonation and enhanced weathering respectively, including the emissions associated with the implementation of the practice. If the application rate is up-scaled to 50 t ha⁻¹, the practice could offset around 60% (considering 50% carbonation and 50% EW) of the GHG emissions attributed to the state in 2016 (**Hypothesis 2** - *Enhanced weathering using basalt rock in São Paulo state, Brazil, has a high carbon capture potential*).

In Chapter 3, the RothC soil C model was modified to simulate changes in soil C stock with additions of sugarcane-derived biochar and fresh residues. Applied to Brazilian sugarcane systems, the model predicted a substantial increase in soil C stocks when the residues are used for biochar production, exceeding the potential increase in soil C stock when the same residues are applied fresh on the field. Scaling to the total sugarcane area of the State, the application of the biochar produced from available sugarcane residues would capture 50 Mt of CO₂e yr⁻¹, which is 31% of the emissions attributed to the State in 2016 (**Hypothesis 3** - *Biochar produced from available sugarcane residues in São Paulo state, Brazil, has a high carbon sequestration potential*).

CHAPTER 6

In Chapter 4, I compare the global warming impact of using sugarcane residues in a combine heat and power plant with scenarios where the residues are turned into biochar and applied to sugarcane fields. Results of the LCA show that, on average, allocating 100%, 50%, and 25% of the available residues to biochar production can sequester a net 6.3 ± 0.5 , 3.6 ± 0.3 , and 1.8 ± 0.3 t CO₂e ha⁻¹ yr⁻¹, respectively. The C capture arises mainly from the C stored as biochar in the soil, while meeting the electricity deficit created by pyrolysis accounts for most of the emissions to implement the practice. Applied to the state of São Paulo, this practice could lead to the sequestration of 36 Mt CO₂e yr⁻¹ or 23% of the State's GHG emissions in 2016. Hence, accounting for the emissions to implement the practice reduces the sequestration potential of biochar alone by around 25% (**Hypothesis 4** - *Replacement of combined heat and power units by pyrolysis plants producing biochar and energy lowers the carbon sequestration potential of the practice*).

In Chapter 5, I use LCA to assess the global warming impact of reforesting a parcel in the south-eastern Peruvian Amazon, following deforestation caused by mineral extraction. A soil C model (RothC) as well as above- and below-ground biomass C models were used in parallel to predict the C increase over time following reforestation. The LCA of the plot shows that the act of reforesting is a net GHG emitting process (**Hypothesis 5** - *The act of planting trees emits GHG*) but that the production and application of biochar at the time of transplant, can drive the process to be C negative. Acknowledging the emissions associated to setup the reforestation plot, the C stock saturation of a growing forest, and the increasing risks of C capture reversal, the chapter then warns about the instantaneous and perennial C credits attributed by reforestation platforms to investment towards future tree planting projects (**Hypothesis 6** - *The immediate carbon GHG capture credits attributed to reforestation projects are not a representative nor a reliable accounting method*).

6.2 Intellectual contribution and potential impacts of findings

The LCAs on enhanced weathering and biochar (Chapters 2 and 4) emphasize the need to consider GGRTs in their entirety to show both their potential along with limitations and caveats. This is crucial, given that diverse techniques are increasingly promoted as effective and efficient solutions to remove or sequester atmospheric CO₂. The enhanced

weathering chapter further shows that earlier estimates of the large-scale potential of the method were magnified in part given that processing and transport impacts were underestimated.

The main impacts of these LCAs relate to the more objective quantification of the net GHG impacts of the processes and the components and assist on defining the limits that flip the system from net GHG sequestration to net GHG emitting. This has major implications for the global potential of the systems and provides insights and assistance to both the science and policy communities. Overall, the approaches comprehensively support the use of LCA to better quantify the potential impacts of GGRTs and explore the limits and potential caveats of such practices.

Chapter 3 on the biochar RothC sub-model delivers a reliable estimation of the C sequestration potential of biochar according to different soil types and meteorological parameters, at desired range of defined timescales. The main impact of the biochar sub-model is its potential spread to other users due to its ease of use, operation in the open-source *R* software platform, and its addition to a well-known and validated soil C model (RothC). When applied to sugarcane systems in São Paulo State, the results provide decision-makers with insights on the C abatement potential of using an available resource for biochar production and amendment.

The LCA of Chapter 5 recalls that the act of planting trees is *per se* a GHG emitting process and that planted forests have to overcome a C debt before being considered as a GGRT. The assessed system also shows that complementary practices such as biochar allows for the planting tree process to become C negative. Overall, Chapter 5 points out the current difficulties in properly assessing the C removal impact of growing forests due to accounting issues linked to reversibility risks and stock saturation. It suggests the need to agree on a methodology to close this gap.

6.3 Limitation of the work

The main limitation of this work relates to the limited number of GGRTs and contexts analysed. As stated in the Introduction (Section 1.4), a combination of the available GGRT portfolio will be needed to limit the global average temperature increase to 2°C (Mace *et al.*, 2021). The environmental impacts associated with the setup of GGRTs

depend highly on the GGRT studied and the context in which it is implemented. Hence, to ensure their C capture relevance, the global warming impact of GGRTs should always be assessed prior to deployment, and net capture potentials reported with functional unit(s) that allow comparison (Goglio *et al.*, 2020). This work assessed three GGRTs with focuses on São Paulo State in Brazil (Chapters 2 – 4) and a tropical rainforest reforestation plot (Chapter 5). This narrow sample of contexts and GGRTs limits the universality of recommendations across all GGRTs, but provides a solid evidence base to comment on land-based GGRTs.

Nonetheless, Chapters 2 and 4 (enhanced weathering and biochar LCA, respectively) both focus on São Paulo State and present results in an equivalent functional unit (in t CO_{2e} emitted per t CO_{2e} sequestered). Hence, they provide a preview of the necessary assessments to allow honest GGRT deployment recommendation. Based on the results of the assessments, spreading ground basalt on sugarcane fields (0.11 t CO₂ emitted per t CO₂ sequestered, considering 50% EW and 50% carbonation) would be more efficient than implementing pyrolysis to produce biochar using 50% the available sugarcane residues (0.19 t CO₂ emitted per t CO₂ sequestered). However, this limited recommendation only considers two of the manifolds existing GGRTs. To provide complete and objective recommendations for the policy and science communities, this work should be supplemented by LCAs of additional GGRTs, impact categories, and thorough assessment of potential attendant environmental and social impacts. Time constraints, however, made this impossible.

Another limitation of this work relates to the permanence of its validity. Despite the effectiveness of LCA in untangling the environmental impacts associated with inputs and outputs needed to setup a GGR technologies, LCA results are only valid for a limited period, typically a few years. As the technologies or surrounding processes and environment evolve, the LCA results are rendered obsolete because the emission factors, the process chains, or our scientific understanding of biological process have changed, improved, or been transformed since the original assessment (e.g. GHG intensity of electricity production, electrification of transport, process mechanisation). Hence, as GGRTs evolve or as technologies shifts, LCAs will need to be updated for their conclusion to remain relevant. Nevertheless, the methodologies and interrogations that

arise from the C accounting sphere (e.g. accounting for temporary C storage, model C permanence [cf. Chapter 3], reversibility, or leakage) are echoed in LCA debates and the practices put in place today aim to be valid timelessly thus facilitating and increasing relevance and ease of comparison of future assessments.

6.4 Limitations to the deployment of GGRTs and future challenges

Missing context-specific LCAs are not the only factor impeding the deployment of GGRTs. Limitations to their deployment have been discussed extensively in the literature and the main arguments can be summarize in four points: lack of funding, remaining uncertainties regarding attendant effects, lack of regulated C capture accounting methodologies, and difficulties to insert GGRTs portfolio in integrated assessment models (IAMs) and earth system models (ESMs).

6.4.1 Funding

The IPCC AR5 reported that the consequences of reaching a 2.5°C average global increase in temperature compared to pre-industrial era could cost up to 2% of the global annual income (IPCC, 2014). Although they were informed by only medium quality evidence to support this figure, the expected economic impact of climate change induced catastrophes should have caused a surge in investments into GGRTs. However, to date, there are no official regulatory support to offer a C price high enough to promote them, and the existing C pricing schemes mostly penalize CO₂ emissions but do not reward C capture (with the exception of voluntary C offsetting scheme) (Renforth and Wilcox, 2019; Carton *et al.*, 2020). Although the economic cost of setting up most GGRTs has been assessed, official mechanisms that would remunerate captured C should be investigated as well and set up as fast as possible (Fajardy *et al.*, 2019).

6.4.2 Uncertainties from upscaling and potential damage

It is now agreed that no single GGRT could offset the cumulative GHG emissions to accord with the UNFCCC 2°C limits (Renforth and Wilcox, 2019). Nevertheless, the scale at which some GGRTs are still modelled in C neutrality pathways represent massive upscaling of the technologies, reaching extents never proven before and generating scientific doubts on their feasibility (Lenzi, 2018; Rickels *et al.*, 2019; Ruseva *et al.*, 2020). In addition, GGRTs come with their set of attendant risks or uncertainties

(Honegger, Michaelowa and Roy, 2020). Massive upscaling of one technology also increases the potential impact of its attendant effects, which are sometimes very difficult to foresee (Smith *et al.*, 2019; Mace *et al.*, 2021). For instance, tree-planting practices has been linked with a significant reduction in river flow, which could potentially negatively impact surrounding environments or human activities (Bentley and Coomes, 2020).

Accounting for the impacts of GGRTs beyond their C sequestrations potential is crucial to assess their sustainability (Mace *et al.*, 2021). Smith *et al.* (2019) already reviewed the effect of some GGRTs on the SDGs and the creation of an ecosystem services impact category within LCA has been discussed (Zhang, Anil and Bakshi, 2010; Zhang, Singh and Bakshi, 2010). However, this would require complex additional indicators and high number of context-specific and accurate data, which is a challenge for the scientific community (Antón *et al.*, 2016).

6.4.3 Regulated and accepted accountability practices

Proper accountability of the C capture potential of GGRTs is essential for their deployment and incentivisation of practitioners (Brander *et al.*, 2021). However, most GGRTs relying on biological sinks face issues such as sink saturation, leakage or reversal, sometimes alone or in combination (Brack and King, 2020). Biochar, enhanced weathering, and reforestation are all subject to sink saturation (Brack and King, 2020). For biochar and enhanced weathering, the saturation is linked to the maximum amount that can be applied on a surface of land before expecting detrimental effects, associated to the biochar permanence and silicate mineral dissolution rate, respectively (Fuss *et al.*, 2018). For reforestation, the saturation effect is related to the time needed for the trees to reach maturity and the near stabilisation of the C stock of a mature forest plot (IPCC, 2019).

Regarding permanence and reversal, C captured through biochar or enhanced weathering is considered safe and relatively permanent (Fuss *et al.*, 2018) (with an important range for biochar depending on environmental parameters (Wang *et al.*, 2016)). Carbon captured in biomass and soils, however, is at high risk of being released into the atmosphere through natural or anthropogenic disturbances (Brack and King, 2020; Mace *et al.*, 2021). This risk is increased by climate change itself and the impacts it may have

on the severity and likelihood of natural phenomena responsible for the C sink reversal (Griscom *et al.*, 2017; Anderegg *et al.*, 2020).

Overall, the issues of saturation and potential reversal are context-specific and sometimes difficult to model, especially at large scale. Brander *et al.* (2021) recommend that emissions and removal should be reported only where and as they happen, following what they called a “reality principle”. Following this principle, they argue that considering temporary C storage should include the emissions to maintain or replace the temporary sinks endlessly (Brander *et al.*, 2021). One recent paper reviewed LCAs of GGRTs and reported that there were only a few LCA on reforestation as a GGRTs (although there are many on wood products) (Terlouw *et al.*, 2021). The authors promote the use of dynamic LCAs for this specific GGRT (Levasseur, Brandão, *et al.*, 2012), and support their statement by explaining that a wildfire at the end of a plantation would be considered statically in a conventional LCA and could lead to a net zero C capture. However, there would be a long period where the growing trees would have indeed captured C and reduced radiative forcing (Terlouw *et al.*, 2021). Similarly, Ruseva *et al.* (2020) argues that permanence as a key principle of the GGR definition is a important barrier to large deployment of some land-based C removal practices, and that temporary C storage still offers climate benefits (e.g. avoiding overshoot or temporarily avoiding radiative forcing (Brandão *et al.*, 2013; Brander *et al.*, 2021)). A variety of dynamic LCA methodologies exists but each yield very different results (Levasseur, Lesage, *et al.*, 2012; Almeida *et al.*, 2016). Hence, a reliable accounting methodology should emerge fast to provide a framework for dealing with delayed emissions, temporary storage and uncertainties related to leakage or reversal of GGRTs.

Another issue relates to the uncertainties in the C removal potential of the assessed GGRT (Lomax *et al.*, 2015). As additional complex processes are merged into accounting rules, uncertainties around the values rise. Although presenting uncertainty ranges is essential to truthfully represent the potential impact of a scenario, high uncertainty may lower the confidence in the technique. Hence, working towards proper accountability to presents as truthful values as possible could increase distrust in the practices from policymakers and public hence lower their adoptability (Nemet *et al.*, 2018; Brander *et al.*, 2021).

Once the deployment of GGRT starts, another challenge will be to reliably monitor their progress towards net zero GHG emission objective (Mace *et al.*, 2021). To do so, affordable and reliable measurement and verification tools have to be made available which can prove difficult, especially when dealing with land-based sinks (Smith, 2016; Mace *et al.*, 2021).

6.4.4 Clear representation of GGRTs in IAMs

Although the merging of the impact of GGRTs in IAMs should be done with caution (Section 1.4) (Lenzi, 2018; McLaren *et al.*, 2019), a recent survey showed that the number of GGRTs that can be assessed in IAMs or ESMs is quite small due to lack of necessary resolution, absence of representative processes, and inability to represent several GGRTs in combination (although a broad range of GGRTs will be needed – Section 1.3) (Rickels *et al.*, 2019). In addition, as the Earth system has never been through deliberate removal of CO₂, there are many unknowns on how it will react. IAMs and ESMs experts' current knowledge suggest that the net contribution of GGRTs (highly affected by feedbacks and saturation effects in an interacting C cycle), would probably not linearly expand the C budget (Rickels *et al.*, 2019).

6.5 An opportunity for localized actions

The previous paragraphs list the current challenges to the deployment of GGRTs. While agreement on accountability practices and integration of GGRTs in IAMs need time and research, localized deployment of GGRTs could alleviate attendant risks and promote investment. Indeed, while large scale deployment of most GGRTs usually collide with land, water or resources limitations, and risks of leading to detrimental environmental and social effects (Meadowcroft, 2013; Mace *et al.*, 2021). In some contexts, studies have shown that deployment of specific GGRTs can have local beneficial environmental or social ramifications (Buck, 2018; Mace *et al.*, 2021). Identification of these “highly profitable” contexts for GGRT deployment could be made through LCA using an array of impact categories followed by *in situ* social and scientific field work to evaluate all likely associated effects of their implementation and desirability. First, GGRT screening could be based on resources needed and price suitability. As the quantity and quality of resources are distributed unevenly over the globe, local availability and quality of the resources define the type and potential of the GGRT that could be set up. In addition, the

location dictates labour and land costs, taxation and incentive rates and hence inform the GGRT implementation costs (Fajardy *et al.*, 2019).

Shifting from aggregated global calculation of GGRT's feasibility to targeted local assessment of each practice's maximum potential and benefits could motivate investment, improve acceptability, and accelerate deployment of some practices (Fajardy *et al.*, 2019; Thoni *et al.*, 2020). To identify the ideal context for GGRT deployment, and allow comparison between practices, it is critical that all LCAs report their impact values based on a similar functional unit (e.g. global warming impact in t CO₂e emitted per t CO₂ removed) (Goglio *et al.*, 2020; Terlouw *et al.*, 2021) (Section 1.3). To date, LCAs of GGRTs are still scarce and not all technologies are equally represented. According to a recent review, around 90 LCA studies covering biochar, soil C sequestration, BECCS, afforestation/reforestation, direct air capture and C storage, and enhanced weathering have been published (Terlouw *et al.*, 2021).

6.6 Future of GGRTs

There are two main ways in which GGRTs can be used (Meadowcroft, 2013). The first is to consider GGRTs as emission reduction options along with mitigation options, with the objective of slowing down the increase of atmospheric GHG and avoid exceeding a decided limit. In this scenario, GGRTs will only be deployed after all cheaper mitigation options are implemented (Meadowcroft, 2013), although some land-based GGRTs can be cost beneficial in specific context (e.g. considering yield increase following biochar or basalt application). The second possibility is to consider GGRTs as a response to a critical overshoot of the decided limit and to provide a last alternative to lower atmospheric GHG concentration to level deemed acceptable (Meadowcroft, 2013). The first option buys time for the development of a decarbonised world and allows for the potential delay of mitigation practices. It also recognises the long-term maintenance of GGRTs with known reversibility and saturation risks. The second option would probably not be able to repair the harm done during the period preceding the overshoot and would expose the world to the unknown effects of reaching such high concentration of atmospheric GHG and potentially passing climate tipping points (Meadowcroft, 2013). Both options may overestimate our capacity to meddle with the C cycle (Lenzi, 2018), but the first option should be preferred as it reduces the likelihood of reaching tipping

CHAPTER 6

points and their potentially highly deleterious effects while allowing the set up of assessed beneficial GGRTs.

Irrespective of which approach is chosen, predicting the most efficient GGRTs within contexts beforehand can only help in their deployment efficacy and impact. However, in time, the decision of which GGRTs to deploy will not only be a subject of efficiency and benefits, but will also be a matter of cost and social acceptance. As of April 2021, 28 countries have submitted their “long-term low GHG emission development strategies” (UNFCCC, 2021). These strategies are requested (although not mandatory) by the Paris agreement as an addition to the “Nationally Determined Contributions”. GGRTs are part of most of the long-term low GHG emissions development strategies but not all are envisioned in each country. The UK and US keep in view a broad range of GGRT (e.g. BECCS, DAC, EW, Forestry), Germany on the other hand focuses on wetland and forests. This selection is not based on the potential beneficial impact of one GGRT over the other, nor is it representative of the technological feasibility of the selected GGRT in the country but indicates the political or social acceptability of the GGRT (Thoni *et al.*, 2020). Biochar and enhanced weathering, although considered more secure and long term than afforestation/reforestation, are almost absent from most of the “long-term low GHG emission development strategies” while forestry practices are considered by most (Thoni *et al.*, 2020).

The Berlin Mercator Research Institute on Global Commons and Climate Change report that we have 6.5 years before the atmospheric GHG concentration reach the level to overshoot the 1.5°C limit (MCC, 2021). Hence, regardless of the type of GGRTs, their implementation (along with drastic emission reduction measures) should be emphatically supported. Likewise, despite acknowledged difficulties and current methodological limits, context- and GGRT-specific LCAs should be promoted to provide insights on the deployment scale and location for maximised impact. While research should continue studying potential additional GGRT or complement to GGRTs practice (Horton *et al.*, 2021).

6.7 References

Almeida, J. *et al.* (2016) ‘Greenhouse gas emission timing in life cycle assessment and the global warming potential of perennial energy crops’, *Carbon Management*. Taylor &

Francis, 6(5–6), pp. 185–195. doi: 10.1080/17583004.2015.1109179.

Anderegg, W. R. L. L. *et al.* (2020) ‘Climate-driven risks to the climate mitigation potential of forests’, *Science*, 368(6497), p. eaaz7005. doi: 10.1126/science.aaz7005.

Antón, A. *et al.* (2016) ‘Addressing biodiversity and ecosystem services in life cycle assessment’, *Handbook on Biodiversity and Ecosystem Services in Impact Assessment*, pp. 140–164. doi: 10.4337/9781783478996.00012.

Bentley, L. and Coomes, D. A. (2020) ‘Partial river flow recovery with forest age is rare in the decades following establishment’, *Global Change Biology*, 26(3), pp. 1458–1473. doi: 10.1111/gcb.14954.

Brack, D. and King, R. (2020) ‘Managing Land-based CDR: BECCS, Forests and Carbon Sequestration’, *Global Policy*. doi: 10.1111/1758-5899.12827.

Brandão, M. *et al.* (2013) ‘Key issues and options in accounting for carbon sequestration and temporary storage in life cycle assessment and carbon footprinting’, *The International Journal of Life Cycle Assessment*, 18(1), pp. 230–240. doi: 10.1007/s11367-012-0451-6.

Brander, M. *et al.* (2021) ‘Carbon accounting for negative emissions technologies’, *Climate Policy*. Taylor & Francis, 0(0), pp. 1–19. doi: 10.1080/14693062.2021.1878009.

Buck, H. J. (2018) ‘The politics of negative emissions technologies and decarbonization in rural communities’, *Global Sustainability*, 1, pp. 1–7. doi: 10.1017/sus.2018.2.

Carton, W. *et al.* (2020) ‘Negative emissions and the long history of carbon removal’, *WIREs Climate Change*, 11(6), pp. 1–25. doi: 10.1002/wcc.671.

Fajardy, M. *et al.* (2019) ‘Negative Emissions: Priorities for Research and Policy Design’, *Frontiers in Climate*, 1(October), pp. 1–7. doi: 10.3389/fclim.2019.00006.

Fuss, S. *et al.* (2018) ‘Negative emissions — Part 2: Costs , potentials and side effects’, *Environmental Research Letters*, 13 063002(063002). doi: 10.1088/1748-9326/aabf9f.

Goglio, P. *et al.* (2020) ‘Advances and challenges of life cycle assessment (LCA) of greenhouse gas removal technologies to fight climate changes’, *Journal of Cleaner Production*. Elsevier Ltd, 244, p. 118896. doi: 10.1016/j.jclepro.2019.118896.

CHAPTER 6

Griscom, B. W. *et al.* (2017) 'Natural climate solutions', *Proceedings of the National Academy of Sciences of the United States of America*, 114(44), pp. 11645–11650. doi: 10.1073/pnas.1710465114.

Honegger, M., Michaelowa, A. and Roy, J. (2020) 'Potential implications of carbon dioxide removal for the sustainable development goals', *Climate Policy*. Taylor & Francis, 0(0), pp. 1–21. doi: 10.1080/14693062.2020.1843388.

Horton, P. *et al.* (2021) 'Technologies to deliver food and climate security through agriculture', *Nature Plants*. Springer US, 7(3), pp. 250–255. doi: 10.1038/s41477-021-00877-2.

IPCC (2014) *Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, IPCC. Edited by Core Writing Team, R. K. Pachauri, and L. A. Meyer. Geneva Switzerland. Available at: https://www.ipcc.ch/site/assets/uploads/2018/05/SYR_AR5_FINAL_full_wcover.pdf.

IPCC (2019) *Climate Change and Land - An IPCC Special Report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems - Summary for Policymakers*, IPCC. doi: 10.4337/9781784710644.00020.

Lenzi, D. (2018) 'The ethics of negative emissions', *Global Sustainability*, 1. doi: 10.1017/sus.2018.5.

Levasseur, A., Lesage, P., *et al.* (2012) 'Assessing temporary carbon sequestration and storage projects through land use, land-use change and forestry: comparison of dynamic life cycle assessment with ton-year approaches', *Climatic Change*, 115(3–4), pp. 759–776. doi: 10.1007/s10584-012-0473-x.

Levasseur, A., Brandão, M., *et al.* (2012) 'Valuing temporary carbon storage', *Nature Climate Change*. Nature Publishing Group, 2(1), pp. 6–8. doi: 10.1038/nclimate1335.

Lomax, G. *et al.* (2015) 'Reframing the policy approach to greenhouse gas removal technologies', *Energy Policy*. Elsevier, 78, pp. 125–136. doi: 10.1016/j.enpol.2014.10.002.

Mace, M. J. *et al.* (2021) *Governing large-scale carbon dioxide removal: are we ready? - an update*. New York. Available at: <https://www.c2g2.net/governing-large-scale-carbon-dioxide-removal-are-we-ready/>.

MCC (2021) *Remaining carbon budget*. Available at: <https://www.mcc-berlin.net/en/research/co2-budget.html>.

McLaren, D. P. *et al.* (2019) ‘Beyond “Net-Zero”: A Case for Separate Targets for Emissions Reduction and Negative Emissions’, *Frontiers in Climate*, 1(August), pp. 1–5. doi: 10.3389/fclim.2019.00004.

Meadowcroft, J. (2013) ‘Exploring negative territory Carbon dioxide removal and climate policy initiatives’, *Climatic Change*, 118(1), pp. 137–149. doi: 10.1007/s10584-012-0684-1.

Nemet, G. F. *et al.* (2018) ‘Negative emissions — Part 3 : Innovation and upscaling’, *Environmental Research Letters*.

Renforth, P. and Wilcox, J. (2019) ‘Specialty Grand Challenge: Negative Emission Technologies’, *Frontiers in Climate*, 1(May), pp. 1–4. doi: 10.3389/fclim.2019.00001.

Rickels, W. *et al.* (2019) ‘(Mis)conceptions about modeling of negative emissions technologies’, *Environmental Research Letters*. IOP Publishing, 14(10), p. 104004. doi: 10.1088/1748-9326/ab3ab4.

Ruseva, T. *et al.* (2020) ‘Rethinking standards of permanence for terrestrial and coastal carbon: implications for governance and sustainability’, *Current Opinion in Environmental Sustainability*, 45, pp. 69–77. doi: 10.1016/j.cosust.2020.09.009.

Smith, P. (2016) ‘Soil carbon sequestration and biochar as negative emission technologies’, *Global Change Biology*, 22(3), pp. 1315–1324. doi: 10.1111/gcb.13178.

Smith, P. *et al.* (2019) ‘Land-Management Options for Greenhouse Gas Removal and Their Impacts on Ecosystem Services and the Sustainable Development Goals’, *Annual Review of Environment and Resources*, 44(1), pp. 255–286. doi: 10.1146/annurev-environ-101718-033129.

Terlouw, T. *et al.* (2021) ‘Life cycle assessment of carbon dioxide removal technologies:

CHAPTER 6

a critical review', *Energy & Environmental Science*. Royal Society of Chemistry, (February). doi: 10.1039/d0ee03757e.

Thoni, T. *et al.* (2020) 'Deployment of Negative Emissions Technologies at the National Level: A Need for Holistic Feasibility Assessments', *Frontiers in Climate*, 2(November). doi: 10.3389/fclim.2020.590305.

UNFCCC (2021) *Communication of long-term strategies*. Available at: <https://unfccc.int/process/the-paris-agreement/long-term-strategies> (Accessed: 9 April 2021).

Wang, J. *et al.* (2016) 'Biochar stability in soil: Meta-analysis of decomposition and priming effects', *GCB Bioenergy*, 8(3), pp. 512–523. doi: 10.1111/gcbb.12266.

Zhang, Y. I., Anil, B. and Bakshi, B. R. (2010) 'Accounting for ecosystem services in life cycle assessment part II: Toward an ecologically based LCA', *Environmental Science and Technology*, 44(7), pp. 2624–2631. doi: 10.1021/es900548a.

Zhang, Y. I., Singh, S. and Bakshi, B. R. (2010) 'Accounting for ecosystem services in life cycle assessment part I: A critical review', *Environmental Science and Technology*, 44(7), pp. 2232–2242. doi: 10.1021/es9021156.

CHAPTER 7

Conclusion

“Global warming isn't a prediction. It is happening.”

JAMES HANSEN

CONCLUSION

This work provides first evidence of C capture potential impact of accounting for the emissions associated to setup the enhanced weathering (Chapter 2) and biochar (Chapter 4) practices in São Paulo State, Brazil. It shows that accounting for the emissions to set up the practices significantly reduces their C capture potential (around 10% for EW and 20% for biochar in the assessed context, Section 6.3) but also highlights the most emitting process inside each system and identifies context-specific limits to the implementation of the assessed GGRTs. In addition, Chapter 2 and 4 show that using a standardized functional unit (i.e. t CO₂e emitted by t CO₂ captured) to assess the impact of two different GGRTs over the same area, enables critical comparison between the practices and provide first evidence for most impactful practices to set up in São Paulo State. Chapter 3 and 5, on the other hand, applies and provides C accounting methodologies in the context of sugarcane residue biochar in São Paulo State and reforestation in south-eastern Peru, and discusses their relevance.

Overall, this work promotes the use of LCA as a context-specific tool to identify the most efficient GGRTs and maximise their C capture potential. Indeed, as GGRTs are expected to play a major role amid the race to prevent the dire consequences of climate change, assessing their impact and foreseeing their limitations is essential to guarantee their efficiency. Although LCA require additional context-specific data and advances on other C accounting methodologies to provide reliable comprehensive recommendations on GGRTs, it already possesses the necessary set of tools to consistently assess their potential and should be promoted as such to identify potentially impactful contexts.

As the delay of emissions mitigation measures increases our ultimate reliance on GGRTs, LCA represents the best available tool to provide policymakers with critical insights on most efficient context-specific type and scale of GGRTs.

APPENDICES

Appendix A Supplementary Information – Chapter 2: Assessing the potential of soil carbonation and enhanced weathering through Life Cycle Assessment: a case study for São Paulo State, Brazil

A.1 Foreword

The analysis presented here considers the uncertainties associated with the emissions and the sequestration potential through carbonation and enhanced weathering of one tonne of Basalt through Monte Carlo simulations. The Monte Carlo simulations were made using R software with 10,000 iterations. This document first describes the uncertainty analyses for the emissions part, then follows with the sequestration potential, and ends with the combination of the two and their combined effects on the offsetting distance.

A.2 Data used for the setup of the Monte Carlo simulations

A.2.1 Basalt blasting

Weight of Explosive		Comments
Reported in the LCA	0.145 kg /t of basalt	
Error range	0.0725 – 0.29 kg /t of basalt	Arbitrary
Distribution	Uniform	
Emission factor	5.37 kg CO _{2e} /kg of explosive	From SimaPro 8.3

A.2.2 Loader operation

Volume of Diesel		Comments
Drill Rig & Compressor		
Reported in the LCA	0.159 l /t of basalt	
Error range	0.180 – 0.195 l /t of basalt	Interview with quarry owner (Rosado <i>et al.</i> , 2017)
Distribution	Uniform	
Emission factor	3.2 kg CO _{2e} /l of diesel	From SimaPro 8.3
Front Loader		

Reported in the LCA	0.1875 l /t of basalt	
Error range	0.156 – 0.162 l /t of basalt	Interview with quarry owner (Rosado <i>et al.</i> , 2017)
Distribution	Uniform	
Emission factor	3.2 kg CO ₂ e /l of diesel	From SimaPro 8.3

A.2.3 Transportation

Tortuosity Factor		Comments
Reported in the LCA	1.3	
Error range	1.1 – 1.59	30 measurements in Google Earth
Distribution	Triangular	
Min	1.1	Min of the 30 measurements
Max	1.59	Max of the 30 measurements
Mode	1.3	Mean of the 30 measurements

We decided to compare the offsetting distances according to different truck size. Their emission factors were taken from GOV.UK website (Department for Business, 2018). We assumed the truck to be fully loaded (100% laden)

Emission Factor / Truck Type considered	
Rigid (>3.5 - 7.5 tonnes)	0.258 kg CO ₂ e /tkm
Rigid (>7.5 - 17 tonnes)	0.150 kg CO ₂ e /tkm
Rigid (>17 tonnes)	0.119 kg CO ₂ e /tkm
Articulated (>3.5 – 33 tonnes)	0.0772 kg CO ₂ e /tkm
Articulated (>33 tonnes)	0.0609 kg CO ₂ e /tkm

A.2.4 Crushing

Vibrating Feeder		Comments
Reported in the LCA	0.176 kWh /t of basalt	
Error	0.0132 – 0.176 kWh /t of basalt	(4 observations) As reported in Rosado <i>et al.</i>
Distribution	Lognormal	following Weidema <i>et al.</i> (2013)

Mean of the distribution on the log scale	-1.816	As required for lognormal distribution in R (Based on the number reported in the LCA)
Standard deviation on the log scale	0.3977	As required for lognormal distribution in R (Based on the range provided by Rosado <i>et al.</i> (2017))
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3

Primary Crusher		Comments
Reported in the LCA	0.661 kWh /t of basalt	
Error	/	/
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3

Secondary Crusher		Comments
Reported in the LCA	0.539 kWh /t of basalt	
Error	/	/
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3

Tertiary Crusher		Comments
Reported in the LCA	0.79 kWh /t of basalt	
Error	/	/
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3
Conveyor Belt		Comments
Reported in the LCA	0.264 kWh /t of basalt	
Error	0.0176 – 0.264 kWh /t of basalt	(7 observations) As reported in Rosado <i>et al.</i> (2017)
Distribution	Lognormal	following Weidema <i>et al.</i> (2013)
Mean of the distribution on the log scale	-1.384	As required for lognormal distribution in R (Based on the number reported in the LCA)

Appendix A

Standard deviation on the log scale	0.3259	As required for lognormal distribution in R (Based on the range provided by Rosado <i>et al.</i> (2017))
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3

Vibrating Screen		Comments
Reported in the LCA	0.176 kWh /t of basalt	
Error	0.0132 – 0.176 kWh /t of basalt	(2 observations) As reported in Rosado <i>et al.</i> (2017)
Distribution	Lognormal	following Weidema <i>et al.</i> (2013)
Mean of the distribution on the log scale	-1.752	As required for lognormal distribution in R (Based on the number reported in the LCA)
Standard deviation on the log scale	0.1754	As required for lognormal distribution in R (Based on the range provided by Rosado <i>et al.</i> (2017))
Emission Factor	0.392 kg CO ₂ e /kWh	From SimaPro 8.3

A.2.5 Spreading

Diesel use for basalt spreading		Comments
Reported in the LCA	0.577 kg /t of basalt	Calculated (Crop_LCA)
Error	0.494 – 4.94 kg /t of basalt	(9 observations) As reported in literature*
Distribution	Lognormal	following Weidema <i>et al.</i> (2013)
Mean of the distribution on the log scale	-1.650	As required for lognormal distribution in R (Based on the number reported in the LCA)
Standard deviation on the log scale	1.4835	As required for lognormal distribution in R (Based on the range provided in literature)
Emission Factor	0.624 kg CO ₂ e /kg diesel	From SimaPro 8.3

*(Koga *et al.*, 2003; Grisso *et al.*, 2010; Handler and Nadlinger, 2012; Lutz Böhle, 2014) for lime spreading and mineral fertilizer spreading.

A.3 Sum of all emissions per type of truck

When considering the average quarry to field distance, uncertainty is mainly driven by the emission factor of the truck used for transportation while the combined errors of all other parameters lead to a relative standard deviation of less than 5% (Figure A.1.)

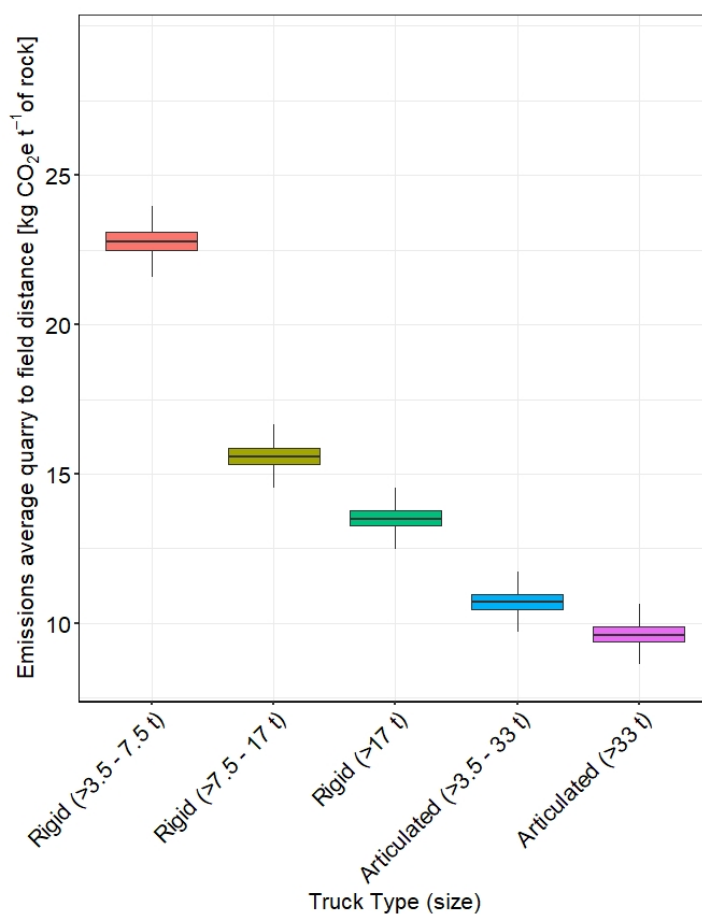


Figure A.1: Average emissions and standard deviation from extraction to application of one tonne of basalt for different type of trucks and considering an average quarry to field distance of 50km in straight line.

Note: In the LCA, the truck used from EcoInvent had an emission factor of 0.168 kg CO₂ /tkm leading to a total emission of 16.8 kg CO₂e /t of basalt (average quarry to field distance). The “Rigid (>7.5 – 17 t)” has the closest emission factor (0.14999 kg CO₂e /tkm) and reaches 15.8 ± 0.46 kg CO₂ /t of basalt.

A.4 Carbon sequestration - Introduction (Theoretical note)

A.4.1 Sequestration through carbonation

The potential CO₂ capture through carbonation is calculated using the CaO and MgO content of the rock and assuming the precipitation of the associated cations (i.e. Ca²⁺ and Mg²⁺) into carbonate minerals (i.e. calcite and magnesite). It is calculated using the equation (2-5) from Renforth, (2012).

A.4.2 Sequestration through enhanced weathering

CO₂ capture through enhanced weathering assumes that all the cations, resulting from the dissolution of the rock, remain dissolved and are transferred to the ocean. From the equation (2-5), only ω changes. According to Kheshgi, (1995), when all cation remains dissolved, the value of ω reaches 1.79 which means that 1.79 moles of CO₂ is sequestered per mole of CaO or MgO. A value of 1.7 for ω was used in other publications on enhanced weathering (Harvey, 2008; Renforth, 2012). Overall, the widely used sequestration value through enhanced weathering of 0.3 t CO₂/t of basalt (Kantola *et al.*, 2017; Beerling *et al.*, 2018; Streffler *et al.*, 2018) results from this equation using average values for CaO and MgO content and a value of 1.7 for ω .

A.5 Methodology

To compute the uncertainty associated with the sequestration potential of basalt through enhanced weathering and carbonation in São Paulo, we used 48 CaO and MgO contents of basalt rock belonging to the Parana Continental Magmatic Province in Brazil (Baggio, Hartmann and Bello, 2016; Braz Machado *et al.*, 2018; Moraes, Seer and Marques, 2018).

We tested these 48 paired observations through Shapiro Wilk: MgO and CaO content are normally distributed. Both data for CaO and MgO content showed a skewness and kurtosis relatively close to 0 which strengthen our assumption to consider them normally distributed.

CaO and MgO content appeared correlated. To confirm this, we used the (cor.test(x,y) in R) which test using the “Pearson’s product-moment correlation”, as recommended when the data are normally distributed, when there are no outliers, and when there are sufficient

observations. The correlation coefficient obtained was 0.936 and was taken into account in the Monte Carlo analysis when randomly sampling from the two normal distributions.

Each Monte Carlo was made for a ω value ranging from 1 (carbonation) to 1.7 by increment of 0.1 and a final and max value of $\omega = 1.79$ (maximum EW).

A.6 Results

The following figure shows the sequestration potential trend in t CO₂e /t of basalt according to different values of for ω (above) and the distribution curve for $\omega = 1$ (bottom left) and for $\omega = 1.79$ (bottom right) representing the sequestration potential through carbonation and enhanced weathering, respectively.

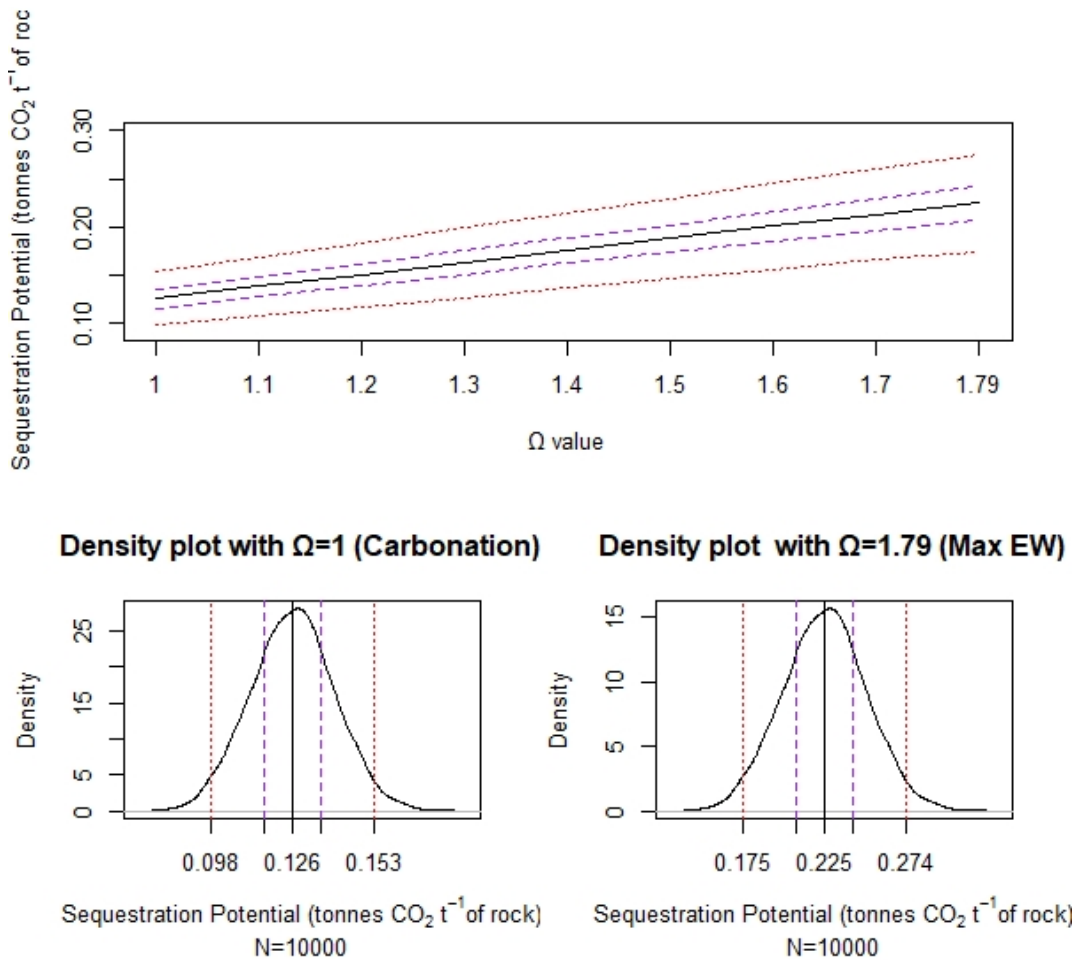


Figure A.2: Above: sequestration potential trend in t CO₂e /t of basalt for increasing value of ω . Bottom: distribution curve with $\omega = 1$ (left) and $\omega = 1.79$ (right) representing carbonation and enhanced weathering, respectively. The Dotted purple line represents the 25 & 75th percentile and the red line dotted line, the 2.5 & 97.5th percentile.

A.7 Discussion

The Monte Carlo simulation shows that as the ω value increases from 1 to 1.79, the standard deviation on the distribution increases. However, this increase is only due to the increasing sequestration value as the relative standard deviation is equal in both values (11.32%).

The relative standard deviation is the result of the division of the standard deviation by the mean value multiplied by 100 to have it in percent. A Relative Standard Deviation (or Coefficient of variation) of 11.32% means that in both cases the “regular” standard deviation is 11.32% of the mean.

In this case it shows that, despite what can be seen in the figure, the increase in ω does not mean that the values are increasingly spread around the mean but that the spread stays the same. The “regular” standard deviation value increases proportionally with the mean

A.8 Effect of truck type and omega (ω) value on offsetting distances.

The truck type and the ω value are the main factors influencing the emissions and sequestration potential of the practice, respectively.

To combine both of these factors we decided to have a look at how they concomitantly influence the offsetting distance (i.e. the distance [km] at which the emissions offset the sequestration potential).

The formula to calculate the offsetting distance is the following:

$$\begin{aligned} & \text{Offsetting Distance} \\ &= \frac{\text{Potential Sequestration} - \text{Emission}_{(\text{Blasting} + \text{Crushing} + \text{Loader Operation} + \text{Spreading})}}{\text{Emission Factor}_{\text{Truck_type}} * \text{Tortuosity ratio}} \end{aligned} \quad (\text{A-1})$$

A.9 Result

The graph hereunder shows the effect of the type of truck and the ω value on the offsetting distance. The offsetting distance of the practice using a fully loaded small truck (high emission factor) and considering carbonation is very low. While the offsetting distance of the practice using a fully loaded big truck (low emission factor) and considering full transport of the dissolved cations to the ocean (EW) is sensibly bigger.

Errors of the other parameters (MgO & CaO content; tortuosity ratio, emissions through blasting, crushing, loader operation, spreading) are taken into account in this graph.

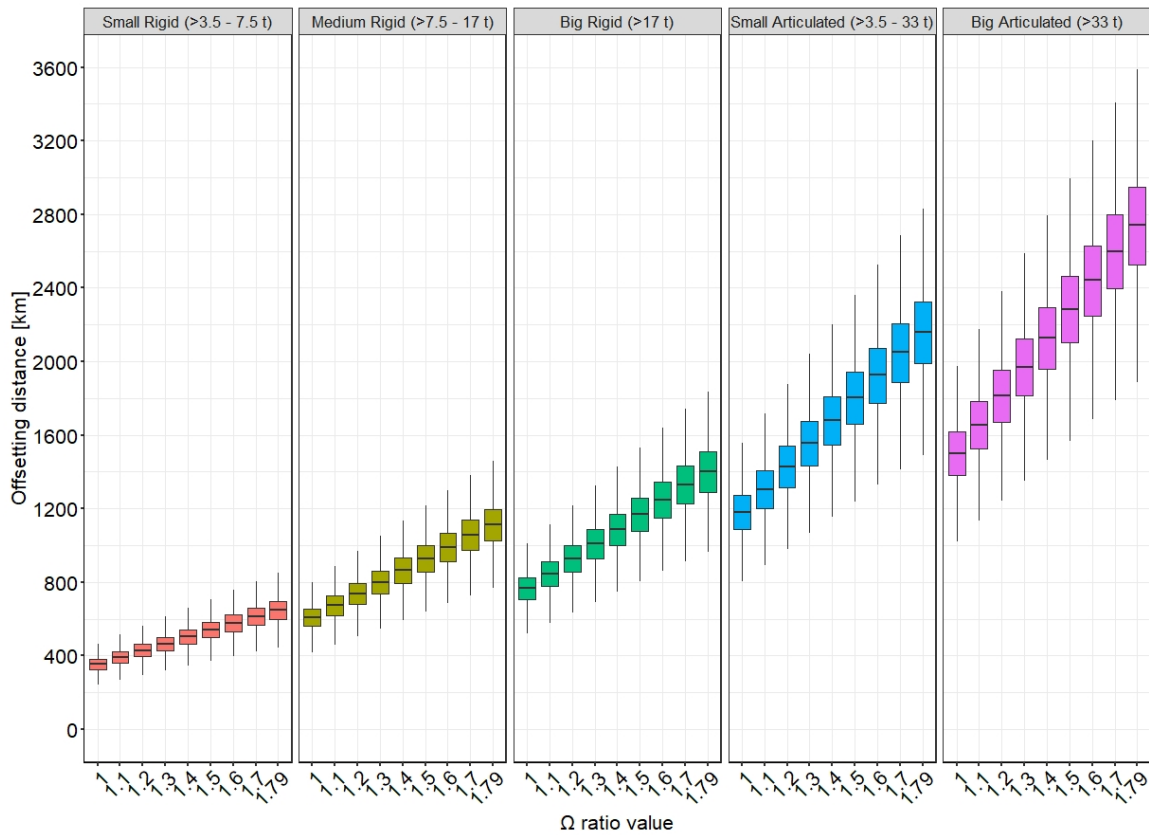


Figure A.3: Effect of Emission Factor (truck type) and ω value on the offsetting distance. The ω value of 1 and 1.79 represent Carbonation and EW, respectively. The error bars represent the propagation of the uncertainty on emissions associated with all the other processes as well as the uncertainty on sequestration potential according to the ω value.

This graph highlights the impact of the most uncertain parameters; i) what percentage of the cations will be transferred to the ocean; and ii) which truck will be use to set up the practice

A.10 References

Baggio, S. B., Hartmann, L. A. and Bello, R. M. S. (2016) 'Paralavas in the cretaceous paran volcanic province, Brazil - a genetic interpretation of the volcanic rocks containing phenocrysts and glass', *Anais da Academia Brasileira de Ciencias*, 88(4), pp. 2167–2193. doi: 10.1590/0001-3765201620150088.

Beerling, D. J. *et al.* (2018) 'Farming with crops and rocks to address global climate, food and soil security', *Nature Plants*, 4(3), pp. 138–147. doi: 10.1038/s41477-018-0108-y.

Braz Machado, F. *et al.* (2018) 'Geochemistry of the Northern Paraná Continental Flood Basalt (PCFB) Province: implications for regional chemostratigraphy', *Brazilian Journal of Geology*, 48(2), pp. 177–199. doi: 10.1371/journal.pone.0018728.

Department for Business, E. & I. S. (2018) *Greenhouse gas reporting: conversion factors 2018*. Available at: <https://www.gov.uk/government/publications/greenhouse-gas-reporting-conversion-factors-2018> (Accessed: 6 December 2018).

Grisso, R. *et al.* (2010) 'Predicting Diesel Fuel Consumption', *Fuel*, (June), pp. 2004–2006.

Handler, F. and Nadlinger, M. (2012) *Strategies for saving fuel with tractors*.

Harvey, L. D. D. (2008) 'Mitigating the atmospheric CO₂ increase and ocean acidification by adding limestone powder to upwelling regions', *Journal of Geophysical Research: Oceans*, 113(4), pp. 1–21. doi: 10.1029/2007JC004373.

Kantola, I. B. *et al.* (2017) 'Potential of global croplands and bioenergy crops for climate change mitigation through deployment for enhanced weathering', *Biology Letters*, (13). doi: <http://dx.doi.org/10.1098/rsbl.2016.0714>.

Kheshgi, H. S. (1995) 'Sequestering Atmospheric Carbon Dioxide by Increasing Ocean Alkalinity', *Energy*, 20(9), pp. 915–922. doi: 0360-5442(95)00035-6.

Koga, N. *et al.* (2003) 'Fuel consumption-derived CO₂ emissions under conventional and reduced tillage cropping systems in northern Japan', *Agriculture, Ecosystems and Environment*, 99(1–3), pp. 213–219. doi: 10.1016/S0167-8809(03)00132-4.

Lutz Bühle (2014) *Biological and chemical parameters and life cycle assessment of the integrated generation of solid fuel and biogas from biomass*.

Moraes, L. C. de, Seer, H. J. and Marques, L. S. (2018) 'Geology, geochemistry and petrology of basalts from Paraná Continental Magmatic Province in the Araguari, Uberlândia, Uberaba and Sacramento regions, Minas Gerais state, Brazil', *Brazilian Journal of Geology*, 48(0), pp. 221–241. doi: 10.1590/2317-4889201820170091.

Renforth, P. (2012) ‘The potential of enhanced weathering in the UK’, *International Journal of Greenhouse Gas Control*. Elsevier Ltd, 10, pp. 229–243. doi: 10.1016/j.ijggc.2012.06.011.

Strefler, J. *et al.* (2018) ‘Potential and costs of carbon dioxide removal by enhanced weathering of rocks’, *Environmental Research Letters*, 13(3), p. 034010. doi: 10.1088/1748-9326/aaa9c4.

Weidema, B. P. *et al.* (2013) *Data quality guideline for the ecoinvent database version 3*. Available at: <http://www.ecoinvent.org/database/methodology-of-ecoinvent-3/methodology-of-ecoinvent-3.html>.

Appendix B Supplementary Information – Chapter 3: Modelling the potential for soil carbon sequestration using biochar from sugarcane residues in Brazil

B.1 Meteorological and soil parameters

Characteristics of the soil types covering São Paulo state's sugarcane area were obtained from the Harmonized World Soil Database (FAO, 2012) (Table B.1).

Table B.1: Main characteristics of the soil type selected as given by the harmonized world soil database (FAO, 2012).

Parameter	Soil 1	Soil 2	Soil 3	Soil 4	Soil 5	Soil 6	Soil 7
Soil Unit Symbol (FAO 90)	FRr	LXh	FRr	FRh	ACf	ARo	ACf
Soil Unit Name (FAO 90)	Rhodic Ferralsols	Haplic Lixisols	Rhodic Ferralsols	Haplic Ferralsols	Ferric Acrisols	Ferrali ^c Arenosols	Ferric Acrisols
Top Soil Texture	Fine	Coarse	Coarse	Fine	Coarse	Coarse	Medium
Topsoil Sand Fraction (%)	13	91	84	52	88	92	67
Topsoil Silt Fraction (%)	25	3	4	7	2	3	7
Topsoil Clay Fraction (%)	62	6	12	41	10	5	26
Topsoil USDA Texture Classification	Clay (heavy)	Sand	Loamy sand	Sandy clay	Loamy sand	Sand	Sandy clay loam
Topsoil Bulk Density (kg/dm ³)	1.20	1.49	1.44	1.45	1.52	1.52	1.40
Topsoil Organic Carbon (% weight)	1.20	0.30	0.67	1.90	0.60	0.84	0.97

Topsoil refers to the 0 – 30 cm depth layer

Appendix B

Latitude, temperature and precipitation data from 10 meteorological stations representing São Paulo state sugarcane area were obtained from the CLIMWAT database (FAO, 2006) (Table B.2).

Table B.2: Latitude, longitude, yearly average temperature and monthly precipitation values of the different meteorological stations considered from ClimWat database (FAO, 2006).

Meteorological Station	Latitude (degree)	Longitude (degree)	Average Temperature (°C)	Average Precipitation (mm/month)	Average Evapotranspiration (mm/month) *
Pocos de Caldas	-21.85	-46.56	18.3	141.5	93.0
Franca	-20.55	-47.43	19.1	135.3	95.4
Frutal	-20.03	-48.93	24.0	127.5	140.9
Tres Lagoas	-20.78	-51.63	24.6	108.7	151.8
Ribeirao Preto	-21.18	-47.71	20.4	128.8	104.9
Piracicaba	-22.71	-47.63	20.7	103.4	108.3
Bauru	-22.31	-49.06	20.7	121.4	108.2
Avare	-23.1	-48.91	20.8	110.2	108.6
Londrina	-23.38	-51.18	21.3	135.2	113.4
Campinas	-22.9	-47.08	19.7	116.2	100.9

*Thornthwaite potential evapotranspiration was calculated using the R software package “SPEI” (Beguería and Vicente-Serrano, 2017) and divided by 0.75, as suggested by the RothC user guide (Coleman and Jenkinson, 2014).

Out of 70 potential soil type/meteorological station combination, only 36 actually represented in São Paulo State (Table B.3).

Table B.3: The actual existing soil type/meteorological station combinations present in the state of São Paulo.

	Soil 1	Soil 2	Soil 3	Soil 4	Soil 5	Soil 6	Soil 7
Tres Lagoas	X	X	X				
Pocos de Caldas	X			X	X		
Ribeirao Preto	X	X	X	X	X		
Piracicaba			X	X	X	X	

	Soil 1	Soil 2	Soil 3	Soil 4	Soil 5	Soil 6	Soil 7
Londrina	X	X	X				
Frutal	X	X	X				
Franca	X				X		
Campinas			X	X	X		
Bauru	X	X	X	X		X	
Avare	X		X		X	X	X

The spatial distribution of the different soil types and sugarcane fields as well as the locations of the meteorological stations are shown in Figure B.1.

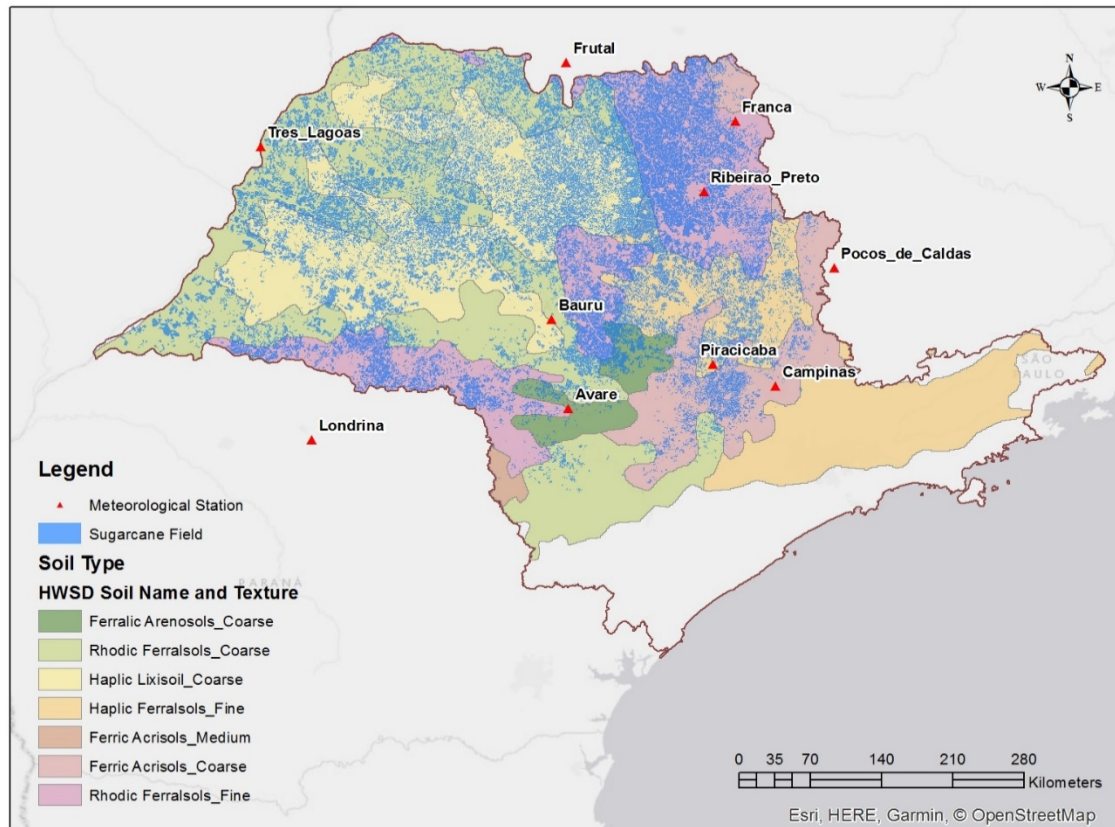


Figure B.1: The study area (State of São Paulo – Brazil) with; (i) the 7 different soil types according to the Harmonized World Soil Database (FAO, 2012), (ii) the location of the meteorological stations considered in the study (FAO, 2006) and (iii) the sugarcane fields according to the CANASAT project (Theodor Rudorff, 2014). Map generated using ArcGIS 10.5.1 (ESRI, 2011).

Table B.4: Parameter settings for soil carbon and biochar production calculations.

Parameter	Value	Unit	Reference
Area in Sugarcane in ha (2014 - all form)	5,768,172	ha	(Theodor Rudorff, 2014)
Estimated sugarcane yield in São Paulo (2018-19)	74,037	TC* ha ⁻¹	(CONAB, 2018)
Filter Cake yearly addition	0.733	tonnes ha ⁻¹	(Cardoso <i>et al.</i> , 2019)
Vinasse yearly addition	31	m ³ ha ⁻¹	(Cardoso <i>et al.</i> , 2019)
Filter Cake Carbon content	32.5	%	(George <i>et al.</i> , 2010)
Vinasse carbon content	1.99	kg m ³	(de Oliveira <i>et al.</i> , 2013)
Average Bagasse production (dm)	140	kg dm TC ^{-1*}	(Waldheim, Monis and Verde Leal, 2001; Khatiwada <i>et al.</i> , 2016)
Bagasse Carbon content	44.6	%	(Hassuani, Leal and Macedo, 2005)
Average trash per tonne of cane (dm)	140	kg dm TC ^{-1*}	(Waldheim, Monis and Verde Leal, 2001; Hassuani, Leal and Macedo, 2005)
Trash losses in sugarcane harvest	5	% TC*	(Sampaio <i>et al.</i> , 2019)
Trash Carbon content	45.27	%	(Hassuani, Leal and Macedo, 2005; Galdos <i>et al.</i> , 2009)
Amount of trash to be left on field	7	tonnes dm ha ⁻¹	(Hassuani, Leal and Macedo, 2005; Carvalho <i>et al.</i> , 2017; Vasconcelos <i>et al.</i> , 2018; Silva <i>et al.</i> , 2019; Popin <i>et al.</i> , 2020)
Yearly carbon input from sugarcane root system	1.88	tonnes ha ⁻¹	(Carvalho <i>et al.</i> , 2017)
Yearly carbon input from root exudates	65	% of root C content	(Bolinder <i>et al.</i> , 2007)
Sugarcane trash biochar (550°C) C content (% dm)	41.01	%	(Cross and Sohi, 2013)
Sugarcane bagasse biochar (550°C) C content (% dm)	63.78	%	(Cross and Sohi, 2013)
Bagasse Biochar production Yield (550°C)	31.3	%	(Quirk <i>et al.</i> , 2012)

Parameter	Value	Unit	Reference
Trash Biochar production Yield (550°C)	33.6	%	(Quirk <i>et al.</i> , 2012)
Biochar weight considered labile	3	%	(Wang <i>et al.</i> , 2016)
Biochar weight considered recalcitrant	97	%	(Wang <i>et al.</i> , 2016)
Sugarcane biochar C lost after 100 years (525°C)	11.9	%	(Zimmerman, 2010)
Soil thickness for modelling activities	30	cm	(FAO, 2012)
DPM:RPM ratio for modelling activities	1.44	-	(Coleman and Jenkinson, 2014)

* TC = tonne of cane

B.2 Calculation – biochar production (example for Scenario 1)

In Scenario 1, 1.29 and 4.62 t C ha⁻¹ yr⁻¹ of the available trash and bagasse respectively (i.e. 100% of each) are supplied to the pyrolysis plant. These figures correspond to 2.85 t and 10.365 t dm of trash and bagasse, respectively, ha⁻¹ yr⁻¹ (see carbon content of trash and bagasse Table B.4).

From there, the equation is:

$$TotalBCC = Wt(dm) * tBCY * tBCCc + Wb(dm) * bBCY * bBCCc \quad (B-1)$$

With:

TotalBCC: total biochar carbon applied [t biochar C ha⁻¹ yr⁻¹]

Wt(dm): trash dry weight available [t ha⁻¹ yr⁻¹]

tBCY: trash biochar production yield [%]

tBCCc: trash biochar carbon content [%]

Wb(dm): bagasse dry weight available [t ha⁻¹ yr⁻¹]

bBCY: bagasse biochar production yield [%]

bBCCc: bagasse biochar carbon content [%]

Using values for scenario 1 and from the Table B.4 hereabove:

$$TotalBCC = 2.85 * 0.336 * 0.4101 + 10.365 * 0.313 * 0.6378$$

TotalBCC equals to 2.46 t biochar C ha⁻¹ yr⁻¹.

B.3 Changes to RothC model from package ‘SoilR’ coded in R

RothC soil carbon model from the package ‘SoilR’ (Sierra and Mueller, 2015) in R (Core Team, 2018) (hereafter referred as ‘RothC_R’) was amended to correspond better with the output values obtained through the RothC original Windows version. From the original R script, we:

- (i) We added a ‘Soil Cover Factor’, present in the original RothC, which modifies the rate constants and takes the monthly value of 0.6 if the soil is vegetated or 1 if it is bare, with an averaged value considering 9 months covered and 3 months bare for sugarcane (Scarpore *et al.*, 2016).
- (ii) We modified the Top Soil Moisture Deficit (‘fW rate factor modifier’) which, originally, takes the monthly value of 1 if the soil is vegetated or 1.8 if it is bare (Coleman and Jenkinson, 2014) with an averaged value based on the same covered and bare ratio than for the ‘Soil Cover Factor’.
- (iii) We multiplied the Soil Cover Factor (SCF) by the rate modifier for temperature (fT) and the updated rate modifier for soil moisture (fW), so the final modifier (Xi) takes into account the temperature, moisture and soil cover.

B.4 Model value outputs and literature data comparison

The steady state carbon stock values produced by the Windows and R versions of RothC were highly similar (Figure B.2). The slight observed differences may be due to the use of yearly averaged values for the ‘Topsoil Moisture Deficit’ coverage modifying factor and the ‘Soil Cover Factor’ (discussed here above) in the RothC_R whereas in Windows version monthly values were used. Our overall mean value of soil carbon stock at steady state (represented by the purple line in Figure B.2) is consistent with the mean top 30 cm SOC stock values for São Paulo State as reported in the literature (De Oliveira Bordonal *et al.*, 2013; Maia *et al.*, 2014; Carvalho *et al.*, 2017) (see black boxplot in Figure B.2).

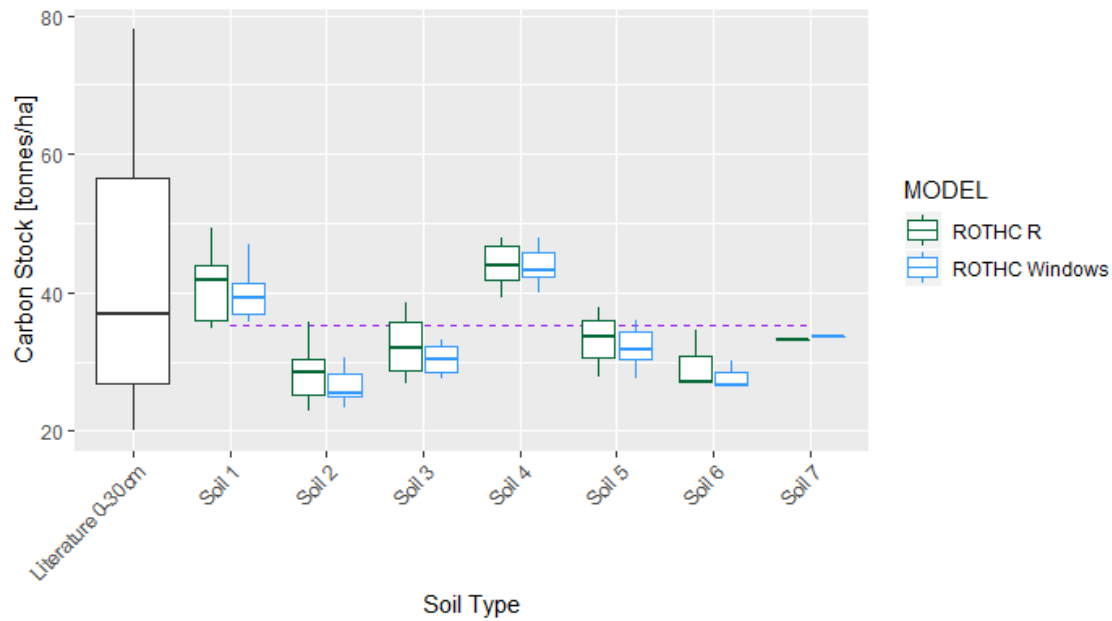


Figure B.2: Soil carbon stock values at steady state obtained through the R (green) and the Windows (blue) version of RothC. The boxplot combines the values obtained for the different meteorological stations relevant to each soil type. The black boxplot represents the spread of the soil carbon stock under sugarcane cultivation in São Paulo found in the literature (De Oliveira Bordonal et al., 2013; Maia et al., 2014; Carvalho et al., 2017). The single line for Soil 7 is due to the only meteorological station attributed to this soil type (see Table B.3). The purple dotted line represents the averaged value when all relevant soils and climate station combinations are considered. The end of the upper (lower) whiskers represent the largest (smallest) value within 1.5 times interquartile range above (below) 75th (25th) percentile, as defined by the ggplot2 package of R software version (3.5.1) (Core Team, 2018).

B.5 Evaluation of the model

B.5.1 Evaluation dataset

Evaluation of our model was made using the data from Liu *et al.* (2020) who measured soil carbon stock increase of a sandy loam soil in China under yearly rotations of wheat-maize, in which both were grown to harvest. The measurement was made after 5 years (10 crop seasons) while biochar application was made 10 times in total (once after each harvest). Data needed for the evaluation of our model are reported in Table B.5.

Table B.5: Evaluation dataset

Parameter	Value	Unit	Reference
Cultivation Period	October to June (wheat) and June to September for maize	-	(Liu <i>et al.</i> , 2020)
Biochar application rate	0; 2.25; 6.75; 11.35	tonnes ha ⁻¹ twice a year	(Liu <i>et al.</i> , 2020)
Biochar feedstock	Rice straw (500°C)	-	(Liu <i>et al.</i> , 2020)
Biochar application depth	15	cm	(Liu <i>et al.</i> , 2020)
Biochar carbon content	51.5	%	(Liu <i>et al.</i> , 2020)
Biochar recalcitrant pool mean residence time	840 [‡]	Years	(Zimmerman, 2010)
Biochar labile content	3 [‡]	%	(Foereid, Lehmann and Major, 2011; Woolf and Lehmann, 2012)
Soil clay content	22.6	%	(Liu <i>et al.</i> , 2020)
Soil sand content	46.3	%	(Liu <i>et al.</i> , 2020)
Site latitude	35.0 N	Degree	(Liu <i>et al.</i> , 2020)
Mean annual temperature	13.9	°C	(Liu <i>et al.</i> , 2020)
Mean annual rainfall	605	mm	(Liu <i>et al.</i> , 2020)
Average soil carbon stock at steady state	16.65	tonnes C ha ⁻¹	(Liu <i>et al.</i> , 2020)
Carbon content measurement depth	15	cm	(Liu <i>et al.</i> , 2020)
Fresh yearly carbon input*	0.99	tonnes C ha ⁻¹	(Coleman and Jenkinson, 2014)

Parameter	Value	Unit	Reference
DPM:RPM ratio	1.44	-	(Coleman and Jenkinson, 2014)

* Calculated using climate, soil, and management data in RothC.

‡ Considered as identical to our assessment using sugarcane biochar.

B.5.2 Additional evaluation work

Long-term experiments reporting soil carbon stock changes after biochar addition are scarce. In addition to the first evaluation dataset from Liu *et al.* (Liu *et al.*, 2020) (shown in the manuscript), a second attempt to evaluate our model was made using the data from Rogovska *et al.* (2014) who measured soil carbon content and bulk density of a Mollisol in Iowa (US) under maize cultivation two years after biochar application. The same data was used by Archontoulis *et al.* (2016) to validate their biochar sub-model for the Agricultural Production System Simulator (APSIM).

Parameter settings for the additional evaluation of our model are reported in Table B.6.

Table B.6: Additional evaluation dataset

Parameter	Value	Unit	Reference
Maize cultivation period	May to October	-	(Rogovska <i>et al.</i> , 2014)
Biochar application rate	0; 19; 38; 58; 77; 96	tonnes ha ⁻¹	(Rogovska <i>et al.</i> , 2014)
Biochar feedstock	Mixed hardwood	-	(Rogovska <i>et al.</i> , 2014)
Majority of biochar particle size	< 1	mm	(Rogovska <i>et al.</i> , 2014)
Biochar application depth	30	cm	(Rogovska <i>et al.</i> , 2014)
Biochar carbon content	78	%	(Rogovska <i>et al.</i> , 2014)
Biochar lost during application	2	%	(Archontoulis <i>et al.</i> , 2016)
Biochar recalcitrant pool mean residence time	500	Years	(Archontoulis <i>et al.</i> , 2016)

Parameter	Value	Unit	Reference
Biochar labile content	13	%	(Archontoulis <i>et al.</i> , 2016)
Soil clay content	21	%	(Archontoulis <i>et al.</i> , 2016)
Soil sand content	41.6	%	(Archontoulis <i>et al.</i> , 2016)
Soil organic carbon content	2.03	%	(Archontoulis <i>et al.</i> , 2016)
Site latitude	42.0	Degree	(Archontoulis <i>et al.</i> , 2016)
Mean annual temperature	9.3	°C	(Archontoulis <i>et al.</i> , 2016)
Mean annual rainfall	900	mm	(Archontoulis <i>et al.</i> , 2016)
Average soil carbon stock at steady state	33.6	tonnes C ha ⁻¹	(Rogovska <i>et al.</i> , 2014)
Carbon content measurement depth	15	cm	(Rogovska <i>et al.</i> , 2014)
Fresh yearly carbon input*	2.554	tonnes C ha ⁻¹	(Coleman and Jenkinson, 2014)
DPM:RPM ratio	1.44	-	(Coleman and Jenkinson, 2014)
Additional fresh C input at the beginning of the experiment‡	9.1	tonnes C ha ⁻¹	(Rogovska <i>et al.</i> , 2014)

* Calculated using climate, soil, and management data in RothC.

‡ The experiment includes additional maize straw input on the field after biochar application to avoid loss by erosion.

Comparisons between simulated and measured values (Figure B.3) shows good agreement up to the application rate of 38 t of biochar ha⁻¹. However, the difference between simulated and measured soil carbon stock is large for the application rates of 58, 77, and 96 t of biochar ha⁻¹. Similar results were obtained by Archontoulis *et al.* (Archontoulis *et al.*, 2016) during their model validation attempt of their APSIM biochar sub-model.

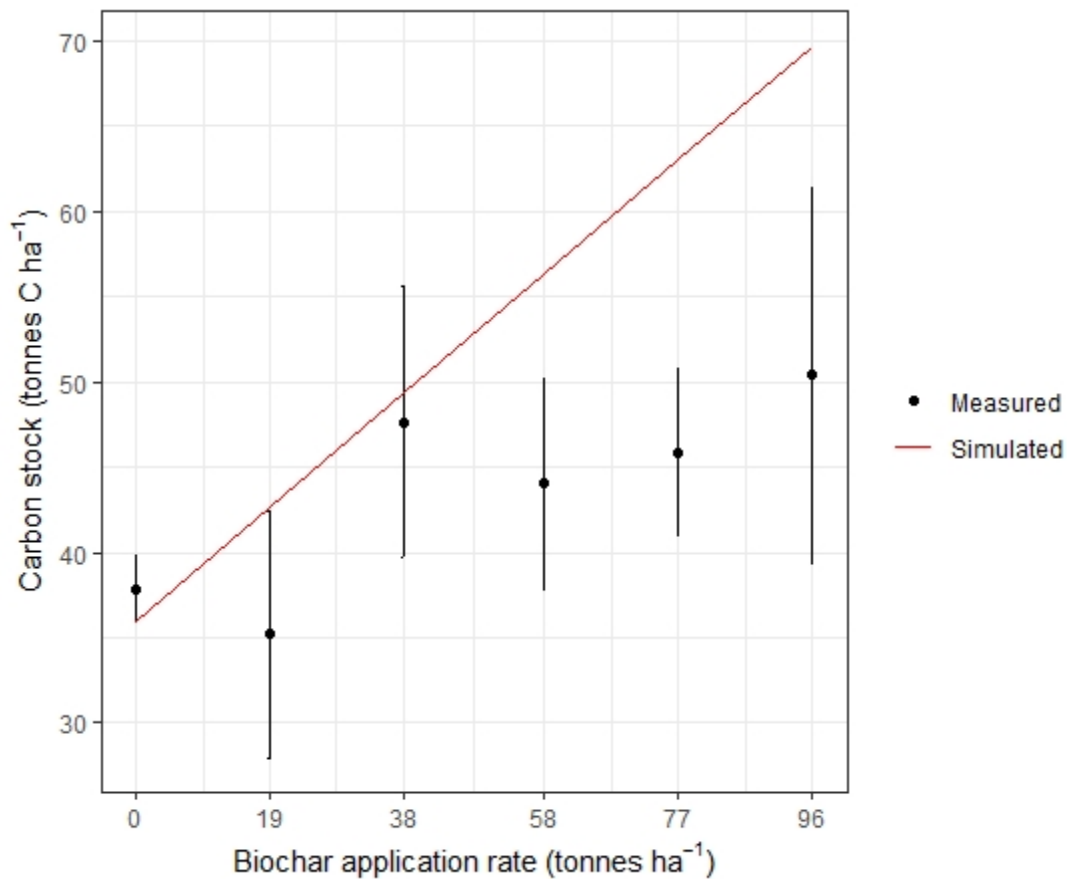


Figure B.3: Comparison between measured (Rogovska et al., 2014) (black dots $\pm$ SD) and simulated (red line) values using our modified RothC model. The soil carbon measured values originates from Rogovska et al. (2014) two years after biochar application.

The sudden decrease in measured soil carbon stock from 58 t of biochar ha⁻¹ onward translates a substantial loss of carbon. This loss is not expected to be due to an increase in mineralization rate of the biochar with increasing application rate (the opposite should occur, according to a recent review (Chao, Zhang and Wang, 2018)). Hence, this particular loss of biochar must be the consequence of factors, which are not taken into consideration by the RothC carbon model.

The literature suggests that biochar could be transported vertically through the soil profile towards deeper soil layers due to rainfall and/or bioturbation (Haefele *et al.*, 2011; Obia *et al.*, 2016), effectively making it absent from the topsoil carbon measurements. The share of biochar found under the application depth after one year ranges from 0.5% to 14.75% (Major *et al.*, 2010; Haefele *et al.*, 2011; Singh *et al.*, 2015; Obia *et al.*, 2016; Ventura *et al.*, 2019). The main factors influencing the vertical migration of biochar

particle are thought to be the biochar particle size (Obia *et al.*, 2016), the soil texture and porosity (Haefele *et al.*, 2011; Obia *et al.*, 2016), and the rainfall amount (Obia *et al.*, 2016). We interpolated the potential amount of biochar that could have been lost to deeper soil layers during the experiment of Rogovska *et al.* (2014) from the percent loss reported in the literature based on the sand content of the soils studied. The loss of biochar from leaching in Rogovska *et al.* (2014) was estimated at 5.9% of the applied biochar per year. This value is around half the values reported by Obia *et al.* (2016) for similar rainfall amounts ($\approx 1000 \text{ mm year}^{-1}$) and biochar particle size ($\leq 1 \text{ mm}$) but higher sand content. Accounting for this loss brings our simulated values closer to the measurements (Figure B.4).

Lateral transportation (erosion) of biochar is also expected to be an important pathway of biochar in soils (Foereid, Lehmann and Major, 2011). Recent studies suggest that 20% to 40% of the biochar could be lost by erosion a year or two after its application, even when the biochar is mixed in the first 10 cm of soil (Major *et al.*, 2010; Obia *et al.*, 2016). In Rogovska *et al.* (2014), the additional fresh organic matter input after biochar application aimed at reducing this potential erosion. No additional organic matter was reported for the second year of the experiment, which may have led to the loss of some biochar particles, however, the erosion of biochar is expected to decrease with time, as biochar particles are incorporated into aggregates (Foereid, Lehmann and Major, 2011). The lack of data prevents any precise assessment of biochar loss by erosion. Staying in the range of the number reported in the literature, we considered that an approximated 15% of the biochar applied was lost to erosion two years after its application (Figure B.4).

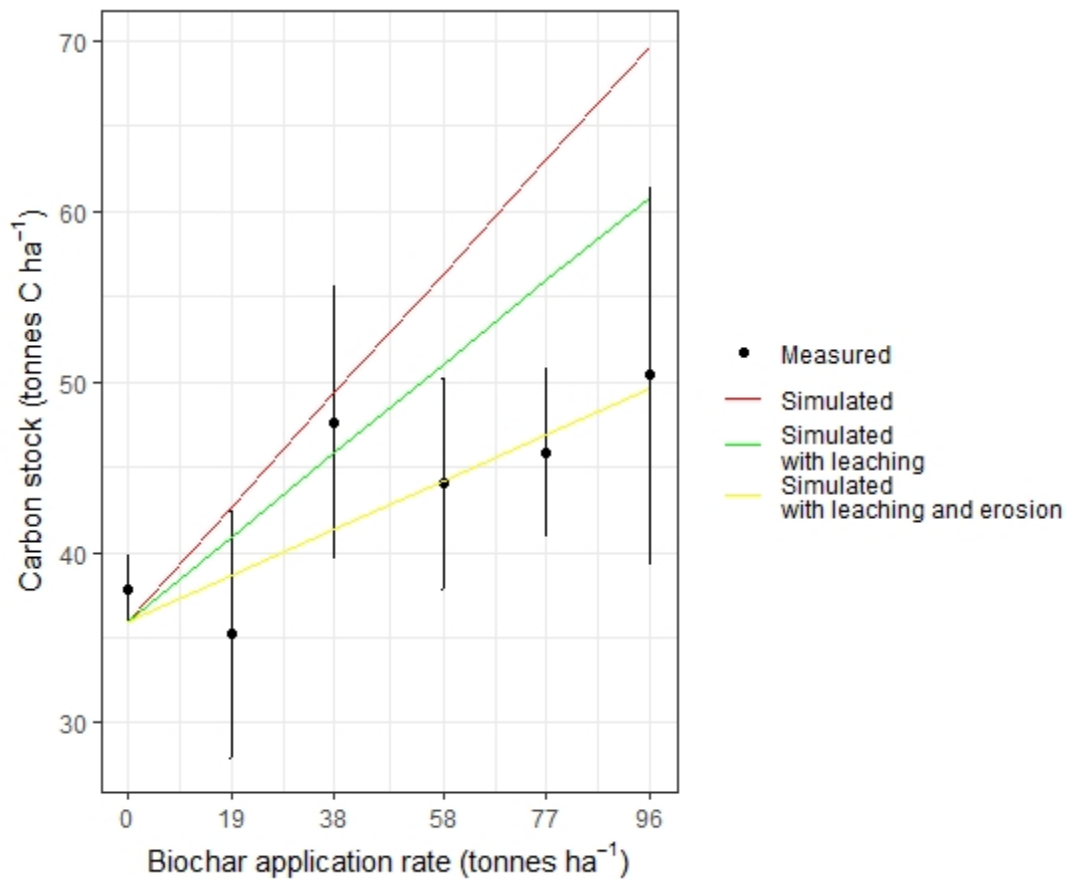


Figure B.4: Carbon stock (t ha^{-1}) 2 yr after biochar addition. Comparison between measured values (Rogovska *et al.*, 2014) (black dots $\pm$ SD), simulated (red line) values using our modified RothC model, simulated values accounting for potential leaching of biochar particles (green line), and simulated values accounting for potential downward (leaching) and lateral migration (erosion) of biochar particles (yellow line).

The loss of biochar through percolation and erosion, when reported, usually exceeds the loss through mineralization and is a known parameter causing underestimation of persistence of biochar in soils in field studies (Singh *et al.*, 2015; Leng *et al.*, 2019).

The reported, although scarcely documented, loss of biochar from its application layer either by leaching or erosion may render the evaluation of biochar models tedious. However, it does not impede its carbon sequestration potential as biochar in subsoil layers or deposited in aquatic environments may be more stable than when stored in the topsoil (Masiello and Druffel, 1998; Kögel-Knabner *et al.*, 2010).

B.5.3 Relevance of incubation studies

Incubation studies offer another type of measurement where biochar mineralization rate is precisely determined while avoiding other type of losses. However, they are often criticized for not representing the soil-plant system realistically enough (Wang *et al.*, 2016). Regarding biochar mineralization, critics argue that the optimal decomposition condition and usually short-term experimental duration leads to higher biochar decomposition rates than in field conditions (Kuzyakov, Bogomolova and Glaser, 2014; Wang *et al.*, 2016), while others expect an underestimation of the mineralization rate in incubation studies due to the impossibility of representing the complexity of real-world conditions at small scale (Leng *et al.*, 2019). Figure B.5 compares our modelled mineralization rate with values from the literature.

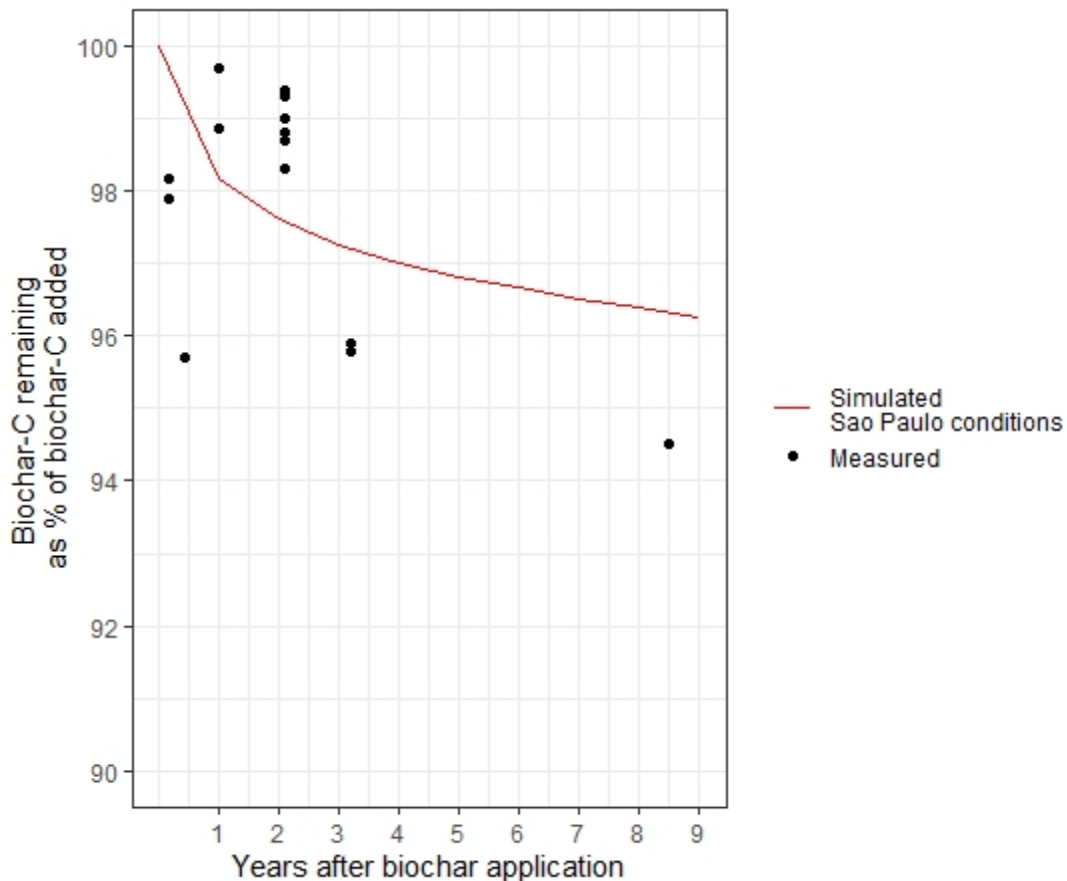


Figure B.5: Biochar-C remaining as percent of biochar-C added over time. Our simulated values (red line) are based on meta-analysis values (Wang *et al.*, 2016) applied on São Paulo conditions. The black dots are data points collected from literature (Kuzyakov *et al.*, 2009; Fang, Singh and Singh, 2014; Kuzyakov, Bogomolova and Glaser, 2014; Dharmakeerthi *et*

al., 2015; Fang et al., 2019) studying biochar mineralization in various condition through incubation experiments. The model reflects the general trend of these disparate experiments, which show, not-surprisingly, substantial scatter. Without longer term incubation experimental data, we cannot conclude if the model shows systematic bias nor do the experimental data contradict the model's predictions.

The main factors affecting biochar mineralization rate in incubation studies are the incubation time, the soil characteristics, and the biochar properties (Chao, Zhang and Wang, 2018). The results gathered (black dots - Figure B.5) originates from studies where biochar was made at a temperature between 400°C and 600°C (Kuzyakov *et al.*, 2009; Fang, Singh and Singh, 2014; Kuzyakov, Bogomolova and Glaser, 2014; Dharmakeerthi *et al.*, 2015; Fang *et al.*, 2019), which is representative of our simulated 550°C biochar.

B.6 References

Archontoulis, S. V. *et al.* (2016) 'A model for mechanistic and system assessments of biochar effects on soils and crops and trade-offs', *GCB Bioenergy*, 8(6), pp. 1028–1045. doi: 10.1111/gcbb.12314.

Beguéría, S. and Vicente-Serrano, S. M. (2017) 'SPEI: Calculation of the Standardised Precipitation-Evapotranspiration Index.' Available at: <https://cran.r-project.org/package=SPEI>.

Bolinder, M. A. *et al.* (2007) 'An approach for estimating net primary productivity and annual carbon inputs to soil for common agricultural crops in Canada', *Agriculture, Ecosystems and Environment*, 118(1–4), pp. 29–42. doi: 10.1016/j.agee.2006.05.013.

Cardoso, T. F. *et al.* (2019) 'A regional approach to determine economic, environmental and social impacts of different sugarcane production systems in Brazil', *Biomass and Bioenergy*. Elsevier Ltd, 120(April 2018), pp. 9–20. doi: 10.1016/j.biombioe.2018.10.018.

Carvalho, J. L. N. *et al.* (2017) 'Contribution of above- and belowground bioenergy crop residues to soil carbon', *GCB Bioenergy*, 9(8), pp. 1333–1343. doi: 10.1111/gcbb.12411.

Chao, L., Zhang, W. D. and Wang, S. L. (2018) 'Understanding the dominant controls on biochar decomposition using boosted regression trees', *European Journal of Soil Science*,

69(3), pp. 512–520. doi: 10.1111/ejss.12534.

Coleman, K. and Jenkinson, D. S. (2014) *RothC - A model for the turnover of carbon in soil Model - Model description and users guide*. Harpenden: Routledge. Available at: https://www.rothamsted.ac.uk/sites/default/files/RothC_guide_WIN.pdf.

CONAB (2018) *Acompanhamento da Safra Brasileira - Cana-de-açúcar - Terceiro levantamento | Dezembro 2018*. Available at: <https://www.conab.gov.br/info-agro/safras/cana>.

Core Team, R. (2018) *R: A language and environment for statistical computing. R Foundation for Statistical Computing*. Vienna. Available at: <https://www.r-project.org/>.

Cross, A. and Sohi, S. P. (2013) ‘A method for screening the relative long-term stability of biochar’, *GCB Bioenergy*, 5(2), pp. 215–220. doi: 10.1111/gcbb.12035.

Dharmakeerthi, R. S. *et al.* (2015) ‘Organic carbon dynamics in soils with pyrogenic organic matter that received plant residue additions over seven years’, *Soil Biology and Biochemistry*. Elsevier Ltd, 88, pp. 268–274. doi: 10.1016/j.soilbio.2015.06.003.

ESRI (2011) *ArcGIS Desktop*. Redlands, CA: Environmental System Research Institute. Available at: <http://desktop.arcgis.com>.

Fang, Y. *et al.* (2019) ‘Interactive carbon priming, microbial response and biochar persistence in a Vertisol with varied inputs of biochar and labile organic matter’, *European Journal of Soil Science*, 70(5), pp. 960–974. doi: 10.1111/ejss.12808.

Fang, Y., Singh, B. P. and Singh, B. (2014) ‘Temperature sensitivity of biochar and native carbon mineralisation in biochar-amended soils’, *Agriculture, Ecosystems and Environment*. Elsevier B.V., 191, pp. 158–167. doi: 10.1016/j.agee.2014.02.018.

FAO (2006) ‘CLIMWAT 2.0 for CROPWAT’. Rome: Water Development and Management Unit and the Climate Change and Bioenergy Unit of FAO. Available at: <http://www.fao.org/land-water/databases-and-software/climwat-for-cropwat/en/>.

FAO (2012) *Harmonized world soil database v1.2 | FAO SOILS PORTAL, Food and Agriculture Organization of the United Nations*. Available at: www.fao.org/soils-portal/soil-survey/soil-maps-and-databases/harmonized-world-soil-database-v12/en/

(Accessed: 15 March 2019).

Foereid, B., Lehmann, J. and Major, J. (2011) 'Modeling black carbon degradation and movement in soil', *Plant and Soil*, 345(1), pp. 223–236. doi: 10.1007/s11104-011-0773-3.

Galdos, M. V. *et al.* (2009) 'Simulation of Soil Carbon Dynamics under Sugarcane with the CENTURY Model', *Soil Science Society of America Journal*, 73(3), p. 802. doi: 10.2136/sssaj2007.0285.

George, P. A. O. *et al.* (2010) 'Residue from sugarcane juice filtration (filter cake): Energy use at the sugar factory', *Waste and Biomass Valorization*, 1(4), pp. 407–413. doi: 10.1007/s12649-010-9046-2.

Haefele, S. M. *et al.* (2011) 'Effects and fate of biochar from rice residues in rice-based systems', *Field Crops Research*. Elsevier B.V., 121(3), pp. 430–440. doi: 10.1016/j.fcr.2011.01.014.

Hassuani, S. J., Leal, M. R. L. V. and Macedo, I. de C. (2005) *Biomass Power Generation - Sugar cane bagasse and trash*. First. Edited by S. J. Hassuani, M. R. L. V. Leal, and I. de C. Macedo. Piracicaba Brasil: PNUD - Programa das Nações Unidas para o Desenvolvimento And CTC - Centro de Tecnologia Canavieira.

Khatiwada, D. *et al.* (2016) 'Optimizing ethanol and bioelectricity production in sugarcane biorefineries in Brazil', *Renewable Energy*. Elsevier Ltd, 85, pp. 371–386. doi: 10.1016/j.renene.2015.06.009.

Kögel-Knabner, I. *et al.* (2010) 'Biogeochemistry of paddy soils', *Geoderma*. Elsevier B.V., 157(1–2), pp. 1–14. doi: 10.1016/j.geoderma.2010.03.009.

Kuzyakov, Y. *et al.* (2009) 'Black carbon decomposition and incorporation into soil microbial biomass estimated by ^{14}C labeling', *Soil Biology and Biochemistry*. Elsevier Ltd, 41(2), pp. 210–219. doi: 10.1016/j.soilbio.2008.10.016.

Kuzyakov, Y., Bogomolova, I. and Glaser, B. (2014) 'Biochar stability in soil: Decomposition during eight years and transformation as assessed by compound-specific ^{14}C analysis', *Soil Biology and Biochemistry*. Elsevier Ltd, 70, pp. 229–236. doi:

10.1016/j.soilbio.2013.12.021.

Leng, L. *et al.* (2019) 'Biochar stability assessment by incubation and modelling: Methods, drawbacks and recommendations', *Science of the Total Environment*. Elsevier B.V., 664, pp. 11–23. doi: 10.1016/j.scitotenv.2019.01.298.

Liu, Y. *et al.* (2020) 'Successive straw biochar amendments reduce nitrous oxide emissions but do not improve the net ecosystem economic benefit in an alkaline sandy loam under a wheat–maize cropping system', *Land Degradation & Development*, 31(7), pp. 868–883. doi: 10.1002/ldr.3495.

Maia, S. M. F. *et al.* (2014) 'Payback time for soil carbon and sugar-cane ethanol', *Nature Climate Change*, 4(7), pp. 605–609. doi: 10.1038/nclimate2239.

Major, J. *et al.* (2010) 'Fate of soil-applied black carbon: Downward migration, leaching and soil respiration', *Global Change Biology*, 16(4), pp. 1366–1379. doi: 10.1111/j.1365-2486.2009.02044.x.

Masiello, C. A. and Druffel, E. R. M. (1998) 'Black Carbon in Deep-Sea Sediments', *Science*, 280(5371), pp. 1911–1913. doi: 10.1126/science.280.5371.1911.

Obia, A. *et al.* (2016) 'Vertical and lateral transport of biochar in light-textured tropical soils', *Soil and Tillage Research*. Elsevier B.V., 165, pp. 34–40. doi: 10.1016/j.still.2016.07.016.

de Oliveira, B. G. *et al.* (2013) 'Soil greenhouse gas fluxes from vinasse application in Brazilian sugarcane areas', *Geoderma*, 200–201, pp. 77–84. doi: 10.1016/j.geoderma.2013.02.005.

De Oliveira Bordonal, R. *et al.* (2013) 'Greenhouse gas mitigation potential from green harvested sugarcane scenarios in São Paulo State, Brazil', *Biomass and Bioenergy*. Elsevier Ltd, 59, pp. 195–207. doi: 10.1016/j.biombioe.2013.08.040.

Popin, G. V. *et al.* (2020) 'Sugarcane straw management for bioenergy: effects of global warming on greenhouse gas emissions and soil carbon storage', *Mitigation and Adaptation Strategies for Global Change*. Mitigation and Adaptation Strategies for Global Change, 25(4), pp. 559–577. doi: 10.1007/s11027-019-09880-7.

- Quirk, R. G. *et al.* (2012) 'Utilization of Biochar in Sugarcane and Sugar-Industry Management', *Sugar Tech*, 14(4), pp. 321–326. doi: 10.1007/s12355-012-0158-9.
- Rogovska, N. *et al.* (2014) 'Biochar impact on Midwestern Mollisols and maize nutrient availability', *Geoderma*. Elsevier B.V., 230–231, pp. 340–347. doi: 10.1016/j.geoderma.2014.04.009.
- Sampaio, I. L. M. *et al.* (2019) 'Electricity Production from Sugarcane Straw Recovered Through Bale System: Assessment of Retrofit Projects', *BioEnergy Research*. BioEnergy Research, 12(4), pp. 865–877. doi: 10.1007/s12155-019-10014-9.
- Scarpore, F. V. *et al.* (2016) 'Sugarcane land use and water resources assessment in the expansion area in Brazil', *Journal of Cleaner Production*, 133(June), pp. 1318–1327. doi: 10.1016/j.jclepro.2016.06.074.
- Sierra, C. A. and Mueller, M. (2015) 'Package " SoilR " - Models of Soil Organic Matter Decomposition', p. 177. Available at: <https://cran.r-project.org/web/packages/SoilR/index.html>.
- Silva, A. G. B. *et al.* (2019) 'How Much Sugarcane Straw is Needed for Covering the Soil?', *BioEnergy Research*. BioEnergy Research, 12(4), pp. 858–864. doi: 10.1007/s12155-019-10008-7.
- Singh, B. P. *et al.* (2015) 'In situ persistence and migration of biochar carbon and its impact on native carbon emission in contrasting soils under managed temperate pastures', *PLoS ONE*, 10(10), pp. 1–20. doi: 10.1371/journal.pone.0141560.
- Theodor Rudorff, B. F. (2014) *Canasat - INPE*. Available at: <http://www.dsr.inpe.br/laf/canasat/tabelas.html> (Accessed: 27 August 2019).
- Vasconcelos, A. L. S. *et al.* (2018) 'Greenhouse gas emission responses to sugarcane straw removal', *Biomass and Bioenergy*. Elsevier Ltd, 113(February), pp. 15–21. doi: 10.1016/j.biombioe.2018.03.002.
- Ventura, M. *et al.* (2019) 'Biochar mineralization and priming effect in a poplar short rotation coppice from a 3-year field experiment', *Biology and Fertility of Soils*. Biology and Fertility of Soils, 55(1), pp. 67–78. doi: 10.1007/s00374-018-1329-y.

Waldheim, L., Monis, M. and Verde Leal, M. R. (2001) 'Biomass Power Generation: Sugar Cane Bagasse and Trash', in Bridgwater, A. . (ed.) *Progress in Thermochemical Biomass Conversion*. Oxford, UK: Blackwell Science Ltd, pp. 509–523. Available at: http://www.sucre-ethique.org/IMG/pdf/CTC_energy_-_biomass_1_.pdf.

Wang, J. *et al.* (2016) 'Biochar stability in soil: Meta-analysis of decomposition and priming effects', *GCB Bioenergy*, 8(3), pp. 512–523. doi: 10.1111/gcbb.12266.

Woolf, D. and Lehmann, J. (2012) 'Modelling the long-term response to positive and negative priming of soil organic carbon by black carbon', *Biogeochemistry*, 111(1–3), pp. 83–95. doi: 10.1007/s10533-012-9764-6.

Zimmerman, A. R. (2010) 'Abiotic and Microbial Oxidation of Laboratory-Produced Black Carbon (Biochar)', *Environmental science & technology*, 44(4), pp. 1295–1301. doi: 10.1021/es903140c.

Appendix C Supplementary Information – Chapter 4: An anticipatory life cycle assessment of the use of biochar from sugarcane residues as a greenhouse gas removal technology

C.1 Scenario Description

C.1.1 Gas as the marginal electricity supply

Scenario 1 accounts for the carbon footprint of the necessary NG infrastructure increment to cover the electricity deficit and the updated emission factor of the country energy mix after accounting for the increased in NG use. In 2017, Brazil produced 54,000 GWh from 12,510 MW installed NG capacity (EPE, 2017). According to Dale *et al.* (2013), the infrastructure for a Brazilian combined natural gas power plant accounts for 49 t CO₂e MW⁻¹ installed capacity. Applied to the 2017 Brazilian installed power and electricity production, and considering 20 yr lifetime, the climate change impact of the infrastructure reaches 0.639 g CO₂e kWh⁻¹ (Dale *et al.*, 2013). This value is in agreement with our assessment using the Ecoinvent3 database applied to Brazilian conditions, i.e. 0.735 g CO₂e kWh⁻¹ for the infrastructure of a 100 MW natural gas power plant (Wernet *et al.*, 2016). Most of the GHG emissions from a natural gas plant are emitted during operation, and range between 360 and 575 g CO₂e kWh⁻¹ while the emission during the construction and decommissioning of the power plant are usually considered insignificant (Weisser, 2007).

C.1.2 Wind as the marginal electricity production supply

Scenario 2 accounts for the carbon footprint of the needed wind turbine infrastructure increment to cover the electricity deficit and the updated emission factor of the country energy mix after accounting for increased wind electricity production. In 2017 the total wind turbine capacity across Brazil was 12,325 MW and produced 42,000 GWh (EPE, 2017). Following the same principle as for NG, we determined the number of wind turbines necessary to cover the electricity deficit. The climate change impact of the infrastructure needed for wind energy in Brazil is 620 t CO₂e MW⁻¹ (Dale *et al.*, 2013). Applied to the production values given by the EPE (2017), and considering a lifetime of 20 years (Wernet *et al.*, 2016; Lucena and Lucena, 2019), the carbon footprint is 9.8 g

CO₂e kWh⁻¹ of electricity to the grid. This number is similar to the values using Ecoinvent3 considering a 2 MW Brazilian wind turbine (i.e. 8.3 g CO₂e kWh⁻¹). Additional estimates of GHG emissions of wind turbine infrastructure include 9.73 g CO₂e kWh⁻¹ (Guezuraga, Zauner and Pölz, 2012), 11.44 g CO₂e kWh⁻¹ for an onshore European 2 MW wind turbine (Sacchi *et al.*, 2019) and 7.1 g CO₂e kWh⁻¹ for a Brazilian 1.5 MW wind turbine (Oebels and Pacca, 2013). The lower climate change impact of the Brazilian wind turbine is due to the decarbonised energy mix of Brazil leading to lower GHG emissions for the local manufacturing of wind turbine parts (Oebels and Pacca, 2013). When considering wind energy, unlike conventional fuel-based technologies, most of the emissions arise from the manufacture and installation phases (Weisser, 2007; Oebels and Pacca, 2013; Sacchi *et al.*, 2019). A recent LCA shows that the raw material and installation phase of a wind turbine in Jordan accounted for 8.97 gCO₂e kWh⁻¹ while the operation phase emits 0.0278 g CO₂e kWh⁻¹ (Gomaa *et al.*, 2019). Overall, between 72–90% of wind turbine climate change impact comes from the installation and material, with the total impact between 8–30 g CO₂e kWh⁻¹ depending on geography and wind regimes (Weisser, 2007).

C.1.3 25% Wind and 75% gas as the marginal mix electricity production mean

Scenario 3 follows the same principle as the first two but considers that 75% of the electricity deficit is covered by NG and 25% by wind.

C.1.4 Brazil marginal electricity mix for 2040

Recently, Vandepaer *et al.* (2019), compared different marginal electricity mixes based on the consequential life cycle inventory of Ecoinvent V3.4. This consequential life cycle inventory prioritizes the reaction to a change of electricity demand through the installation of new production capacity by the predicted long-term marginal supplier. Considering the year 2030 as the year at which an increase in demand is registered and the year 2040 as the year at which all the installations are operational, the marginal electricity mix of Brazil will have a global warming impact (IPCC 2013, GWP 100 yr impact category) of 0.136 kg CO₂e kWh⁻¹ net electricity supplied (Vandepaer *et al.*, 2019).

C.2 Biochar Producing System description

C.2.1 Biomass grinding

Before pyrolysis, sugarcane residues are ground to a smaller particle size. According to several sources, updated with our biochar production yield, the energy required for grinding averages 91 kWh tBC⁻¹ (Roberts *et al.*, 2010; Wernet *et al.*, 2016; Brassard *et al.*, 2018; AMISY, 2020b). This figure is likely to be overestimated, as the biomass does not need to be ground too finely to increase the non-condensable share of the by-products and the potential energy production from pyrolysis (Basu, 2010).

C.2.2 The Biochar plant

Our scenarios consider biochar to be produced by a conventional industrial slow pyrolyzer at around 550°C. The products of the pyrolysis are biochar, syngas and bio-oil in a ratio that reflects the biomass and pyrolysis plant used (Basu, 2010). Slow pyrolysis was selected because it favours biochar over other products when compared to other pyrolysis options (Basu, 2010). While biochar characteristic and properties vary according to the production temperature (Kambo and Dutta, 2015), biochar made at around 550 °C usually shows good characteristic mix for its use as a soil amendment (Lehmann, 2007). In our scenarios the bio-oil is not collected but rather combusted in the gas phase (Gabra *et al.*, 2001; Basu, 2010). This approach has been reported in previous studies (Park *et al.*, 2014; Wang *et al.*, 2014; Roy and Dias, 2017; Azzi, Karlton and Sundberg, 2019). The gas released during pyrolysis is used to generate electricity via boiler or gas turbine (Quirk *et al.*, 2012; de Oliveira Vilela *et al.*, 2014; You *et al.*, 2017; Oldfield *et al.*, 2018).

We based the energy production on available data and set the results using the sugarcane biochar yield (32.4%). The average surplus electricity produced by the production of biochar through slow pyrolysis is 355 kWh tBC⁻¹ (Roberts *et al.*, 2010; Hammond *et al.*, 2011; Quirk *et al.*, 2012; Muñoz *et al.*, 2017; Rajabi Hamedani *et al.*, 2019). This number includes the use of some heat from pyrolysis to dry the feedstock. According to Leme *et al.* (2018), an auto-thermal Brazilian batch slow pyrolysis plant, producing charcoal from eucalyptus wood for use in the steel industry produces enough syngas (used in a steam Rankine cycle) to generate 930 kWh of electricity per tonne of charcoal produced including heat loss for biomass drying. Similarly, Shen *et al.* (2020), compared the potential electricity production between biochar production and combustion of wood

waste in Singapore. They report that 0.69 and 2.06 kWh per kg of wood waste could be produced during pyrolysis and combustion, respectively (Shen *et al.*, 2020). Our results are 0.11 kWh and 0.96 kWh per kg of sugarcane residue through pyrolysis and CHP, respectively. Our estimate of electricity production through pyrolysis is therefore probably underestimated, although sugarcane residues have a lower heating value than wood proposed in the above-mentioned articles (Raveendran and Ganesh, 1996). In comparison, another study looking at pyrolysis of winter oilseed rape straw found that each kg of residue produced 0.44 kWh of electricity when pyrolyzed at 400°C (including drying and losses) (Thers *et al.*, 2019).

C.2.3 Biochar pellets

Following pyrolysis, the biochar powder is compressed into pellets to facilitate transport, spreading, and avoid losses. The average electricity necessary to pelletize biochar is 61 kWh tBC⁻¹ (Bartocci *et al.*, 2016; AMISY, 2020a).

C.2.4 Loading operation

Once in pellet form, the biochar is loaded in the truck. Loading operation diesel use and emission factors were collected from a previous study in São Paulo condition (Rosado *et al.*, 2017).

C.2.5 Transport of biochar to the field

The sugar content of the cane degrades rapidly after harvest, so sugarcane processing plants are situated close to sugarcane fields (Hassuani, Leal and Macedo, 2005). The current average distance from the field to the mill is 31 km (Waack and Neves, 1998; Hassuani, Leal and Macedo, 2005; Seabra *et al.*, 2011; Cardoso *et al.*, 2018, 2019; Sampaio *et al.*, 2019). In our scenarios trucks deliver biochar to the field margins and then return empty to the processing plant (Weidema *et al.*, 2013). Delivered biochar is loaded in the fertilizer spreading machine, requiring a second loading operation.

C.2.6 Biochar spreading

In our scenarios we considered biochar to be deployed as a separate operation but using spreaders designed for conventional lime or mineral fertilizer. Since tillage does not occur during four years of ratooning (Nihei, Hayashi and Shirota, 2015) the 4.2, 2.1 or 1.05 t

ha⁻¹ (depending on the sub-scenario) of biochar pellets are applied to the surface straw blanket, which has a biomass of 7 t dm ha⁻¹. Subsequent operations using heavy machinery include the application of herbicide, insecticide and/or fertilizer between rows during ratoon growth (Nihei, Hayashi and Shirota, 2015; Bordonal *et al.*, 2018). Such management on straw blanket is assumed to minimize lateral displacement of biochar, so we do not account for any losses of biochar arising from erosion. Strictly, movement of biochar to water bodies and eventually oceans should not increase its decomposition and reduce its carbon sequestration potential (Masiello and Druffel, 1998), but its effects on the soil-plant systems might be altered and protocols for carbon accounting. In each fifth year the accumulated biochar would be more thoroughly integrated in the topsoil during the next planting operation (Nihei, Hayashi and Shirota, 2015). A fertilizer spreader will use on average 2.1 kg of diesel t⁻¹ for lime or mineral fertilizer (Koga *et al.*, 2003; Grisso *et al.*, 2010; Handler and Nadlinger, 2012; Lutz Böhle, 2014), which agrees with the number proposed by Roberts *et al.* (2010) for biochar spreading.

C.3 Data

Table C.1: System information

Parameter	Data	Unit	Data Origin	Comment
Sugarcane field in SP	5,768,172	ha	(Theodor Rudorff, 2014)	
Sugarcane field in Brazil	10,189,208	ha	(FAO, 2019)	
Sugarcane Yield SP 2018-19	74.04	t ha ⁻¹ yr ⁻¹	(CONAB, 2018)	
Filter Cake addition	0.73	t ha ⁻¹ yr ⁻¹	(Cardoso <i>et al.</i> , 2019)	
Vinasse addition	31.00	m ³ yr ⁻¹	(Cardoso <i>et al.</i> , 2019)	
Filter Cake Carbon content	32.50	%	(George <i>et al.</i> , 2010)	
Vinasse carbon content	1.99	g litre ⁻¹	(de Oliveira <i>et al.</i> , 2013)	
Average Bagasse yield after processing (dry weight)	140	kg tC ⁻¹	(Waldheim, Monis and Verde Leal, 2001;	

Parameter	Data	Unit	Data Origin	Comment
Average straw per tonne of stalk (dry weight)	140	kg tC ⁻¹	Khatiwada <i>et al.</i> , 2016) (Waldheim, Monis and Verde Leal, 2001; Hassuani, Leal and Macedo, 2005)	
Straw loss during transport	5.00	% of tC yield	(Sampaio <i>et al.</i> , 2019) (Hassuani, Leal and Macedo, 2005;	
Amount of straw to be left on field	7.00	t ha ⁻¹ yr ⁻¹	Vasconcelos <i>et al.</i> , 2018; Gomes <i>et al.</i> , 2019; Silva <i>et al.</i> , 2019; Popin <i>et al.</i> , 2020)	
Sugarcane biochar (525°C) %C lost after 100 years	11.90	%	(Zimmerman, 2010)	
Sugarcane straw biochar (550°C) C content (% dry mass)	41.01	%	(Cross and Sohi, 2013)	
Sugarcane bagasse biochar (550°C) C content (% dry mass)	63.78	%	(Cross and Sohi, 2013)	
Bagasse Biochar production Yield (550°C)	31.30	%	(Quirk <i>et al.</i> , 2012)	
Straw Biochar production Yield (550°C)	33.60	%	(Quirk <i>et al.</i> , 2012)	
Amount of straw sent to CHP (baseline)	2.85	t ha ⁻¹ yr ⁻¹	Calculated	- Straw loss - Straw blanketing
Amount of bagasse sent to CHP (baseline)	10.37	t ha ⁻¹ yr ⁻¹	(Khatiwada <i>et al.</i> , 2016)	

Parameter	Data	Unit	Data Origin	Comment
tC ⁻¹ = ton of fresh cane harvested				

Table C.2: Electricity baseline mill

Parameter	Data	Unit	Data Origin
Electricity needed to run the mill	30	kWh tC ⁻¹	(Cardoso <i>et al.</i> , 2019)
Electricity needed to run the mill	32	kWh tC ⁻¹	(Klein <i>et al.</i> , 2018)
Electricity needed to run the mill	28	kWh tC ⁻¹	(Seabra and Macedo, 2011)
Electricity needed to run the mill	25	kWh tC ⁻¹	(Pippo and Luengo, 2013)
Electricity needed to run the mill	30	kWh tC ⁻¹	(Pippo and Luengo, 2013)
Surplus electricity to the grid (bagasse)	34	kWh tC ⁻¹	(Cardoso <i>et al.</i> , 2019)
Surplus electricity to the grid (bagasse)	104.60	kWh tC ⁻¹	(Dias <i>et al.</i> , 2011)
Surplus electricity to the grid (bagasse)	70.80	kWh tC ⁻¹	(Sampaio <i>et al.</i> , 2019)
Surplus electricity to the grid (bagasse)	77.50	kWh tC ⁻¹	(Sampaio <i>et al.</i> , 2019)
Surplus electricity to the grid (bagasse)	69.60	kWh tC ⁻¹	(Olivério and Ferreira, 2010)
Surplus electricity to the grid (bagasse)	77.34	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	83.10	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	85.58	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	91.09	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	77.48	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	83.02	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	87.61	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (bagasse)	93.22	kWh tC ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.16	kWh kg of straw ⁻¹	(Dias <i>et al.</i> , 2011)
Surplus electricity to the grid (straw)	3.39	kWh kg of straw ⁻¹	(Cardoso <i>et al.</i> , 2019)

Parameter	Data	Unit	Data Origin
Surplus electricity to the grid (straw)	1.45	kWh kg of straw ⁻¹	(Klein <i>et al.</i> , 2018)
Surplus electricity to the grid (straw)	1.23	kWh kg of straw ⁻¹	(Sampaio <i>et al.</i> , 2019)
Surplus electricity to the grid (straw)	1.06	kWh kg of straw ⁻¹	(Sampaio <i>et al.</i> , 2019)
Surplus electricity to the grid (straw)	1.07	kWh kg of straw ⁻¹	(Klein <i>et al.</i> , 2018)
Surplus electricity to the grid (straw)	1.25	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.24	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.31	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.67	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.27	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.27	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.37	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.38	kWh kg of straw ⁻¹	(Alves <i>et al.</i> , 2015)
Surplus electricity to the grid (straw)	1.75	kWh kg of straw ⁻¹	(Seabra and Macedo, 2011)

Table C.3: Electricity pyrolysis

Parameter	Data	Unit	Data Origin	Comment
Biochar average yield	32.40	%	(Quirk <i>et al.</i> , 2012)	
Gas engine conversion efficiency	37.00	%	(Quirk <i>et al.</i> , 2012)	In the range of Hammond <i>et al.</i> (2011) (Hammond <i>et al.</i> , 2011)
Biochar maximum application rate	4.20	tBC ha ⁻¹	Calculated	

Parameter	Data	Unit	Data Origin	Comment
Biochar amount per tonne of harvested cane	0.06	tBC tC ⁻¹	Calculated	
Pyrolysis electricity/Energy production	0.50	MWh tonne dry feedstock ⁻¹	(Quirk <i>et al.</i> , 2012)	Sugarcane straw BC (550°C) Electricity production
Pyrolysis electricity/Energy production	0.50	MWh tonne dry feedstock ⁻¹	(Quirk <i>et al.</i> , 2012)	Sugarcane bagasse BC (550°C) Electricity production
Pyrolysis electricity/Energy production	0.70	MWh tonne dry feedstock ⁻¹	(Hammond <i>et al.</i> , 2011)	Electricity production
Pyrolysis electricity/Energy production	4,899.00	MJ tonne dry feedstock ⁻¹	(Roberts <i>et al.</i> , 2010)	Positive net energy in the system
Pyrolysis electricity/Energy production	6,467	MJ tBC ⁻¹	(Muñoz <i>et al.</i> , 2017)	Excess energy produced through pyrolysis
Pyrolysis electricity/Energy production	5,787	MJ tonne of dry feedstock ⁻¹	(Roberts <i>et al.</i> , 2010)	Excess syngas heat energy
Pyrolysis electricity/Energy production	1.01	GJ tBC ⁻¹	(Rajabi Hamedani <i>et al.</i> , 2019)	Electricity production during pyrolysis
Pyrolysis electricity/Energy production	665.00	kWh tBC ⁻¹	(Yang <i>et al.</i> , 2017)	
Pyrolysis electricity/Energy production	406.62	kWh tBC ⁻¹	(Yang <i>et al.</i> , 2018)	

Table C.4: Electricity biochar processing

Parameter	Data	Unit	Data Origin	Comment
Biomass milling	16.60	kWh tBC ⁻¹	(AMISY, 2020b)	electrical wood hammer mill

Parameter	Data	Unit	Data Origin	Comment
Wood chipping, industrial residual wood, stationary electric chipper	197.50	kWh tBC ⁻¹	(Wernet <i>et al.</i> , 2016)	analysed through cumulative energy demand (CED)
Biomass grinding	123.12	kWh tBC ⁻¹	(Brassard <i>et al.</i> , 2018)	
Biomass chipping	27.63	kWh tBC ⁻¹	(Roberts <i>et al.</i> , 2010)	
Biochar pellet	72.00	kWh tBC ⁻¹	(AMISY, 2020a)	High Efficiency Vertical Ring Die Wood Pellet Machine
Biochar pellet	0.05	kWh kg of pellet ⁻¹	(Bartocci <i>et al.</i> , 2016)	

Table C.5: Additional processes

Parameter	Data	Unit	Data Origin
Distance field - mill	19	km	(Hassuani, Leal and Macedo, 2005)
Distance field - mill	25	km	(Cardoso <i>et al.</i> , 2018)
Distance field - mill	32	km	(Cardoso <i>et al.</i> , 2019)
Distance field - mill	50	km	(Waack and Neves, 1998)
Distance field - mill	37	km	(Sampaio <i>et al.</i> , 2019)
Distance field - mill	21	km	(Seabra <i>et al.</i> , 2011)
Loading operation	0.19	l of diesel tBC ⁻¹	(Rosado <i>et al.</i> , 2017)
Spreading operation	2.15	kg of diesel tBC ⁻¹	(Koga <i>et al.</i> , 2003; Grisso <i>et al.</i> , 2010; Handler and Nadlinger, 2012; Lutz Böhle, 2014)

Table C.6: Yearly addition of N containing amendment with their nitrogen content and the total N applied per hectare per year

Nitrogen Source	Application rate (kg ha ⁻¹ yr ⁻¹) (Cardoso <i>et al.</i> , 2019)	Nitrogen content	Data Origin	Applied N (kg ha ⁻¹ yr ⁻¹)
N as Urea (fertilizer)	86.80	/	(Cardoso <i>et al.</i> , 2019)	86.8
Vinasse	31 m ³	0.595 kg N m ⁻³	(Macedo, Seabra and Silva, 2007)	18.44

Filter Cake	733.00	12.5 kg N tonne ⁻¹	(Chagas <i>et al.</i> , 2016)	9.162
Straw	7000 dm (scenario)	6.09 g N kg ⁻¹	(Menandro <i>et al.</i> , 2017)	42.63

Table C.7: Emission factors

Parameter	Data	Unit	Data Origin	Comment
Transport	0.171	kg CO ₂ e tkm ⁻¹	(Wernet <i>et al.</i> , 2016)	Transport, freight, lorry 16-32 metric ton, EURO4 {RoW} transport, freight, lorry 16-32 metric ton, EURO4 Cut-off, U
Diesel for loader operation	3.2	kg CO ₂ e l of diesel ⁻¹	(Wernet <i>et al.</i> , 2016)	Diesel, combusted in industrial equipment/US
Production of diesel	0.507	kg CO ₂ e kg of diesel ⁻¹	(Wernet <i>et al.</i> , 2016)	Diesel, low-sulfur {BR} diesel production, low- sulphur, petroleum refinery operation Cut- off, U
Diesel market	0.548	kg CO ₂ e kg of diesel ⁻¹	(Wernet <i>et al.</i> , 2016)	Diesel, low-sulfur {BR} market for diesel, low- sulfur Cut-off, U
Diesel for spreading operation	5.557	kg CO ₂ e kg of diesel ⁻¹	(Wernet <i>et al.</i> , 2016)	Diesel, burned in agricultural machinery {GLO} diesel, burned in agricultural machinery Cut-off, U
Phosphate fertiliser, as P ₂ O ₅	1.691	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{RoW} triple superphosphate production Cut-off, U
Urea, as N	3.237	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{RoW} production Cut-off, U
Soil pH raising agent, as CaCO ₃	0.043	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{GLO} lime to generic market for soil pH raising agent Cut-off, U
Concrete block	0.074	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{BR} market for concrete block Cut-off, U
Steel, unalloyed	1.823	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{GLO} market for Cut- off, U

Parameter	Data	Unit	Data Origin	Comment
Aluminium, wrought alloy	12.81 1	kg CO ₂ e kg ⁻¹	(Wernet <i>et al.</i> , 2016)	{GLO} market for Cut-off, U
GWP - CH ₄	28	kg CO ₂ e kg ⁻¹	(Bréon <i>et al.</i> , 2013)	
GWP - NO _x	-11	kg CO ₂ e kg ⁻¹	(Bréon <i>et al.</i> , 2013)	
GWP - CO	2.65	kg CO ₂ e kg ⁻¹	(Bréon <i>et al.</i> , 2013)	
GWP - N ₂ O	265	kg CO ₂ e kg ⁻¹	(Bréon <i>et al.</i> , 2013)	

Table C.8: Average amendment application per hectare per year taking into account 4/5 years of ratooning (Cardoso *et al.*, 2019)

Nutrient	Application rate (kg ha ⁻¹ yr ⁻¹)	Application rate (kg yr ⁻¹ tC ⁻¹) *	Amendment reduction due biochar application (kg ha ⁻¹ yr ⁻¹)
N as urea	86.8	1.172	9.55
P ₂ O ₅	36	0.486	3.6
K ₂ O	120.8	1.631	/
limestone	400	5.403	57 (per tBC)
Ashes	457	6.173	/

*74.037 tC ha⁻¹

Table C.9: Pyrolysis plant construction material

Parameter	Data	Unit	Data Origin	Comment
Concrete	194	kg tBC ⁻¹	(Roberts <i>et al.</i> , 2010)	Considering 200 days yr ⁻¹ at 7 h day ⁻¹
Steel	62	kg tBC ⁻¹	(Roberts <i>et al.</i> , 2010)	Considering 200 days yr ⁻¹ at 7 h day ⁻¹
Aluminium	0.40	kg tBC ⁻¹	(Roberts <i>et al.</i> , 2010)	Considering 200 days yr ⁻¹ at 7 h day ⁻¹

Parameter	Data	Unit	Data Origin	Comment
Pyrolysis Plant workshop and office building	53	kg CO ₂ e tBC ⁻¹	(Yang <i>et al.</i> , 2016)	
Pyrolysis Plant equipment	25	kg CO ₂ e tBC ⁻¹	(Yang <i>et al.</i> , 2016)	
Separation and purification of the pyrolysis products	93	kWh tBC ⁻¹	(Yang <i>et al.</i> , 2016)	
Auxiliary systems and domestic electricity consumption	154	kWh tBC ⁻¹	(Yang <i>et al.</i> , 2016)	

C.4 Distribution – Monte Carlo

Natural Gas Emission Factor		Unit	Comments
Reported in the LCA	0.196	kg CO ₂ e kWh ⁻¹	
Range	0.1957 - 0.1958	kg CO ₂ e kWh ⁻¹	Based on infrastructure associated emission
Distribution	Pert		
N	2		

Wind Emission Factor		Unit	Comments
Reported in the LCA	0.125	kg CO ₂ e kWh ⁻¹	
Range	0.1246 - 0.1276	kg CO ₂ e kWh ⁻¹	Based on infrastructure associated emission
Distribution	Pert		
N	5		

Natural Gas (75%) Wind (25%) Emission Factor		Unit	Comments
Reported in the LCA	0.178	kg CO ₂ e kWh ⁻¹	
Range	0.1603 - 0.1960	kg CO ₂ e kWh ⁻¹	± 10 %
Distribution	Pert		
N	3		

Appendix C

Year 2040 Emission Factor		Unit	Comments
Reported in the LCA	0.1362	kg CO ₂ e kWh ⁻¹	
Range	0.12258 – 0.14982	kg CO ₂ e kWh ⁻¹	± 10 %
Distribution	Pert		
N	3		

Surplus electricity generated by Bagasse		Unit	Comments
Reported in the LCA	79.68	kWh tC ⁻¹	
Range	34.9 – 104.6	kWh tC ⁻¹	Literature
Distribution	Pert		
N	13		

Surplus electricity generated by Straw		Unit	Comments
Reported in the LCA	52.03	kWh tC ⁻¹	
Range	40.76 – 130.24	kWh tC ⁻¹	Literature
Distribution	Pert		
N	15		

Electric demand sugarcane mill		Unit	Comments
Reported in the LCA	29	kWh tC ⁻¹	
Range	25 – 32	kWh tC ⁻¹	Literature
Distribution	Pert		
N	5		

Surplus electricity generated during pyrolysis		Unit	Comments
Reported in the LCA	326.3	kWh tBC ⁻¹	
Range	162.3 – 665	kWh tBC ⁻¹	Literature
Distribution	Pert		
N	7		

Electric demand of grinding		Unit	Comments
Reported in the LCA	91.2	kWh tBC ⁻¹	
Range	16.6 – 197.5	kWh tBC ⁻¹	Literature
Distribution	Pert		
N	4		

Electric demand of pelletizing		Unit	Comments
Reported in the LCA	61	kWh tBC ⁻¹	
Range	50 – 72	kWh tBC ⁻¹	Literature
Distribution	Pert		
N	2		

Soil sequestration potential of BC		Unit	Comments
Reported in the LCA	116.5	kg CO ₂ t ⁻¹ harvested cane	
Range	96.5 – 136.5	kg CO ₂ t ⁻¹ harvested cane	Previous Study
Distribution	Pert		
N	3		

Distance between the pyrolysis plant and the field		Unit	Comments
Reported in the LCA	30.7	km	
Range	19 – 50	km	Literature
Distribution	Uniform		
N	6		

Loader Operation		Unit	Comments
Reported in the LCA	0.188	l diesel tBC ⁻¹	
Range	0.18 – 0.195	l diesel tBC ⁻¹	Literature
Distribution	Lognormal		

N	2		
---	---	--	--

Lime/Fertiliser spreader		Unit	Comments
Reported in the LCA	2.15	kg diesel tBC ⁻¹	
Range	0.49 – 4.94	kg diesel tBC ⁻¹	Literature
Distribution	Lognormal		
N	9		

Emission factor carbon monoxide		Unit	Comments
Reported in the LCA	2.65	kg CO ₂ kg ⁻¹	
Range	2 – 3.3	kg CO ₂ kg ⁻¹	Literature
Distribution	Uniform		
N	2		

C.5 Results

The following table shows the contribution analysis of each scenario (marginal energy)/sub-scenario (biochar production) combination.

The processes legend is as follow:

- BC_Soil: CO₂ captured based on the carbon content of biochar amended to soil
- Ferti_Red: Fertilizer reduction due to biochar application
- N2OEm_Red: Nitrous oxide emission reduction due to biochar application
- Elec_BC_Prod: Emission reduction due to the electricity production during pyrolysis
- Em_Grind: Emissions associated with the grinding process
- Em_Spread: Emissions associated with the spreading process
- Em_Transp: Emissions associated with the transport process
- Em_Pellet: Emissions associated with the pelletizing process
- Elec_Deficit: Emissions associated with covering the electricity deficit left by pyrolysis as compared with CHP
- Em_Loader: Emissions associated with the loading process

- Em_Building: Emissions associated with the construction of the pyrolysis plant.

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
NG	100%	BC_Soil	8.619	3.2E-01	Emission Reduction	95.70%
NG	100%	Ferti_Red	0.040	NA	Emission Reduction	0.40%
NG	100%	N2OEm_Red	0.055	NA	Emission Reduction	0.60%
NG	100%	Elec_BC_Prod	0.292	2.9E-02	Emission Reduction	3.20%
NG	100%	Em_Grind	-0.079	1.4E-02	Emission	2.50%
NG	100%	Em_Spread	-0.050	1.3E-02	Emission	1.60%
NG	100%	Em_Transp	-0.025	2.6E-03	Emission	0.80%
NG	100%	Em_Pellet	-0.050	2.4E-03	Emission	1.60%
NG	100%	Elec_Deficit	-2.478	7.7E-02	Emission	77.00%
NG	100%	Em Loader	-0.007	-5.4E-05	Emission	0.20%
NG	100%	Em_Building	-0.531	NA	Emission	16.50%
NG	50%	BC_Soil	4.315	1.6E-01	Emission Reduction	95.60%
NG	50%	Ferti_Red	0.039	NA	Emission Reduction	0.90%
NG	50%	N2OEm_Red	0.026	NA	Emission Reduction	0.60%
NG	50%	Elec_BC_Prod	0.134	1.3E-02	Emission Reduction	3.00%
NG	50%	Em_Grind	-0.036	6.5E-03	Emission	3.60%
NG	50%	Em_Spread	-0.023	5.8E-03	Emission	2.30%
NG	50%	Em_Transp	-0.011	1.2E-03	Emission	1.10%
NG	50%	Em_Pellet	-0.023	1.1E-03	Emission	2.30%
NG	50%	Elec_Deficit	-0.659	3.1E-02	Emission	65.90%
NG	50%	Em Loader	-0.003	-2.5E-05	Emission	0.30%
NG	50%	Em_Building	-0.244	NA	Emission	24.40%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
NG	25%	BC_Soil	2.149	8.2E-02	Emission Reduction	94.80%
NG	25%	Ferti_Red	0.038	NA	Emission Reduction	1.70%
NG	25%	N2OEm_Red	0.013	NA	Emission Reduction	0.60%
NG	25%	Elec_BC_Prod	0.067	6.6E-03	Emission Reduction	3.00%
NG	25%	Em_Grind	-0.018	3.2E-03	Emission	3.60%
NG	25%	Em_Spread	-0.012	2.9E-03	Emission	2.30%
NG	25%	Em_Transp	-0.006	6.0E-04	Emission	1.10%
NG	25%	Em_Pellet	-0.011	5.5E-04	Emission	2.30%
NG	25%	Elec_Deficit	-0.328	1.5E-02	Emission	65.90%
NG	25%	Em_Loader	-0.002	-1.2E-05	Emission	0.30%
NG	25%	Em_Building	-0.122	NA	Emission	24.40%
Yr-2040	100%	BC_Soil	8.624	3.2E-01	Emission Reduction	96.60%
Yr-2040	100%	Ferti_Red	0.040	NA	Emission Reduction	0.50%
Yr-2040	100%	N2OEm_Red	0.055	NA	Emission Reduction	0.60%
Yr-2040	100%	Elec_BC_Prod	0.203	2.0E-02	Emission Reduction	2.30%
Yr-2040	100%	Em_Grind	-0.055	9.9E-03	Emission	2.30%
Yr-2040	100%	Em_Spread	-0.050	1.3E-02	Emission	2.10%
Yr-2040	100%	Em_Transp	-0.025	2.6E-03	Emission	1.00%
Yr-2040	100%	Em_Pellet	-0.035	1.8E-03	Emission	1.50%
Yr-2040	100%	Elec_Deficit	-1.724	6.6E-02	Emission	72.90%
Yr-2040	100%	Em_Loader	-0.007	-5.5E-05	Emission	0.30%
Yr-2040	100%	Em_Building	-0.469	NA	Emission	19.80%
Yr-2040	50%	BC_Soil	4.316	1.6E-01	Emission Reduction	96.50%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
Yr-2040	50%	Ferti_Red	0.039	NA	Emission Reduction	0.90%
Yr-2040	50%	N2OEm_Red	0.026	NA	Emission Reduction	0.60%
Yr-2040	50%	Elec_BC_Prod	0.093	9.6E-03	Emission Reduction	2.10%
Yr-2040	50%	Em_Grind	-0.025	4.5E-03	Emission	3.40%
Yr-2040	50%	Em_Spread	-0.023	5.9E-03	Emission	3.10%
Yr-2040	50%	Em_Transp	-0.011	1.2E-03	Emission	1.50%
Yr-2040	50%	Em_Pellet	-0.016	8.5E-04	Emission	2.10%
Yr-2040	50%	Elec_Deficit	-0.458	2.4E-02	Emission	60.90%
Yr-2040	50%	Em_Loader	-0.003	-2.5E-05	Emission	0.40%
Yr-2040	50%	Em_Building	-0.216	NA	Emission	28.60%
Yr-2040	25%	BC_Soil	2.150	8.1E-02	Emission Reduction	95.70%
Yr-2040	25%	Ferti_Red	0.038	NA	Emission Reduction	1.70%
Yr-2040	25%	N2OEm_Red	0.013	NA	Emission Reduction	0.60%
Yr-2040	25%	Elec_BC_Prod	0.047	4.7E-03	Emission Reduction	2.10%
Yr-2040	25%	Em_Grind	-0.013	2.3E-03	Emission	3.40%
Yr-2040	25%	Em_Spread	-0.012	2.9E-03	Emission	3.10%
Yr-2040	25%	Em_Transp	-0.006	6.1E-04	Emission	1.50%
Yr-2040	25%	Em_Pellet	-0.008	4.2E-04	Emission	2.10%
Yr-2040	25%	Elec_Deficit	-0.228	1.2E-02	Emission	60.90%
Yr-2040	25%	Em_Loader	-0.002	-1.3E-05	Emission	0.40%
Yr-2040	25%	Em_Building	-0.107	NA	Emission	28.60%
NG (75%) - Wind (25%)	100%	BC_Soil	8.620	3.2E-01	Emission Reduction	96.00%
NG (75%) - Wind (25%)	100%	Ferti_Red	0.040	NA	Emission Reduction	0.50%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
NG (75%) - Wind (25%)	100%	N2OEm_Red	0.055	NA	Emission Reduction	0.60%
NG (75%) - Wind (25%)	100%	Elec_BC_Prod	0.266	2.7E-02	Emission Reduction	3.00%
NG (75%) - Wind (25%)	100%	Em_Grind	-0.072	1.3E-02	Emission	2.40%
NG (75%) - Wind (25%)	100%	Em_Spread	-0.051	1.3E-02	Emission	1.70%
NG (75%) - Wind (25%)	100%	Em_Transp	-0.025	2.6E-03	Emission	0.80%
NG (75%) - Wind (25%)	100%	Em_Pellet	-0.046	2.4E-03	Emission	1.50%
NG (75%) - Wind (25%)	100%	Elec_Deficit	-2.257	8.6E-02	Emission	76.00%
NG (75%) - Wind (25%)	100%	Em_Loader	-0.007	-5.5E-05	Emission	0.20%
NG (75%) - Wind (25%)	100%	Em_Building	-0.513	NA	Emission	17.30%
NG (75%) - Wind (25%)	50%	BC_Soil	4.312	1.6E-01	Emission Reduction	95.90%
NG (75%) - Wind (25%)	50%	Ferti_Red	0.039	NA	Emission Reduction	0.90%
NG (75%) - Wind (25%)	50%	N2OEm_Red	0.026	NA	Emission Reduction	0.60%
NG (75%) - Wind (25%)	50%	Elec_BC_Prod	0.122	1.2E-02	Emission Reduction	2.70%
NG (75%) - Wind (25%)	50%	Em_Grind	-0.033	5.9E-03	Emission	3.60%
NG (75%) - Wind (25%)	50%	Em_Spread	-0.023	5.8E-03	Emission	2.50%
NG (75%) - Wind (25%)	50%	Em_Transp	-0.011	1.2E-03	Emission	1.20%
NG (75%) - Wind (25%)	50%	Em_Pellet	-0.021	1.1E-03	Emission	2.30%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
NG (75%) - Wind (25%)	50%	Elec_Deficit	-0.600	3.1E-02	Emission	64.70%
NG (75%) - Wind (25%)	50%	Em Loader	-0.003	-2.5E-05	Emission	0.30%
NG (75%) - Wind (25%)	50%	Em_Building	-0.236	NA	Emission	25.40%
NG (75%) - Wind (25%)	25%	BC_Soil	2.149	8.0E-02	Emission Reduction	95.10%
NG (75%) - Wind (25%)	25%	Ferti_Red	0.038	NA	Emission Reduction	1.70%
NG (75%) - Wind (25%)	25%	N2OEm_Red	0.013	NA	Emission Reduction	0.60%
NG (75%) - Wind (25%)	25%	Elec_BC_Prod	0.061	6.1E-03	Emission Reduction	2.70%
NG (75%) - Wind (25%)	25%	Em_Grind	-0.017	2.9E-03	Emission	3.60%
NG (75%) - Wind (25%)	25%	Em_Spread	-0.011	2.9E-03	Emission	2.50%
NG (75%) - Wind (25%)	25%	Em_Transp	-0.006	6.0E-04	Emission	1.20%
NG (75%) - Wind (25%)	25%	Em_Pellet	-0.010	5.5E-04	Emission	2.30%
NG (75%) - Wind (25%)	25%	Elec_Deficit	-0.299	1.5E-02	Emission	64.70%
NG (75%) - Wind (25%)	25%	Em Loader	-0.002	-1.3E-05	Emission	0.30%
NG (75%) - Wind (25%)	25%	Em_Building	-0.117	NA	Emission	25.40%
Wind	100%	BC_Soil	8.630	3.3E-01	Emission Reduction	96.80%
Wind	100%	Ferti_Red	0.040	NA	Emission Reduction	0.50%
Wind	100%	N2OEm_Red	0.055	NA	Emission Reduction	0.60%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
Wind	100%	Elec_BC_Prod	0.187	1.8E-02	Emission Reduction	2.10%
Wind	100%	Em_Grind	-0.051	8.9E-03	Emission	2.30%
Wind	100%	Em_Spread	-0.050	1.3E-02	Emission	2.30%
Wind	100%	Em_Transp	-0.025	2.6E-03	Emission	1.10%
Wind	100%	Em_Pellet	-0.032	1.6E-03	Emission	1.50%
Wind	100%	Elec_Deficit	-1.589	5.0E-02	Emission	71.80%
Wind	100%	Em Loader	-0.007	-5.5E-05	Emission	0.30%
Wind	100%	Em_Building	-0.458	NA	Emission	20.70%
Wind	50%	BC_Soil	4.314	1.6E-01	Emission Reduction	96.60%
Wind	50%	Ferti_Red	0.039	NA	Emission Reduction	0.90%
Wind	50%	N2OEm_Red	0.026	NA	Emission Reduction	0.60%
Wind	50%	Elec_BC_Prod	0.086	8.4E-03	Emission Reduction	1.90%
Wind	50%	Em_Grind	-0.023	4.1E-03	Emission	3.30%
Wind	50%	Em_Spread	-0.023	5.8E-03	Emission	3.30%
Wind	50%	Em_Transp	-0.011	1.2E-03	Emission	1.60%
Wind	50%	Em_Pellet	-0.015	7.2E-04	Emission	2.10%
Wind	50%	Elec_Deficit	-0.422	2.0E-02	Emission	59.60%
Wind	50%	Em Loader	-0.003	-2.5E-05	Emission	0.40%
Wind	50%	Em_Building	-0.211	NA	Emission	29.70%
Wind	25%	BC_Soil	2.150	8.0E-02	Emission Reduction	95.80%
Wind	25%	Ferti_Red	0.038	NA	Emission Reduction	1.70%
Wind	25%	N2OEm_Red	0.013	NA	Emission Reduction	0.60%
Wind	25%	Elec_BC_Prod	0.043	4.2E-03	Emission Reduction	1.90%
Wind	25%	Em_Grind	-0.012	2.1E-03	Emission	3.30%

Marginal Energy	Biochar Production	Processes	Value s (t CO₂ ha⁻¹)	SD (t CO₂ ha⁻¹)	Group	Contribution within Group
Wind	25%	Em_Spread	-0.011	2.9E-03	Emission	3.30%
Wind	25%	Em_Transp	-0.006	6.0E-04	Emission	1.60%
Wind	25%	Em_Pellet	-0.007	3.5E-04	Emission	2.10%
Wind	25%	Elec_Deficit	-0.210	1.0E-02	Emission	59.60%
Wind	25%	Em_Loader	-0.002	-1.3E-05	Emission	0.40%
Wind	25%	Em_Building	-0.105	NA	Emission	29.70%

C.6 References

Alves, M. *et al.* (2015) ‘Surplus electricity production in sugarcane mills using residual bagasse and straw as fuel’, *Energy*. Elsevier Ltd, 91, pp. 751–757. doi: 10.1016/j.energy.2015.08.101.

AMISY (2020a) *High Efficiency Vertical Ring Die Wood Pellet Machine, Wood Pellet Mill*. Available at: <https://www.wood-pellet-mill.com/product/wood-pellet-mill/vertical-ring-die-wood-pellet-mill.html> (Accessed: 15 January 2020).

AMISY (2020b) *Wood Hammer Mill, Customized Wood Crushing Machine for Wood Pellet Plant*. Available at: <https://www.wood-pellet-mill.com/product/wood-crusher/wood-hammer-mill.html> (Accessed: 15 January 2020).

Azzi, E. S., Karlton, E. and Sundberg, C. (2019) ‘Prospective Life Cycle Assessment of Large-Scale Biochar Production and Use for Negative Emissions in Stockholm’, *Environmental Science & Technology*. American Chemical Society, 53(14), pp. 8466–8476. doi: 10.1021/acs.est.9b01615.

Bartocci, P. *et al.* (2016) ‘Biochar pellet carbon footprint’, *Chemical Engineering Transactions*, 50, pp. 217–222. doi: 10.3303/CET1650037.

Basu, P. (2010) *Biomass Gasification and Pyrolysis*. Elsevier. doi: 10.1016/C2009-0-20099-7.

Bordonal, R. de O. *et al.* (2018) ‘Sustainability of sugarcane production in Brazil. A review’, *Agronomy for Sustainable Development*. Agronomy for Sustainable

Development, 38(2). doi: 10.1007/s13593-018-0490-x.

Brassard, P. *et al.* (2018) 'Pyrolysis of switchgrass in an auger reactor for biochar production: A greenhouse gas and energy impacts assessment', *Biomass and Bioenergy*. Elsevier Ltd, 116(June), pp. 99–105. doi: 10.1016/j.biombioe.2018.06.007.

Bréon, F.-M. *et al.* (2013) 'Anthropogenic and natural radiative forcing', in *Climate Change 2013 the Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, pp. 659–740. doi: 10.1017/CBO9781107415324.018.

Cardoso, T. F. *et al.* (2018) 'Economic, environmental, and social impacts of different sugarcane production systems', *Biofuels, Bioproducts and Biorefining*, 12(1), pp. 68–82. doi: 10.1002/bbb.1829.

Cardoso, T. F. *et al.* (2019) 'A regional approach to determine economic, environmental and social impacts of different sugarcane production systems in Brazil', *Biomass and Bioenergy*. Elsevier Ltd, 120(April 2018), pp. 9–20. doi: 10.1016/j.biombioe.2018.10.018.

Chagas, M. F. *et al.* (2016) 'Environmental and economic impacts of different sugarcane production systems in the ethanol biorefinery', *Biofuels, Bioproducts and Biorefining*, 6(3), pp. 246–256. doi: 10.1002/bbb.

CONAB (2018) *Acompanhamento da Safra Brasileira - Cana-de-açúcar - Terceiro levantamento | Dezembro 2018*. Available at: <https://www.conab.gov.br/info-agro/safras/cana>.

Cross, A. and Sohi, S. P. (2013) 'A method for screening the relative long-term stability of biochar', *GCB Bioenergy*, 5(2), pp. 215–220. doi: 10.1111/gcbb.12035.

Dale, A. T. *et al.* (2013) 'Modeling future life-cycle greenhouse gas emissions and environmental impacts of electricity supplies in Brazil', *Energies*, 6(7), pp. 3182–3208. doi: 10.3390/en6073182.

Dias, M. O. S. *et al.* (2011) 'Second generation ethanol in Brazil: Can it compete with

electricity production?’, *Bioresource Technology*. Elsevier Ltd, 102(19), pp. 8964–8971. doi: 10.1016/j.biortech.2011.06.098.

EPE (2017) *The Ten-Year Expansion Plan 2027*. Rio de Janeiro. Available at: http://www.epe.gov.br/sites-en/publicacoes-dados-abertos/publicacoes/PublicacoesArquivos/publicacao-195/PDE_2027_Executive_Summary.pdf.

FAO (2019) *FAOSTAT, Food and Agriculture Organization of the United Nations - Statistic Division*. Available at: <http://www.fao.org/faostat/en/#data/QC> (Accessed: 21 October 2019).

Gabra, M. *et al.* (2001) ‘Evaluation of cyclone gasifier performance for gasification of sugar cane residue - Part 1: Gasification of bagasse’, *Biomass and Bioenergy*, 21(5), pp. 351–369. doi: 10.1016/S0961-9534(01)00043-5.

George, P. A. O. *et al.* (2010) ‘Residue from sugarcane juice filtration (filter cake): Energy use at the sugar factory’, *Waste and Biomass Valorization*, 1(4), pp. 407–413. doi: 10.1007/s12649-010-9046-2.

Gomaa, M. R. *et al.* (2019) ‘Evaluating the Environmental Impacts and Energy Performance of a Wind Farm System Utilizing the Life-Cycle Assessment Method: A Practical Case Study’, *Energies*, 12(17), p. 3263. doi: 10.3390/en12173263.

Gomes, L. C. *et al.* (2019) ‘Modelling and mapping soil organic carbon stocks in Brazil’, *Geoderma*. Elsevier, 340(December 2017), pp. 337–350. doi: 10.1016/j.geoderma.2019.01.007.

Grisso, R. *et al.* (2010) ‘Predicting Diesel Fuel Consumption’, *Fuel*, (June), pp. 2004–2006.

Guezuraga, B., Zauner, R. and Pölz, W. (2012) ‘Life cycle assessment of two different 2 MW class wind turbines’, *Renewable Energy*. Elsevier Ltd, 37(1), pp. 37–44. doi: 10.1016/j.renene.2011.05.008.

Hammond, J. *et al.* (2011) ‘Prospective life cycle carbon abatement for pyrolysis biochar systems in the UK’, *Energy Policy*. Elsevier, 39(5), pp. 2646–2655. doi:

10.1016/j.enpol.2011.02.033.

Handler, F. and Nadlinger, M. (2012) *Strategies for saving fuel with tractors*.

Hassuani, S. J., Leal, M. R. L. V. and Macedo, I. de C. (2005) *Biomass Power Generation - Sugar cane bagasse and trash*. First. Edited by S. J. Hassuani, M. R. L. V. Leal, and I. de C. Macedo. Piracicaba Brasil: PNUD - Programa das Nações Unidas para o Desenvolvimento And CTC - Centro de Tecnologia Canavieira.

Kambo, H. S. and Dutta, A. (2015) 'A comparative review of biochar and hydrochar in terms of production, physico-chemical properties and applications', *Renewable and Sustainable Energy Reviews*. Elsevier, 45, pp. 359–378. doi: 10.1016/j.rser.2015.01.050.

Khatiwada, D. *et al.* (2016) 'Optimizing ethanol and bioelectricity production in sugarcane biorefineries in Brazil', *Renewable Energy*. Elsevier Ltd, 85, pp. 371–386. doi: 10.1016/j.renene.2015.06.009.

Klein, B. C. *et al.* (2018) 'Beyond ethanol, sugar, and electricity: a critical review of product diversification in Brazilian sugarcane mills', *Biofuels, Bioproducts and Biorefining*, pp. 1–13. doi: 10.1002/BBB.1969.

Koga, N. *et al.* (2003) 'Fuel consumption-derived CO₂ emissions under conventional and reduced tillage cropping systems in northern Japan', *Agriculture, Ecosystems and Environment*, 99(1–3), pp. 213–219. doi: 10.1016/S0167-8809(03)00132-4.

Lehmann, J. (2007) 'Bio-energy in the black', *Frontiers in Ecology and the Environment*, p. 6. doi: 10.1890/1540-9295(2007)5[381:BITB]2.0.CO;2.

Leme, M. M. V. *et al.* (2018) 'Electricity generation from pyrolysis gas produced in charcoal manufacture: Technical and economic analysis', *Journal of Cleaner Production*, 194, pp. 219–242. doi: 10.1016/j.jclepro.2018.05.101.

Lucena, J. D. A. Y. and Lucena, K. Â. A. (2019) 'Wind energy in Brazil: An overview and perspectives under the triple bottom line', *Clean Energy*, 3(2), pp. 69–84. doi: 10.1093/ce/zkz001.

Lutz Böhle (2014) *Biological and chemical parameters and life cycle assessment of the integrated generation of solid fuel and biogas from biomass*.

- Macedo, I. C., Seabra, J. E. A. and Silva, J. E. A. R. (2007) 'Green house gases emissions in the production and use of ethanol from sugarcane in Brazil: The 2005/2006 averages and a prediction for 2020', *Biomass and Bioenergy*, 32(7), pp. 582–595. doi: 10.1016/j.biombioe.2007.12.006.
- Masiello, C. A. and Druffel, E. R. M. (1998) 'Black Carbon in Deep-Sea Sediments', *Science*, 280(5371), pp. 1911–1913. doi: 10.1126/science.280.5371.1911.
- Menandro, L. M. S. *et al.* (2017) 'Comprehensive assessment of sugarcane straw: implications for biomass and bioenergy production', *Biofuels, Bioproducts and Biorefining*, 11(3), pp. 488–504. doi: 10.1002/bbb.1760.
- Muñoz, E. *et al.* (2017) 'Environmental hotspots in the life cycle of a biochar-soil system', *Journal of Cleaner Production*, 158, pp. 1–7. doi: 10.1016/j.jclepro.2017.04.163.
- Nihei, T., Hayashi, H. and Shirota, R. (2015) 'Characteristics of sugarcane production in the State of São Paulo , Brazil', *Geographical Space*, pp. 53–80. doi: 10.24586/jags.8.1_53.
- Oebels, K. B. and Pacca, S. (2013) 'Life cycle assessment of an onshore wind farm located at the northeastern coast of Brazil', *Renewable Energy*. Elsevier Ltd, 53, pp. 60–70. doi: 10.1016/j.renene.2012.10.026.
- Oldfield, T. L. *et al.* (2018) 'Biochar, compost and biochar-compost blend as options to recover nutrients and sequester carbon', *Journal of Environmental Management*, 218, pp. 465–476. doi: 10.1016/j.jenvman.2018.04.061.
- de Oliveira, B. G. *et al.* (2013) 'Soil greenhouse gas fluxes from vinasse application in Brazilian sugarcane areas', *Geoderma*, 200–201, pp. 77–84. doi: 10.1016/j.geoderma.2013.02.005.
- de Oliveira Vilela, A. *et al.* (2014) 'A new technology for the combined production of charcoal and electricity through cogeneration', *Biomass and Bioenergy*, 69, pp. 222–240. doi: 10.1016/j.biombioe.2014.06.019.
- Olivério, J. L. and Ferreira, F. M. (2010) 'Cogeneration - A new source of income for

sugar and ethanol mills’, in *Proc. Int. Soc. Sugar Cane Technol.*, pp. 1–13. Available at: [https://www.semanticscholar.org/paper/COGENERATION-A-NEW-SOURCE-OF-INCOME-FOR-SUGAR-AND-Olivério-](https://www.semanticscholar.org/paper/COGENERATION-A-NEW-SOURCE-OF-INCOME-FOR-SUGAR-AND-Olivério-Ferreira/7ed359fa22554680fe52445155c3d7fe9106f308)

[Ferreira/7ed359fa22554680fe52445155c3d7fe9106f308](https://www.semanticscholar.org/paper/COGENERATION-A-NEW-SOURCE-OF-INCOME-FOR-SUGAR-AND-Olivério-Ferreira/7ed359fa22554680fe52445155c3d7fe9106f308).

Park, J. *et al.* (2014) ‘Slow pyrolysis of rice straw: Analysis of products properties, carbon and energy yields’, *Bioresource Technology*. Elsevier Ltd, 155, pp. 63–70. doi: 10.1016/j.biortech.2013.12.084.

Pippo, W. A. and Luengo, C. A. (2013) ‘Sugarcane energy use: Accounting of feedstock energy considering current agro-industrial trends and their feasibility’, *International Journal of Energy and Environmental Engineering*, 4(1), pp. 1–13. doi: 10.1186/2251-6832-4-10.

Popin, G. V. *et al.* (2020) ‘Sugarcane straw management for bioenergy: effects of global warming on greenhouse gas emissions and soil carbon storage’, *Mitigation and Adaptation Strategies for Global Change*. Mitigation and Adaptation Strategies for Global Change, 25(4), pp. 559–577. doi: 10.1007/s11027-019-09880-7.

Quirk, R. G. *et al.* (2012) ‘Utilization of Biochar in Sugarcane and Sugar-Industry Management’, *Sugar Tech*, 14(4), pp. 321–326. doi: 10.1007/s12355-012-0158-9.

Rajabi Hamedani, S. *et al.* (2019) ‘Life Cycle Assessment and Environmental Valuation of Biochar Production: Two Case Studies in Belgium’, *Energies*, 12(11), p. 2166. doi: 10.3390/en12112166.

Raveendran, K. and Ganesh, A. (1996) ‘Heating value of biomass and biomass pyrolysis products’, *Fuel*, 75(15), pp. 1715–1720. doi: 10.1016/S0016-2361(96)00158-5.

Roberts, K. G. *et al.* (2010) ‘Life Cycle Assessment of Biochar Systems: Estimating the Energetic, Economic, and Climate Change Potential’, *Environmental Science and Technology*, 44, pp. 827–833. doi: 10.1021/es902266r.

Rosado, L. P. *et al.* (2017) ‘Life cycle assessment of natural and mixed recycled aggregate production in Brazil’, *Journal of Cleaner Production*. Elsevier Ltd, 151, pp. 634–642. doi: 10.1016/j.jclepro.2017.03.068.

- Roy, P. and Dias, G. (2017) 'Prospects for pyrolysis technologies in the bioenergy sector: A review', *Renewable and Sustainable Energy Reviews*. Elsevier Ltd, 77(September), pp. 59–69. doi: 10.1016/j.rser.2017.03.136.
- Sacchi, R. *et al.* (2019) 'Exploring technologically, temporally and geographically-sensitive life cycle inventories for wind turbines: A parameterized model for Denmark', *Renewable Energy*, 132, pp. 1238–1250. doi: 10.1016/j.renene.2018.09.020.
- Sampaio, I. L. M. *et al.* (2019) 'Electricity Production from Sugarcane Straw Recovered Through Bale System: Assessment of Retrofit Projects', *BioEnergy Research*. BioEnergy Research, 12(4), pp. 865–877. doi: 10.1007/s12155-019-10014-9.
- Seabra, J. E. A. *et al.* (2011) 'Life cycle assessment of Brazilian sugarcane products: GHG emissions and energy use', *Biofuels, Bioproducts and Biorefining*, 5(5), pp. 519–532. doi: 10.1002/bbb.289.
- Seabra, J. E. A. and Macedo, I. C. (2011) 'Comparative analysis for power generation and ethanol production from sugarcane residual biomass in Brazil', *Energy Policy*. Elsevier, 39(1), pp. 421–428. doi: 10.1016/j.enpol.2010.10.019.
- Shen, Y. *et al.* (2020) 'Impacts of biochar concentration on the growth performance of a leafy vegetable in a tropical city and its global warming potential', *Journal of Cleaner Production*. Elsevier Ltd, 264, p. 121678. doi: 10.1016/j.jclepro.2020.121678.
- Silva, A. G. B. *et al.* (2019) 'How Much Sugarcane Straw is Needed for Covering the Soil?', *BioEnergy Research*. BioEnergy Research, 12(4), pp. 858–864. doi: 10.1007/s12155-019-10008-7.
- Theodor Rudorff, B. F. (2014) *Canasat - INPE*. Available at: <http://www.dsr.inpe.br/laf/canasat/tabelas.html> (Accessed: 27 August 2019).
- Thers, H. *et al.* (2019) 'Biochar potentially mitigates greenhouse gas emissions from cultivation of oilseed rape for biodiesel', *Science of The Total Environment*. Elsevier B.V., 671, pp. 180–188. doi: 10.1016/j.scitotenv.2019.03.257.
- Vandepaer, L. *et al.* (2019) 'The integration of long-term marginal electricity supply mixes in the ecoinvent consequential database version 3.4 and examination of modeling

choices’, *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 24(8), pp. 1409–1428. doi: 10.1007/s11367-018-1571-4.

Vasconcelos, A. L. S. *et al.* (2018) ‘Greenhouse gas emission responses to sugarcane straw removal’, *Biomass and Bioenergy*. Elsevier Ltd, 113(February), pp. 15–21. doi: 10.1016/j.biombioe.2018.03.002.

Waack, R. S. and Neves, M. F. (1998) *Competitividade no Agribusiness Brasileiro - sistema agroindustrial da cana-de-açúcar, Competitividade no Agribusiness Brasileiro*. Sao Paulo - Brazil. Available at: <https://www.wageningenacademic.com/doi/pdf/10.22434/IFAMR2015.0195?download=true>.

Waldheim, L., Monis, M. and Verde Leal, M. R. (2001) ‘Biomass Power Generation: Sugar Cane Bagasse and Trash’, in Bridgwater, A. . (ed.) *Progress in Thermochemical Biomass Conversion*. Oxford, UK: Blackwell Science Ltd, pp. 509–523. Available at: http://www.sucre-ethique.org/IMG/pdf/CTC_energy_-_biomass_1_.pdf.

Wang, Z. *et al.* (2014) ‘Effects of co-produced biochar in life cycle greenhouse gas emissions of pyrolysis-derived renewable fuels’, *Biofuels, Bioproducts and Biorefining*, 8, pp. 189–204. doi: 10.1002/bbb.

Weidema, B. P. *et al.* (2013) *Data quality guideline for the ecoinvent database version 3*. Available at: <http://www.ecoinvent.org/database/methodology-of-ecoinvent-3/methodology-of-ecoinvent-3.html>.

Weisser, D. (2007) ‘A guide to life-cycle greenhouse gas (GHG) emissions from electric supply technologies’, *Energy*, 32(9), pp. 1543–1559. doi: 10.1016/j.energy.2007.01.008.

Wernet, G. *et al.* (2016) ‘The ecoinvent database version 3 (part I): overview and methodology’, *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 21(9), pp. 1218–1230. doi: 10.1007/s11367-016-1087-8.

Yang, Q. *et al.* (2016) ‘Greenhouse gas emissions of a biomass-based pyrolysis plant in China’, *Renewable and Sustainable Energy Reviews*. Elsevier, 53, pp. 1580–1590. doi: 10.1016/j.rser.2015.09.049.

Yang, Y. *et al.* (2017) 'Combined heat and power from the intermediate pyrolysis of biomass materials: performance, economics and environmental impact', *Applied Energy*. Elsevier Ltd, 191, pp. 639–652. doi: 10.1016/j.apenergy.2017.02.004.

Yang, Y. *et al.* (2018) 'A techno-economic analysis of energy recovery from organic fraction of municipal solid waste (MSW) by an integrated intermediate pyrolysis and combined heat and power (CHP) plant', *Energy Conversion and Management*. Elsevier, 174(August), pp. 406–416. doi: 10.1016/j.enconman.2018.08.033.

You, S. *et al.* (2017) 'Towards practical application of gasification : A critical review from syngas and biochar perspectives', *Critical Reviews in Environmental Science and Technology*. Taylor & Francis, 0(0), pp. 1–48. doi: 10.1080/10643389.2018.1518860.

Zimmerman, A. R. (2010) 'Abiotic and Microbial Oxidation of Laboratory-Produced Black Carbon (Biochar)', *Environmental science & technology*, 44(4), pp. 1295–1301. doi: 10.1021/es903140c

Appendix D Supplementary Information – Chapter 5: Assessing the carbon capture potential of reforestation projects

D.1 Meteorological and soil data

Table D.1: Latitude, longitude, yearly average temperature and monthly precipitation values from the ClimWat database (FAO, 2006) of the meteorological station considered in this study. These meteorological values were assumed in repetition for the modelling activities.

Meteorological station		Puerto Maldonado	
Altitude	266 m.a.s.l.		
Latitude	-12.63	Longitude	-69.2
Month	Average Temperature (°C)	Average Precipitation (mm)	Evapotranspiration (mm)*
January	26.3	317	143.2
February	25.9	317	120.7
March	25.65	273	125.5
April	25.5	149	116
May	24.45	85	101.7
June	23.25	51	82.2
July	23.1	55	83.6
August	24.25	66	100.3
September	25.8	100	122.8
October	26.3	153	139.3
November	26.65	223	144.1
December	26.35	285	144.9

*Thornthwaite potential evapotranspiration was calculated using the R software package “SPEI” (Beguería and Vicente-Serrano, 2017)

Table D.2: Soil characteristics of the case study plot and the reference forest (Román-Dañobeytia et al., 2021).

Characteristic	Unit	Reference Forest	Open sandy area (case study)
pH	/	4.8	5.6
Organic matter	%	3	0.2
CEC	Cmol kg ⁻¹	17.4	5.9
Sand content	%	60	91.7
Clay content	%	13.6	5.1
Silt content	%	26.5	3.1
Bulk density	g cm ⁻³	1.11	1.57
Carbon stock (0-20 cm)			
*	t ha ⁻¹	66.63	6.35

* Calculated

D.2 Nursery

Table D.3: Elements needed for the growth of 1000 seedlings in the nursery (Farfan, 2020).

Amendment	Weight for 1000 seedlings [kg]
Semi carbonized rice husk	100
Saw dust	75
Poultry manure	150
Cow manure	100
Compost	100
NPK 16-8-12 (Basacote)	3
Fertiliser (Magnocal)	3
Diammonium phosphate	3
NPK 14-16-10 (Yaramilla)	3
Potassium chloride	0.3
Fungicide (Cupravit - copper oxychloride)	0.2
Fungicide (Benzomil - C ₁₄ H ₁₈ N ₄ O ₃)	0.3

The nursery pays 9 soles per month for the electricity (Jhon Farfan, *pers. com.*). In 2013 the price per kWh in Madre de Dios was 0.44 soles per kWh (MINEM, 2013).

D.3 Soil carbon modelling data

Table D.4: Data for soil and biomass carbon modelling activities.

Parameter	Value	Unit	Note	Reference
CINCIA planting density	1111	Seedling ha ⁻¹		(Román-Dañobeytia <i>et al.</i> , 2021)
Biochar yield	30	%		(IPCC, 2019a)
soil thickness	20	cm		(Román-Dañobeytia <i>et al.</i> , 2021)
DPM:RPM ratio	0.25	/	For a deciduous or tropical woodland	(Coleman and Jenkinson, 2014)
Clay content	5.13	%		(Román-Dañobeytia <i>et al.</i> , 2021)
Degraded soil carbon stock	6.35	t C ha ⁻¹	Calculated	(Román-Dañobeytia <i>et al.</i> , 2021)
Adjacent forest soil carbon stock	66.63	t C ha ⁻¹	Calculated	(Román-Dañobeytia <i>et al.</i> , 2021)
Time between minimum and maximum litter input	40	years		(Jones <i>et al.</i> , 2019)
Minimum litter input	1.203	t C ha ⁻¹	Inverse RothC	(Coleman and Jenkinson, 2014)
Maximum litter input	11.4258	t C ha ⁻¹	Inverse RothC	(Coleman and Jenkinson, 2014)
Biochar Carbon content	87.6	%		(Lefebvre <i>et al.</i> , 2019)
Biochar weight considered labile	5	%		(Calvelo Pereira <i>et al.</i> , 2011)
Biochar weight considered recalcitrant	95	%		(Calvelo Pereira <i>et al.</i> , 2011)

Parameter	Value	Unit	Note	Reference
Biochar C lost after 100 years	15	%		(IPCC, 2019a)
Median number of trees in old growth tropical forest	565	/		(ter Steege <i>et al.</i> , 2013)

D.4 LCA data

Table D.5: Data and emission factors used for the LCA.

Parameter	Value	Unit	Reference	Comment
Distance biomass collection to biochar production plant	21	km	(Google, 2018)	
Distance biochar production plant to reforestation plot	36	km	(Google, 2018)	
Distance nursery to reforestation plot	128	km	(Google, 2018)	
Boat fuel consumption to reforestation plot	4.25	litres	(Jhon Farfan, pers. com.)	
Weight of seedlings	110	g seedling ⁻¹	(Jhon Farfan, pers. com.)	
GWP - CH ₄	28	kg CO ₂ e kg ⁻¹	(Myhre et al., 2013)	
GWP - NO _x	-11	kg CO ₂ e kg ⁻¹	(Myhre et al., 2013)	
GWP - CO	2.65	kg CO ₂ e kg ⁻¹	(Myhre et al., 2013)	
GWP - N ₂ O	265	kg CO ₂ e kg ⁻¹	(Myhre et al., 2013)	
Density gasoline low sulphur	850	kg m ⁻³	(Shell, 2014)	
Emission factor gasoline	0.88	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Petrol, low-sulfur {RoW} market for Alloc Def, S

Parameter	Value	Unit	Reference	Comment
Emission factor loader operation	3.2	kg CO ₂ e l of diesel ⁻¹	(Wernet et al., 2016)	Diesel, combusted in industrial equipment/US
Emission factor electricity	0.392	kg CO ₂ e kWh ⁻¹	(Wernet et al., 2016)	Electricity, low voltage {PE} market for Alloc Def, S
Emission factor human labour	0.123	kg CO ₂ e hour of work ⁻¹	(Rugani, Panasiuk and Benetto, 2012)	Using the purchasing power parity of Peru
Emission factor transportation	0.2628	kg CO ₂ e tkm ⁻¹	(Wernet et al., 2016)	Transport, freight, lorry 7.5-16 metric ton, EURO3 {RoW} transport, freight, lorry 7.5-16 metric ton, EURO3 Alloc Def, S – Added 20% to account for rough terrain
Loading operation	0.19	l of diesel tBC ⁻¹	(Rosado et al., 2017)	
Emission factor open dump of husks	0.0853	kg CO ₂ e kg of husks ⁻¹	(Wernet et al., 2016)	Waste wood, untreated {GLO} treatment of waste wood, untreated, open dump, very wet infiltration class (1000mm) Cut-off, U (of project Ecoinvent 3 - allocation, cut-off by classification - unit)
Methane production during pyrolysis	30 ± 60	g CH ₄ kg biochar ⁻¹	(IPCC, 2019b)	
Nitrogen oxides production during pyrolysis	0.4 ± 0.3	g NO _x kg biochar ⁻¹	(IPCC, 2019b)	
Carbon monoxide production during pyrolysis	54 ± 35.1	g CO kg biochar ⁻¹	(IPCC, 2019b)	
Nursery amendment	150	km	Estimated	

Appendix D

Parameter	Value	Unit	Reference	Comment
transportation to city				
Electricity for nursery	0.09	kWh seedling ⁻¹	Calculated	
Team number for field work and maintenance	8	people	(Jhon Farfan, pers. com.)	
Weight of team member for field work and maintenance	85	kg	Estimated – including gears	
Number of days needed for field work and number of maintenances	3	day ha ⁻¹	(Jhon Farfan, pers. com.)	
Saw dust	0.0293	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Saw dust, wet, measured as dry mass {GLO} market for Alloc Def, S
Poultry manure	0.196	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Poultry manure, fresh {GLO} nutrient supply from poultry manure, fresh Conseq, S
Cow manure	0.0388	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Manure, solid, cattle {GLO} nutrient supply from manure, solid, cattle Conseq, S
Compost	0.211	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Biowaste {RoW} treatment of, composting Alloc Def, S
NPK 16-8-12 (Basacote)	0.903	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	NPK compound (NPK 15-15-15), at regional storehouse/RER Mass
Fertiliser (Magnocal)	0.903	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	NPK compound (NPK 15-15-15), at regional storehouse/RER Mass
Diammonium phosphate	2.87	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Nitrogen fertiliser, as N {RoW} diammonium

Parameter	Value	Unit	Reference	Comment
				phosphate production Alloc Def, S
NPK 14-16-10 (Yaramilla)	0.903	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	NPK compound (NPK 15-15-15), at regional storehouse/RER Mass
Potassium chloride	0.568	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Potassium chloride, as K ₂ O {GLO} market for Alloc Def, S
Fungicide (Cupravit - copper oxychloride)	11	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Pesticide, unspecified {GLO} market for Alloc Def, S
Fungicide (Benzomil - C ₁₄ H ₁₈ N ₄ O ₃)	8.43	kg CO ₂ e kg ⁻¹	(Wernet et al., 2016)	Benzimidazole-compound {GLO} market for Alloc Def, S

D.5 CINCIA's LCA contribution analysis

Table D.6: Contribution analysis of the LCA related to the setup of CINCIA's reforestation plot.

Processes	Sub-Process	Values (kg C ha ⁻¹)	SD (kg C ha ⁻¹)	Group	Contribution within Group
Biochar plant	Biomass transportation	4.87	0.447	Emission	0.9%
Biochar plant	Loader Operations	1.21	0.0198	Emission	0.2%
Biochar plant	Pyrolysis emissions	298	161	Emission	57.2%
Biochar plant	Biochar Transport	4.92	0.0971	Emission	0.9%
Nursery	Amendments	55.9	16.2	Emission	10.7%
Nursery	Electricity	11.0	/	Emission	2.1%
Nursery	Seedling transport	3.11	0.0380	Emission	0.6%
Field works	On-site fertilizers	25.2	0.0140	Emission	4.8%
Field works	Team Transportation	13.8	0.282	Emission	2.7%

Processes	Sub-Process	Values (kg C ha ⁻¹)	SD (kg C ha ⁻¹)	Group	Contribution within Group
Field works	Nitrous Oxide emissions	76.1	23.3	Emission	14.6%
Field works	Human labour	6.50	/	Emission	1.2%
Field maintenance	Team Transportation	13.8	0.282	Emission	2.7%
Field maintenance	Human labour	6.50	/	Emission	1.2%
Avoided composting	Avoided composting	86.2	/	Emission reduction	8.4%
Biochar in soil	Biochar in soil	942	/	Emission reduction	91.6%

D.6 Distribution - Monte Carlo

Methane emissions during pyrolysis		Unit	Comments
Reported in the LCA	30	kg tBC ⁻¹	Literature
Shape	0.25		
Scale	120		
Distribution	Gamma		
N	3		

Nitrogen oxides emissions during pyrolysis		Unit	Comments
Reported in the LCA	0.4	kg tBC ⁻¹	Literature
Shape	1.78		
Scale	0.225		
Distribution	Gamma		
N	3		

Carbon monoxide emissions during pyrolysis		Unit	Comments
Reported in the LCA	54	kg tBC ⁻¹	Literature
Shape	2.38		
Scale	22.7		
Distribution	Gamma		
N	3		

Emission factor direct emission of N ₂ O-N		Unit	Comments
Reported in the LCA	0.01	kg kg-N ⁻¹	Literature
Min	0.003		
Max	0.3		
Mode	0.01		
Distribution	Triangular		

N	1
---	---

Emission factor indirect emissions N₂O volatilisation		Unit	Comments
Reported in the LCA	0.01	kg kg-N ⁻¹	Literature
Min	0.002		
Max	0.05		
Mode	0.01		
Distribution	Triangular		
N	1		

Emission factor indirect emissions N₂O leaching		Unit	Comments
Reported in the LCA	0.0075	kg kg-N ⁻¹	Literature
Min	0.0005		
Max	0.025		
Mode	0.0075		
Distribution	Triangular		
N	1		

Fraction gasification volatilisation synthetic fertilizer N₂O-N		Unit	Comments
Reported in the LCA	0.1	/	Literature
Min	0.03		
Max	0.3		
Mode	0.1		
Distribution	Triangular		
N	1		

Fraction gasification volatilisation organic fertilizer N₂O-N		Unit	Comments
Reported in the LCA	0.2	/	Literature
Min	0.05		
Max	0.5		
Mode	0.2		
Distribution	Triangular		
N	1		

Fraction leaching N fertilizer		Unit	Comments
Reported in the LCA	0.3	/	Literature
Min	0.1		
Max	0.8		
Mode	0.3		
Distribution	Triangular		
N	1		

Distance between biomass and pyrolysis plant	Unit	Comments
---	------	----------

Appendix D

Reported in the LCA	20	km	Google Maps
Range	15 – 20 – 25	km	
Distribution	Uniform		
N	3		

Distance transport nursery amendment		Unit	Comments
Reported in the LCA	150	km	
Range	100 - 200	km	Arbitrary
Distribution	Uniform		
N	2		

Loader operation		Unit	Comments
Reported in the LCA	0.188	l diesel tBC ⁻¹	
Range	0.18 – 0.195	l diesel tBC ⁻¹	Literature
Distribution	Uniform		
N	2		

Emissions factor truck transportation		Unit	Comments
Reported in the LCA	0.2409	kg CO ₂ tkm ⁻¹	
Range	0.219 - 0.2628	kg CO ₂ tkm ⁻¹	EcoInvent and additional 20% for rough terrain
Distribution	Uniform		
N	2		

Emission factor carbon monoxide		Unit	Comments
Reported in the LCA	2.65	kg CO ₂ kg ⁻¹	
Range	2 – 3.3	kg CO ₂ kg ⁻¹	Literature
Distribution	Uniform		
N	2		

D.7 References

Beguéría, S. and Vicente-Serrano, S. M. (2017) ‘SPEI: Calculation of the Standardised Precipitation-Evapotranspiration Index.’ Available at: <https://cran.r-project.org/package=SPEI>.

Calvelo Pereira, R. *et al.* (2011) ‘Contribution to characterisation of biochar to estimate the labile fraction of carbon’, *Organic Geochemistry*. Elsevier Ltd, 42(11), pp. 1331–1342. doi: 10.1016/j.orggeochem.2011.09.002.

Coleman, K. and Jenkinson, D. S. (2014) *RothC - A model for the turnover of carbon in soil Model - Model description and users guide*. Harpenden: Routledge. Available at: https://www.rothamsted.ac.uk/sites/default/files/RothC_guide_WIN.pdf.

FAO (2006) 'CLIMWAT 2.0 for CROPWAT'. Rome: Water Development and Management Unit and the Climate Change and Bioenergy Unit of FAO. Available at: <http://www.fao.org/land-water/databases-and-software/climwat-for-cropwat/en/>.

Farfan, J. F. (2020) *Produccion de Plantones en el Vivero Tecnificado de Mazuko Para Recuperar Suelos Degradado por Minería Aluvial en Madre de Dios*. Puerto Maldonado.

Google (2018) *Google Earth Pro*. Available at: <https://www.google.com/earth/download/gep/agree.html>.

IPCC (2019a) 'Appendix 4: Method for Estimating the Change in Mineral Soil Organic Carbon Stocks from Biochar Amendments', in *2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories*, pp. 2–6.

IPCC (2019b) 'Volume 2, Chapter 4: Fugitive Emissions - 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories', *IPCC Guidelines for National Greenhouse Gas Inventories*, 2, p. 78. Available at: http://www.ipcc-nggip.iges.or.jp/public/2006gl/pdf/2_Volume2/V2_4_Ch4_Fugitive_Emissions.pdf.

Jones, I. L. *et al.* (2019) 'Above- and belowground carbon stocks are decoupled in secondary tropical forests and are positively related to forest age and soil nutrients respectively', *Science of the Total Environment*. Elsevier B.V., 697, p. 133987. doi: 10.1016/j.scitotenv.2019.133987.

Lefebvre, D. *et al.* (2019) 'Biochar Effects on Two Tropical Tree Species and Its Potential as a Tool for Reforestation', *Forests*. MDPI AG, 10(8), p. 678. doi: 10.3390/f10080678.

MINEM (2013) *Capítulo 5: Por qué cuesta la energía y cómo calculamos nuestra factura*, *Guía Inicial de la Energía*. Available at: http://www.minem.gob.pe/giee/pdf/GUIA_INICIAL_PRIMARIA_CAP5.pdf.

Myhre, G. *et al.* (2013) 'Anthropogenic and Natural Radiative Forcing.', in Jacob, D., Ravishankara, A. R., and Shine, K. (eds) *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. New York: Cambridge University Press. doi: 10.3390/jmse6040146.

Román-Dañobeytia, F. *et al.* (2021) ‘Survival and early growth of 51 tropical tree species in areas degraded by artisanal gold mining in the Peruvian Amazon’, *Ecological Engineering*. Elsevier B.V., 159(December 2020), p. 106097. doi: 10.1016/j.ecoleng.2020.106097.

Rosado, L. P. *et al.* (2017) ‘Life cycle assessment of natural and mixed recycled aggregate production in Brazil’, *Journal of Cleaner Production*. Elsevier Ltd, 151, pp. 634–642. doi: 10.1016/j.jclepro.2017.03.068.

Rugani, B., Panasiuk, D. and Benetto, E. (2012) ‘An input-output based framework to evaluate human labour in life cycle assessment’, *International Journal of Life Cycle Assessment*, 17(6), pp. 795–812. doi: 10.1007/s11367-012-0403-1.

Shell (2014) *Marine Fuels, Shell Trading and Supply*. Available at: <https://www.shell.com/business-customers/marine/fuel/ulsfo.html>.

ter Steege, H. *et al.* (2013) ‘Hyperdominance in the Amazonian Tree Flora’, *Science*, 342(6156), pp. 1243092–1243092. doi: 10.1126/science.1243092.

Wernet, G. *et al.* (2016) ‘The ecoinvent database version 3 (part I): overview and methodology’, *International Journal of Life Cycle Assessment*. The International Journal of Life Cycle Assessment, 21(9), pp. 1218–1230. doi: 10.1007/s11367-016-1087-8.

