Investigating microalgal carbon capture: an experimental and techno-economic study

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Abstract:

Power stations and industrial factories contribute a significant fraction of total anthropogenic CO_2 emissions, which need to be mitigated to reduce their effect on the climate. Commercially available methods of CCUS (Carbon capture and utilisation or storage) rely on toxic chemicals for the capture processes as well as the long-term storage of CO_2 gas. The use of photosynthetic microalgae for CCUS offers the potential for a sustainable capture system, which can both reduce emissions and produce renewable products. Previous studies have focused largely on the products (particularly biofuels) from microalgae, rather than their ability to capture CO_2. In this study, the green microalga *Chlorella* sp. was cultivated with CO_2 concentrations like those present in power-station flue gases (5-15 % v/v) to determine the optimal concentration for growth. The maximum growth rate (0.11 g L^{-1} day^{-1}) and final biomass concentration (2.11 g L^{-1}) for *Chlorella* sp. was found with continuous aeration of 5 % CO2. The CO_2 removal efficiency of the cultures monitored in real-time with a nondispersive infrared (NDIR) sensor. The average CO_2 removal efficiency at 5 % CO_2 was 17.5 % over the 14 days of continuous exposure. A techno-economic assessment of algal-CCUS was performed using the experimental results and a range of different financial and operational scenarios. The assessment highlights that microalgal strain choice and development for CO_2 removal is the key parameter for reducing the cost of the process. This, alongside the reduction of capital expenditure and increasing government incentives for reducing emissions will make algal-carbon capture economically feasible.

Keywords:

Chlorella sp., carbon capture and utilisation or storage, techno-economic assessment

1. Introduction:

Anthropogenic emissions of CO_2 are now a great concern due to their role in climate change. Carbon capture and utilisation and/or storage (CCUS) combines a group of technologies with the aim to reduce CO_2 emissions by capturing, transporting and utilise or store purified CO_2 [1,2]. These technologies allow for the continued use of power and industrial production methods reliant on fossil fuels, creating a pathway towards renewable energy with reductions in emissions now. Current, commercially available capture methods, have several drawbacks including large energy penalties and toxicity from the chemicals used [3,4]. Utilisation of CO_2 for the production of biomass offers a pathway to employ CO_2 emissions as a feedstock for the production of fuels, chemicals and other high value products which can ultimately replace those from fossil fuels [5]. The use of photosynthetic microorganisms, specifically microalgae, is believed to be one of the most promising techniques for biofixation of carbon [6].

Microalgae utilise CO_2 as their main carbon source during photosynthetic growth and do so at up to 100 times faster rates than terrestrial plants [7–10], making them an attractive option for CCUS. Although the premise of algal-CCUS has been around for some time, the efficiencies are very low and the costs very high, limiting commercial progression [11]. Furthermore, the previous focus of algal research on biofuel production means there is a wealth of information regarding lipid and biomass productivity under the supplementation of CO_2 with little information on the potential carbon reduction efficiencies and economics of carbon-capture rather than biomass production.

Previous studies have shown that commercially important microalgal genus such as *Chlorella* and *Scenedesmus* can withstand CO_2 concentrations as high as 80 – 100 % v/v, although the optimal conditions for growth were much lower, *ca.* 10 % CO_2 [12,13]. When it comes to the ability of algal cultures to remove CO_2 from the gas stream, the surrounding literature becomes unclear. There is an overwhelming focus on the organism's ability to produce lipids for biofuels or high value products (HVPs), such as proteins and carotenoids, rather than on how much CO_2 can potentially be captured by the cells. Few studies have looked directly at the ability of

algal cultures to remove CO_2 from a flow of gas. Li *et al.* [7] found that the CO_2 removal efficiency could be as high as 67 % with the green alga, *Scenedesmus obliquus* under optimal conditions, but that increases in CO_2 concentration (from 6 to 18 %) and the flow rate (0.05 to 0.5 vvm) had major detrimental effects on this efficiency. Chiu *et al.* [14] achieved a 58 % reduction in CO_2 concentration when sparging cultures of *Chlorella* sp. with 2 % CO_2 at 0.25 vvm. However, a similar pattern of reduced capture efficiency was seen by increasing the CO_2 concentration; CO_2 removal was as low as 16 % with an inflow of 15 % CO_2. The same group in 2011 found that a CO_2 removal efficiency as high as 95 % could be achieved when coke flue gas was intermittently sparged into cultures, although, as the duration of gas sparging into the culture increased this reduced, reaching as low as 13 % after 40 minutes of unceasing gas flow [15]. The use of intermittent sparging can be seen throughout the literature (sparging for less than a minute an hour) [15–20]. However, the stop-start nature and low potential CO_2 usage of these systems, especially where it is being used to control pH, would not be suited to applications such as CCUS where there could be substantial amounts of CO_2 requiring usage/storage at any given time.

Alongside these examples, there have been many instances where research has focused on the CO_2 removal rate of cultures in terms of the grams CO_2 removed per litre of culture per day (g L^{-1} d^{-1}), as seen in Table 1. However, with the large variety of cultivation systems, aeration rates and CO_2 concentrations being applied to cultures this gives a poor basis for comparison between the different microalgal species. In many of the cases seen in Table 1, the fixation rate of carbon, R_{CO2} (g_{CO2} L^{-1} d^{-1}), is calculated using the following equation:

$$R_{CO2} = P \times C_c \times \frac{M_{CO2}}{M_C} \quad (1)$$

Where P is the productivity of the culture in g L^{-1} d^{-1}, C_C is the carbon content of the dry biomass, assumed at ~ 50 %, and M_{CO2} and M_C are the molecular weights of CO_2 and carbon, respectively. This gives the assumption that for each kg of dry biomass produced, 1.88 kg of CO_2 is required [21].

Using these assumptions can give dramatically different results when compared to data produced from direct measurements of CO_2 in and out of the system. For example, Li *et al.* [7] directly measured the CO_2 removal efficiency of *Scenedesmus obliquus* and a mutant WUST4, gaining a CO_2 removal of between 40 and 60 % of the CO_2. If the productivity of the culture and experimental conditions described in their publication are used with the assumption described above, a removal of just ~ 1 % (0.17 g_{CO2} L^{-1} d^{-1}) is seen instead of that measured.

Genus and Species	CO_2 (%)	P (g L^{-1} d^{-1})	R_{CO2} (g L^{-1} d^{-1})	Method for determining CO_2 removal	Source
Botryococcus braunii	5	NA	0.497	Real time monitoring	[22]
Chlorella fusca	10*	0.08	0.255	Eq 1.	[16]
Chlorella kessleri	6*	0.087	0.163	Eq 1.	[23]
Chlorella minutissima	10	0.15	0.250	Eq 1.	[24]
***Chlorella* sp.**	5	0.271	0.498	$P_x \times 0.5 \times \frac{M_{CO2}}{M_C}$	[17]
***Chlorella* sp.**	25*	0.52	60%	Real time monitoring	[15]
Chlorella pyrenoidosa	10	0.144	0.260	Eq 1.	[25]
Chlorella vulgaris	5	NA	0.251	Real time monitoring	[22]
Chlorella vulgaris	5	0.11	0.15	Eq 1.	[26]
Dunaliella tertiolecta	3	0.17	0.313	Eq 1.	[27]
Dunaliella tertiolecta	5	NA	0.272	Real time monitoring	[22]
Scenedesmus obliquus	6	0.1	0.188	Eq 1.	[28]
Scenedesmus obliquus	10	0.155	0.288	Eq 1.	[25]
Scenedesmus obliquus	10	0.0653	40.2%	Real time monitoring	[7]
***Scenedesmus obliquus* WUST4**	20	0.0971	59.8%	Real time monitoring	[7]
***Spirulina* sp.**	6	0.2	0.376	Eq 1.	[28]
***Spirulina* sp.**	10*	0.04	0.120	Eq 1.	[16]
Spirulina platensis	5	NA	0.318	Real time monitoring	[22]

Table 1: *Comparison of CO_2 biofixation rates within the literature and the methods used to calculate these values. * Denotes that an intermittent gas flow was used in these experiments rather than a constant flow of CO_2 to the cultures. Where the method is denoted as Eq 1. This represents the equation described previously with C_C assumed as 50 % unless otherwise stated.*

Whilst research has focused on the CO_2 removal rate of different algal systems and the potentially reduced environmental burdens over mature CCS technologies [29–31], the economics of the process have yet to be purposefully looked at with the focus being carbon capture.

This research aims to highlight two key points, which do not appear readily within the literature:

1) Real-time monitoring of algal CO_2 uptake is key to accurate estimations of removal capacity when compared to the traditional use of following culture productivity. This is achieved by monitoring *Chlorella* sp. CO_2 removal capacity using an NDIR (Non Dispersive Infra-Red) sensor and then comparing results to those calculated from the growth rate throughout the experimental period.
2) The economics of algal-CCUS are not readily evaluated in terms of cost of capturing CO_2. Therefore, a techno-economic assessment (TEA) based on the experimental findings from this work, under different financial and operational scenarios is completed to highlight key areas for optimisation and development.

2. Experimental Materials and Methods:

2.1. Microalgal strain, media, and stock maintenance

The freshwater microalga, *Chlorella* sp., was used throughout this work. Stock cultures were maintained in closed 50 mL flasks (working volume 25 mL) under standard conditions of: continuous light (240 µmol m^{-2} s^{-1}), room temperature (20 ± 2 °C) and continuous stirring (120 rpm). All cultures were grown using a modified Bold's Basal medium [32], containing 3-times

nitrogen content and supplemented with vitamin B_{12}. The composition was the following (mg L^{-1}): 750 $NaNO_3$, 25 $CaCl_2.2H_2O$, 75 $MgSO_4.7H_2O$, 75 $K_2HPO_4.3H_2O$, 175 KH_2PO_4, 25 NaCl, 45 Na_2EDTA, 0.582 $FeCl_3.6H_2O$, 0.246 $MnCl_2.4H_2O$, 0.03 $ZnCl_2$, 0.012 $CoCl_2.6H_2O$, 0.024 $Na_2MoO_4.2H_2O$ and 0.001 ng Cyanocobalamin, made with deionised water. Initial media pH was 6.8 and the prepared media was autoclaved at 121 °C, 15 psi to sterilise before storing at room temperature for up to 2 weeks.

2.2. Growth with no supplementation of CO_2/air

Prior to testing growth under supplemental CO_2, the growth of *Chlorella* sp. without any additional aeration was measured as a baseline. Triplicate cultures and a media-only control were grown in 1 L flasks (500 mL working volume). All cultures were inoculated to an optical density (OD) of 0.1 at 695 nm (Spectronic 200E, ThermoFisher Scientific). The cultures were maintained in the same conditions as those mentioned in Section 2.1. While stirred at 120 rpm, a further hand shaking of the flask was performed before each sampling time to ensure a homogenous mixture. 5 mL samples were taken every 2-3 days to measure OD.

2.3. Growth under different CO_2 concentrations

To obtain the optimal CO_2 concentration for *Chlorella* sp. growth, cultures were grown with supplementation of either air, 5% CO_2 mixed in air, 10 % CO_2 mixed in air or 15 % CO_2 mixed in air. All elevated CO_2 concentrations were achieved through mixing pure CO_2 and compressed air via flow rate in a gas-mixing chamber before being administered to each culture. Triplicate cultures and a control containing only media were grown for each CO_2 concentration tested; Figure 1 shows the schematic for experimental set up. Cultures were 500 mL in volume with the hydration flasks containing approximately 200 mL of sterile distilled water each to minimise evaporative losses. Cultures were inoculated with washed cells from the previous experiment to an initial OD of 0.1 at 695 nm.

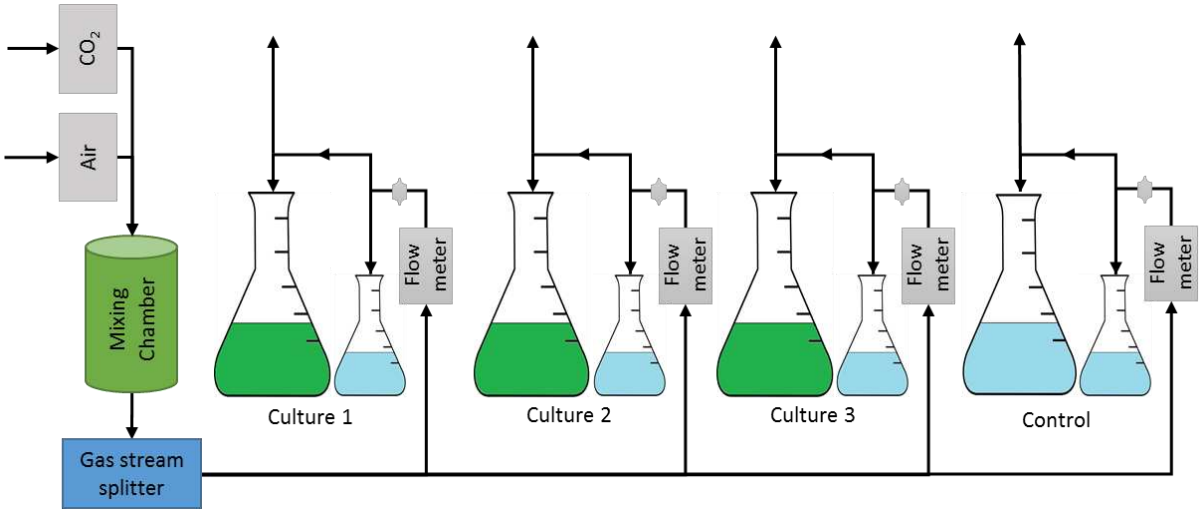


Figure 1: *Experimental set up for the growth of Chlorella sp. with different concentrations of CO_2 mixed in air.*

The flow of gas into each experimental flask was controlled by independent flow meters (FR2000 series, Key Instruments, USA) and gas-flow was maintained at 1 L min^{-1} (2 vvm) for each flask throughout the entire growth period. Prior to reaching the culture, gas flow was filtered through a 0.22 µm bacterial air-vent (Acro 37 TF, Pall, USA) and then hydrated by passing through a 250 mL flask containing deionised water. The gas flow then entered the experimental flask through a glass tube in the silicon stopper. The gas bubbling mixed the cultures sufficiently so that no additional agitation was required to keep cells in suspension.

Cultures were exposed to 24-hours light (200 µmole m^{-2} s^{-1}) and room temperature 20 ± 2 °C throughout the experiment. As previously mentioned, bubbling gases mixed the cultures, however, before sampling each culture was shaken by hand to produce a homogeneous mixture. 5 mL of culture was removed by sterile syringe through a sampling port every 2-3 days. Experiments were run for 14 days or until growth ceased.

2.4. Measuring *Chlorella* sp. mediated CO_2 removal in real-time

The optimal conditions for growth were then used in the same manner for the cultivation of one flask culture (Figure 2) connected to a NDIR sensor [33] (CO2meter.com, USA). The single flask experiment was conducted on three separate occasions. A fourth run with media only was conducted as a control. Before attaching the culture to the gas supply, the CO_2 and air mixture was measured with the sensor over a 0.5-2.5-hour interval to ensure the correct concentration of CO_2 was flowing. The volume flow rates used to maintain the 5 % CO_2 concentration were then maintained throughout the experiment to achieve a uniform gas flow to the culture. The sensor took readings of CO_2 concentration (%), temperature (°C) and humidity (%) every minute throughout the entire growth period (14 days). The experimental set up was the same as stated previously with two exceptions: 1) the flow rate entering the culture was reduced to 0.5 L min^{-1} (1 vvm) to further reduce evaporative losses seen during the preliminary experimentation and 2) the addition of a shaker unit underneath (115 rpm) to keep the culture in suspension as settling of the biomass during the latter half of the experiment became a major concern during the previous experiments.

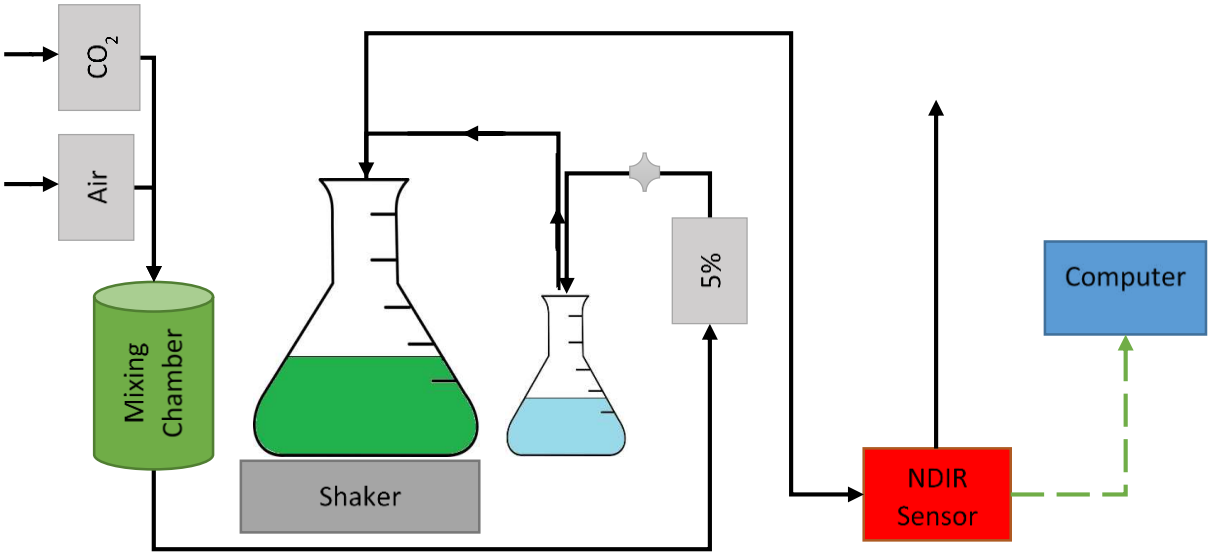


Figure 2: *Experimental set up for real-time monitoring of CO_2 uptake efficiency.*

2.5. Growth and productivity measurements

The growth of cultures was followed by measuring the OD at 695 nm, using sterile media as a blank [17]. A calibration curve of dry cell weight against OD was produced for each spectrophotometer used in these methods by drying washed condensed cells at 105 °C for 24 hours in pre-weighed Eppendorf tubes [34].

The culture productivity, $P\ (g\ L^{-1}\ d^{-1})$, was calculated as the difference in biomass concentration over time:

$$P = \frac{dX}{dt} \tag{2}$$

Where X is the biomass concentration (g L^{-1}) and t is time (days).

2.6. CO_2 removal efficiency measurements

The literature assumes that the CO_2 removal of an algal culture is based upon its productivity and the carbon content of the biomass produced, as described by Eq. 1. Therefore, Eq. 1 is used to give comparison of experimental results seen here with the current literature and for comparison against monitored CO_2 gas reduction efficiency observed in the secondary set of experiments. It is assumed here that *Chlorella* sp. has a 50 % by weight carbon content of its biomass, based on the general formula for microalgal biomass proposed by Chisti [21,35].

The experimental conditions and gas laws are used to calculate the CO_2 flow (g L^{1-} d^{-1}) entering the culture during the experiment. The estimated CO_2 removal efficiency (Estimated RE) of the culture is then calculated as:

$$Estimated\ RE\ (\%) = \frac{RCO_2}{CO2IN} \times 100 \tag{3}$$

Where CO2IN is the amount of CO_2 entering the culture, RCO_2 is the CO_2 removal rate calculated using Eq. 1 and both are presented in the same units.

Where the CO_2 concentration is measured in real time using the NDIR sensor, the monitored CO_2 removal efficiency (Monitored RE) is calculated as:

$$Monitored\ RE\ (\%) = \frac{CO2IN - CO2OUT}{CO2IN} \times 100 \quad (4)$$

2.7. Statistical analysis

All experiments were conducted in triplicate. Analysis of variance (ANOVA) was carried out to study the difference between conditions, with a significant difference recognised where $p < 0.05$.

3. Techno-economic analysis

The experimental data for biomass production and CO_2 removal efficiency were then used as inputs for a TEA based on a theoretical facility of modular photobioreactor (PBR) units. The aim of which is to gain estimates of the current CO_2 capture costs to allow for comparison with mature technologies, as well as to highlight areas for optimisation and further research.

The methodology used is based on that employed within the current literature for algal-biorefineries and biofuel production and adjusted for the use as an option for CCUS.

3.1. Facility description and scope definition

The facility size is set to 0.5-ha and consists of an array of modular PBR units along with a laboratory and office space as seen in Tredici *et al.* [36]. The TEA is based on a modular PBR unit of 300 L; the Phycoflow® (Varicon Aqua, UK), Figure 3, as capital and operational information for these units is readily available to the authors.

Figure 3: *The modular PBR, Phycoflow® from VariconAqua. The unit has a working volume of 300 L, composed of a 100 L plastic tank and a serpentine borosilicate bioreactor. The PBR is encased in a Sunlite multiwall protective structure with 83 % light penetration.* [37]

It is assumed that the CO_2 supply for algal growth will come from a source point (power station *etc.*) and that either the algal facility will be onsite and therefore transportation is not included or that the provider will supply the transportation needed to the facility and therefore is assumed outside the scope of the analysis. It is also assumed that the biomass is harvested and sold on as a feedstock for fuels, HVPs or back to the CO_2 source point for combined heat and power (CHP) generation and therefore this too is excluded from the scope, which is described further in Figure 4.

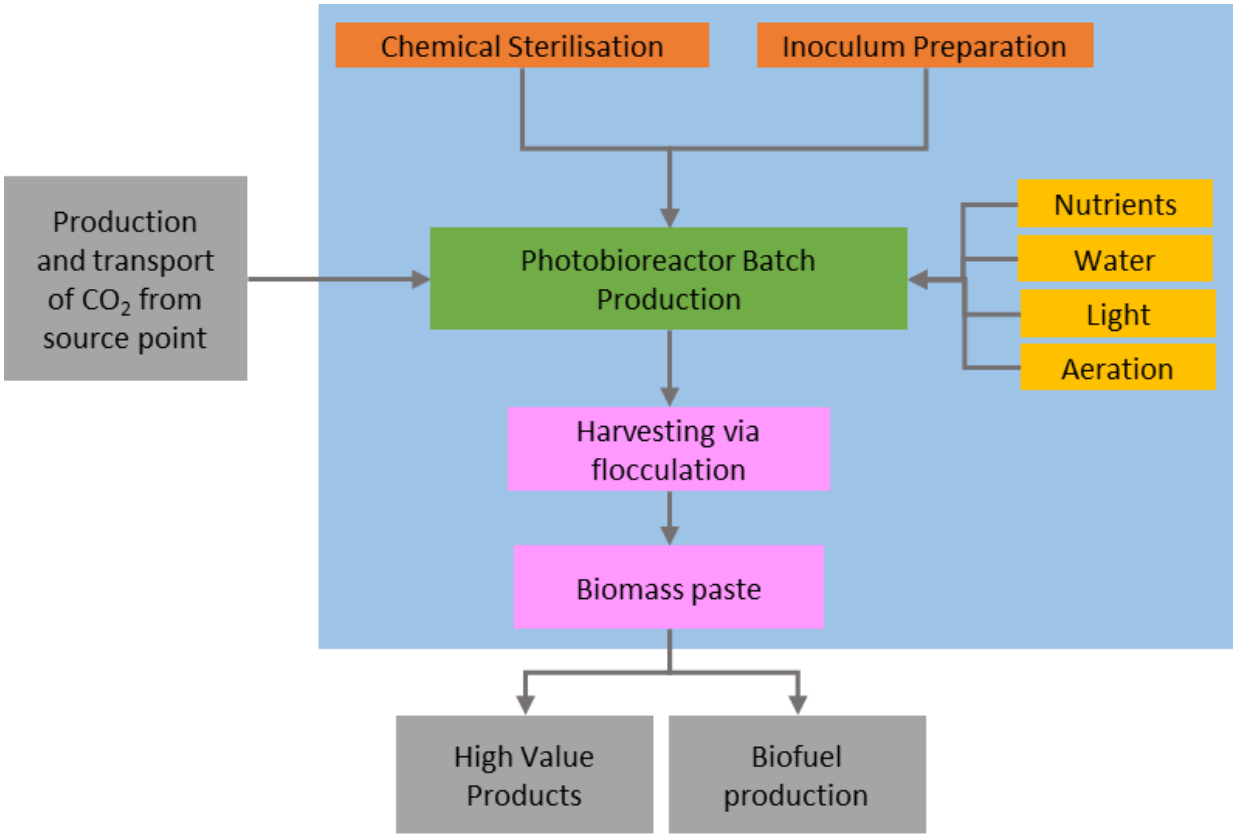


Figure 4: *Scope of the TEA. All processes and elements inside the shaded area are covered within the analysis. It is assumed that either the facility is located adjacent to the CO_2 source point or that gas provider will cover the cost of transportation to the facility.*

3.2. CapEx and OpEx

The capital and operational expenditure (CapEx and OpEx, respectively) were calculated over the chosen project lifetime of 20 years [29,38–44] with a reference year of 2018, in GBP (£). Financial inputs such as contingency, depreciation and discount rate are all taken from the literature, and the electricity and water prices are assumed to be the UK average cost and at a standard rate for Yorkshire, UK (Table 2). The price of land was taken from the average price for industrial land in the UK [45]. A breakdown of capital items can be found in Supplementary Table 1.

The total CapEx figure is a combination of the total direct CapEx (TDC) (major equipment, land, and buildings) and total indirect CapEx (TIC) (contingency planning and installation):

$$Total\ CapEx = TDC + TIC \quad (5)$$

263

264 For net cash flow (NCF) calculations, depreciation of physical assets such as equipment is
265 applied [36], in a linear manner over the items lifetime with a salvage value of zero:

$$Annual\ Depreciation\ \% = \frac{ItemCost}{ItemLifespan} \quad (6)$$

266

267 The total annual OpEx was calculated to include three major items: direct cultivation OpEx
268 (DCO), annual labour OpEx and indirect:

$$Total\ OpEx = DCO + Labour + Indirect\ OpEx \quad (7)$$

269

270 The DCO is calculated using the experimental data that has been scaled appropriately. This
271 element includes all the nutritional, water, heat, and energy inputs required for the cultivation
272 , harvesting of the biomass and cleaning/sterilisation of the PBR for the next batch cultivation.
273 The use of lower grade or wholesale chemicals was considered in this calculation rather than
274 the use of laboratory-grade chemicals used within the experimentation. It is assumed this
275 change does not affect the algae's growth or CO_2 fixation rate.

276 Labour costs are included in this TEA. There is no consistency with the literature for labour
277 cost inclusion and the assumptions made differ dramatically between each publication
278 [38,41,42,46–49] . Here, the UK average salary for scientific technicians and laboratory
279 supervisors are used with a 60 % overhead for additional services.

280 The indirect OpEx is included to cover the cost of maintenance and insurance which will be
281 charged annually. Both the maintenance and insurance costs are assumed to be percentages
282 (5 and 10 %, respectively) of the combined total of DCO for all units and annual labour OpEx:

$$Indirect\ OpEx = 15\% \times (DCO + Labour) \quad (8)$$

283

284 The revenues from the process include both sale of the wet biomass produced and the
285 potential of carbon credits or the avoidance of carbon taxation by the emissions reduction. A
286 minimum selling price (MSP) for the biomass paste produced and a Cost of Capture of CO_2
287 (CoC) were calculated using the financial information over the entire project lifetime
288 annualised:

$$MSP(£kg^{-1}) = \frac{(CapEx + OpEx - Revenue)}{Biomass\ Yield} \quad (9)$$

289

290 For the calculation of Cost of Capture (CoC) for the CO_2, the wet biomass is assumed to be
291 sold at £0.34 kg^{-1} [50]**.** The CoC is then calculated as:

$$CoC\ (£\ tonne_{CO2}^{-1}) = \frac{(CapEx + OpEx - Revenue)}{CO_2 Captured} \quad (10)$$

292

293 Where CO_2Captured is the amount of CO_2 taken out of the gas stream over all the PBR units
294 annually in tonnes. Both values allow comparison between this work and the literature.

Item	Value	Description	Reference
Project lifetime	20 years	Average within the literature	Davis et al., 2011; Doshi et al., 2017; Gallagher, 2011; Ventura et al., 2013; Wiesberg et al., 2017; Xin et al., 2016; Zamalloa et al., 2011; Zhang et al., 2017
Reference year	2018	Year to which all prices are originally set against	
Construction period	1.5 years	No operation for the first 1.5 years of the project lifetime due to construction/testing of facility	Davis et al., 2013; de Queiroz Fernandes Araújo et al., 2015
Electricity price	£0.1344 kWh^{-1}	UK average cost	Energy Saving Trust, 2018
Water price	£1.348 m^{-3}	Standard tariff	Yorkshire Water, 2018

Sewage price	£1.59 m^{-3}	Standard tariff	Yorkshire Water, 2018
Maintenance cost	5%	Of TDC	Tredici et al., 2016
Contingency cost	15%	Of TDC	Nagarajan et al., 2013; Ou et al., 2015
Labour overheads	60%	Of salary costs	Brownbridge et al., 2014
Depreciation	Straight line	Items depreciate linearly over their product lifetime with zero salvage value	Amer et al., 2011; Doshi et al., 2017; Tredici et al., 2016

Table 2: *Financial assumptions and prices used within the TEA model.*

3.3. Scenario analysis

Once the baseline values for CoC and the overall cost breakdown were calculated, six different operational and financial scenarios were input to the TEA, descriptions of all the scenarios can be found in Table 3. The results from each scenario were analysed against the baseline and each other based on cost breakdown and CoC.

This scenario based analysis is seen throughout the literature as a way of showing how different financial, political and technological situations can either increase or decrease the economic viability of algal based remediation and biofuel production [9,31,60–63,38,39,41,50,56–59]. The scenarios selected in this work posit reasonable improvements in algal productivity, operational management or investment requirements, all of which can be seen as near-term or optimistic goals, similar to methods used in Hoffman *et al.* [58].

Scenario 1 assumes that the laboratory data does not scale to commercial and therefore offers a 'worst-case scenario' where the biomass production is reduced by 50 %. Scenario 2 is the baseline case used in the initial TEA set up. Scenario 3 offers reduction in CapEx of 25 %, assuming that a wholesale/trade discount can be applied to the major equipment purchases. Scenario 4 assumes reductions in operational costs, for example, there is no charge to water or nutrients due to using a waste-water stream and that power is provided cheaply on site.

Scenario 5 combines both assumptions from 3 and 4 to give an 'optimal' cost reduction. Scenario 6 assumes that a CO_2 credit for the capture process is applied at £50 $tonne_{CO2}^{-1}$. This is based on the USA's 45Q Carbon tax of $50 $tonne_{CO2}^{-1}$ and the EU emissions trading scheme value of £16 $tonne_{CO2}^{-1}$, which would likely rise in the coming years to reach emissions targets. Scenario 7 assumes there has been strain optimisation and the algae have an increased CO_2 capture efficiency.

Scenario	Description
1	Sub optimal, there are issues with scaling up the experimental results and therefore the biomass productivity and CO_2 capture efficiency are halved compared to baseline.
2	Baseline, the original information used to create the TEA.
3	Reduction in CapEx, due to the high volume of equipment being purchased a bulk-order discount of 25 % is applied to all major equipment purchasing, including the PBR units.
4	Reduction in OpEx, assuming there is no change in the biomass productivity or CO_2 capture efficiency, the cost of operational expenditures such as lighting, media nutrients and heating are no longer required and omitted.
5	Combination of both CapEx and OpEx reduction, both scenarios 3 and 4 combined.
6	CO_2 credits, again assuming there is no change to the biomass productivity or CO_2 capture efficiency, there is an introduction of a 'carbon credit' where a revenue of £50/tonne captured CO_2 is applied.
7	Improvements in efficiency, without the biomass productivity changing, the efficiency of the capture process is doubled.

Table 3: *Financial and Operational scenarios used to evaluate how changing parameters in a realistic manner affects the cost breakdown and CoC for this algal-CCUS option. Further information on changes can be found in Supplementary Table 2.*

3.4. Sensitivity analysis

A single-parameter sensitivity analysis was then conducted for each scenario to see the effect increasing capture efficiency would have on the CoC value. Each scenario was run with CO_2 removal efficiencies increasing from 0 – 100 %. The test was done using the Microsoft Excel Data Table function where the efficiency was the only parameter affected.

A two-parameter sensitivity analysis using the same methods was then conducted for the baseline scenario against changing CO_2 removal efficiency (as above) and increasing CO_2 credit values from 0 – 300 £ $tonne_{CO2}^{-1}$.

4. Results:

4.1. Growth of Chlorella sp. under different CO_2 concentrations

The growth of *Chlorella* sp. under the five different CO_2 conditions tested is shown in Figure 5. The slowest growth was seen for the control cultures where no aeration was applied. Here the biomass does not exceed 0.2 g L^{-1} at the end of the growth period. The air (0.04% CO_2) aerated cultures grew significantly faster than the controls but slower than all the remaining conditions, reaching a final biomass concentration of 1.3 g L^{-1} at 2 weeks.

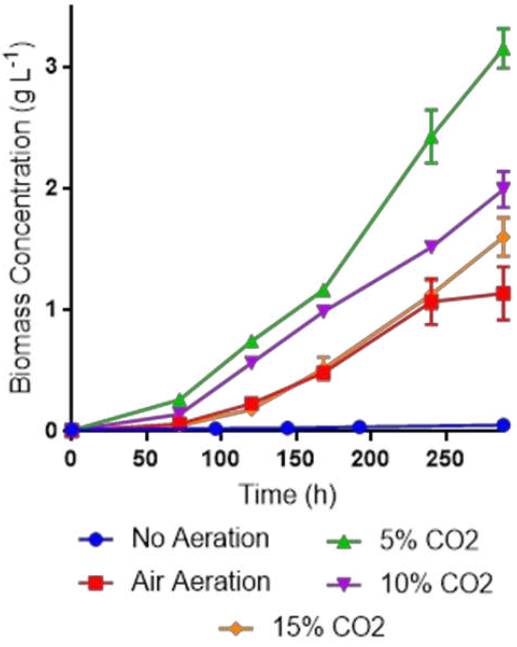


Figure 5: *Chlorella sp. growth under different CO_2 environments ranging from no aeration to 15 % CO_2 mixed in air. Each experiment consists of 3 replicates and a media-only blank to check for contamination.*

345 The highest growth was seen when 5 % CO_2 was added to the cultures. From the graph, it
346 can be seen the 5 % CO_2 cultures were still in their linear growth phase as the experiment
347 ended. Increasing the CO_2 concentration further to 10 and 15 % CO_2 did not further improve
348 the growth of *Chlorella* sp., actually decreasing the final biomass concentration at the end of
349 the experiment. The cultures grown with 10 % and 15 % CO_2 reached a maximum biomass
350 concentration of 1.99 and 1.60 g L^{-1}, respectively. When the average culture productivity was
351 compared against the non-aerated control cultures, all aerated cultures grew significantly
352 faster ($P<0.05$).

353 Although giving a lower final total biomass yield than 5 % CO_2, conditions of both 10 and 15
354 % CO_2 gave a higher biomass concentration than aeration with air alone, suggesting the
355 cultures are carbon limited when supplied with air only. This is further supported, as the only
356 independent variable within the experiments was the aeration carbon dioxide concentration.
357 Therefore, it can be assumed that carbon is the limiting factor when no air or air alone is
358 supplied and that around 5% CO_2 is optimal for this species of microalgae.

359

360 **4.2.** *Chlorella* sp. growth and real-time monitoring of CO_2 exiting the culture

361 The previous experiment showed that, of the concentrations tested, 5 % CO_2 gave the highest
362 growth rate for *Chlorella* sp. and was therefore the CO_2 concentration used in the next phase
363 of experiments. Figure 6 shows the biomass growth and CO_2 removal efficiency of the cultures
364 over the 14-day experimental period. The CO_2 removal efficiency is based on the difference in
365 concentration entering and exiting the system as measured by the NDIR sensor (Eq. 4). The
366 growth of the cultures shows a similar pattern to the 5 % CO_2 cultures for the previous

experiments, with the cultures still being in the linear growth phase at the end of the experiment. After two weeks, the final average biomass for the cultures was 2.11 g L^{-1}.

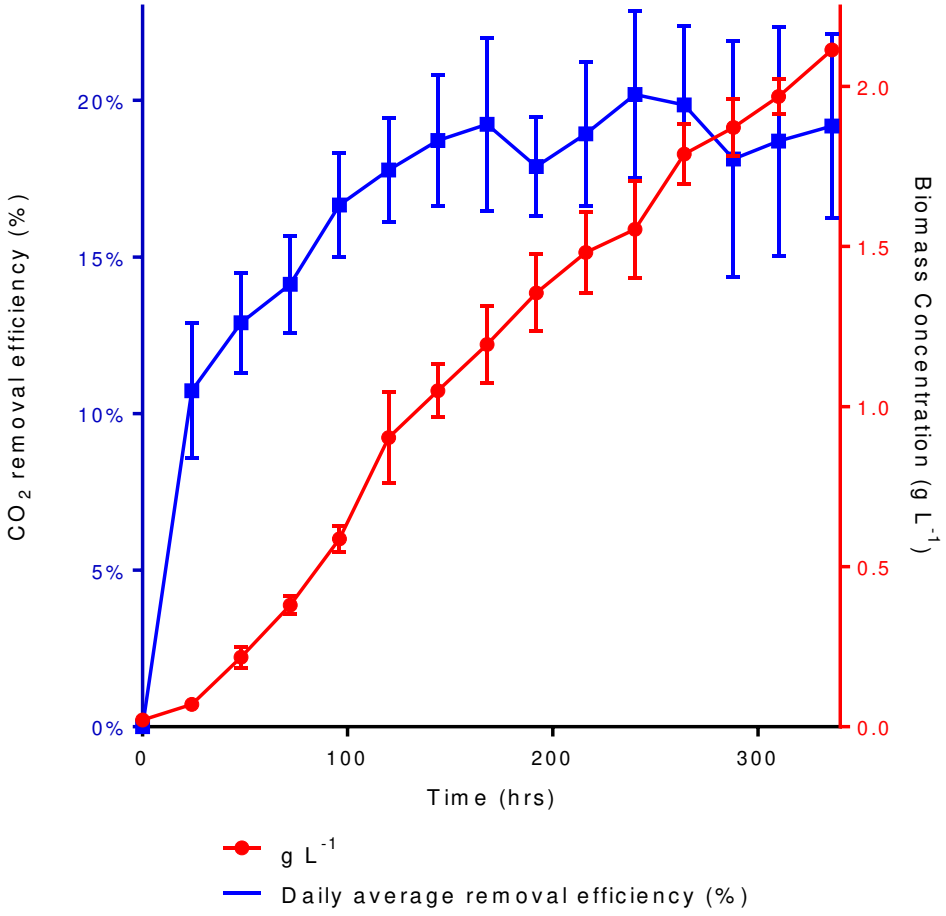


Figure 6: *The growth of Chlorella sp. grown under continuous aeration with 5 % CO_2 (red) and the CO_2 removal efficiency of the culture measured by real-time CO_2 measurements of the off-gas (blue). The results are averages from three replicate cultures.*

The CO_2 removal efficiency of the culture changes over the cultivation time. At the beginning of the experiment where the biomass concentration is very low the CO_2 removal efficiency is very low, reaching only 10 % after 24 hours of growth. As the biomass concentration increases

the CO_2 removal efficiency does as well, until a plateau at ~ 17.5 % efficiency is reached in the latter half of the experiment, during the linear growth phase.

4.3. CO_2 removal comparison

To allow comparison with information available within the literature (Table 1), the average culture productivities for each experiment were used to calculate the R_{CO2} as described by Eq. 1. The average productivity of the triplicate cultures for each condition was used along with the assumption that *Chlorella* sp. has a carbon content of 50 %, based on the approximate molecular formula for microalgal biomass proposed by Chisti [21]. Figure 7 shows the R_{CO2} values for each experimental condition. The highest R_{CO2} was seen for 5 % CO_2 during the first experiment, where the final biomass concentration was also the highest. This R_{CO2} value of 0.31 g_{CO2} L^{-1} d^{-1} is higher than the average of those presented in Table 1 (0.279 g_{CO2} L^{-1} d^{-1}).

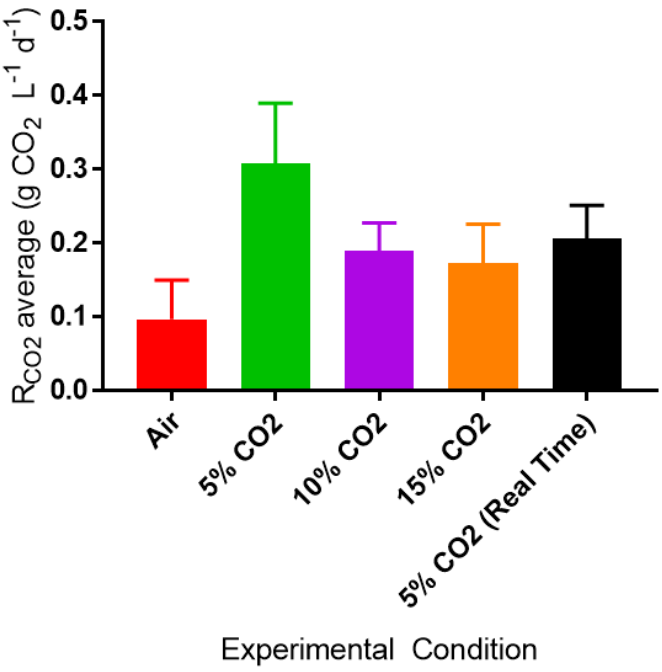


Figure 7: *The average R_{CO2} for Chlorella sp. grown under different CO_2 concentrations over a two-week period. The R_{CO2} value is calculated using Eq. 1 from the literature to allow for*

393 *comparison with the surrounding literature. The 5 % CO_2 Real Time (final column) denotes*
394 *the second experiment where CO_2 removal was measured in real-time also.*

395

396 Although the R_{CO2} values mentioned above are higher than seen elsewhere in the literature,
397 these values do not tell us how much CO_2 was removed from the inflowing gas stream. For
398 aeration with air, 5 %, 10 % and 15 % CO_2 in air, the maximum Estimated RE (when using
399 Eq. 1) are 7.58 %, 0.14 %, 0.04 % and 0.03 %, respectively. Although the biomass production
400 has been visibly improved by increasing CO_2 concentration, the CO_2 availability now
401 supersedes the difference in growth meaning a lower percentage of that available is actually
402 used, when comparing the air and 5 % CO_2 experiments.

403 Table 4 shows the comparison of these results to the real-time monitored CO_2 removal
404 efficiency from the second experiment. As can be seen, there is a large difference in the CO_2
405 removal efficiency, with the real-time monitoring showing a much larger CO_2 removal
406 efficiency than that predicted by Eq. 1. One reason for this may be the fact that Eq. 1 assumes
407 that the carbon content of the biomass remains as a fixed value. The carbon content of the
408 biomass may fluctuate over time leading to a higher or lower CO_2 consumption at any given
409 time point [64], missed by the assumptions made in the equation. Another reason would be
410 the production of excreted products [65] or storage of dissolved inorganic carbon in vacuoles
411 within the cells [66]. Neither of these would be seen in the measurement of biomass
412 productivity, which the equation relies on. The media only control for this experiment (data not
413 shown) highlights that there is an initial capture of CO_2 by the media in the first hour of bubbling
414 but after this the carbon balance is maintained and the media does not take up any additional
415 CO_2 from the gas inlet over the entire 14-day period.

416

Experimental conditions	Average Productivity ($g\ L^{-1}\ d^{-1}$)	RCO_2 based on Eq. 1 ($g\ CO_2\ L^{-1}\ d^{-1}$)	RCO_2 based on NDIR sensor ($g\ CO_2\ L^{-1}\ d^{-1}$)	Estimated RE
5 % CO_2, 1 vvm, real time monitoring	0.11	0.21	11.53	17.27% Monitored RE 0.08% Estimated RE
Air, 2 vvm	0.05	0.10		7.58%
5 % CO_2, 2 vvm	0.17	0.31		0.14%
10 % CO_2, 2 vvm	0.10	0.19		0.04%
15 % CO_2, 2 vvm	0.09	0.17		0.03%

Table 4*: Productivities, RCO_2 and CO_2 removal efficiency for Chlorella sp. grown under different CO_2 conditions.*

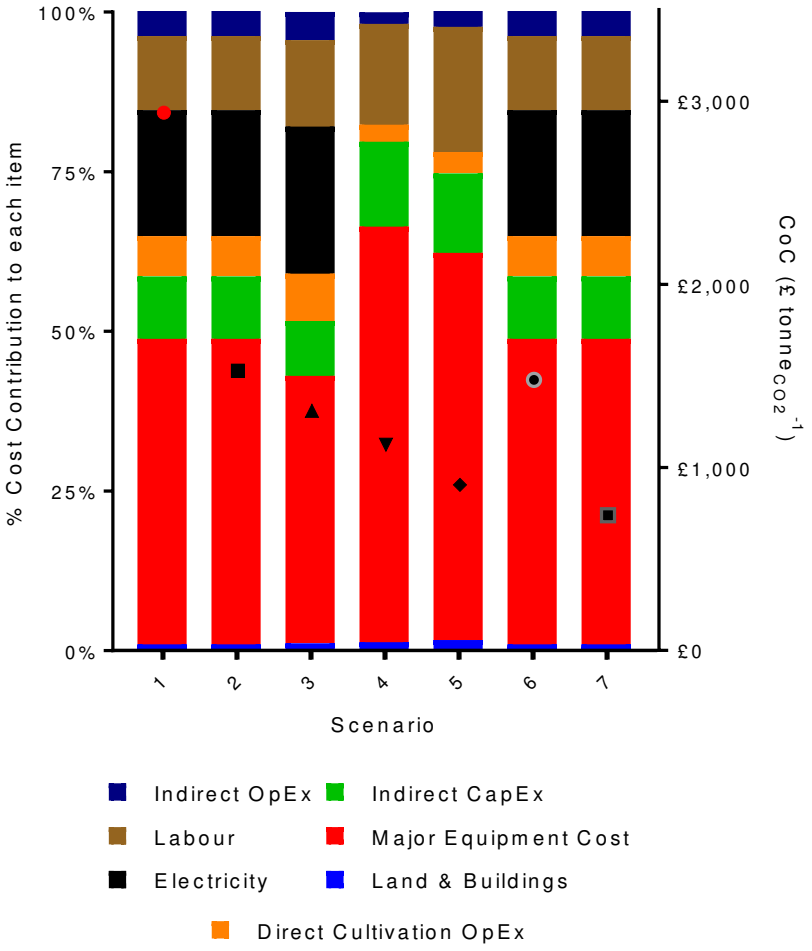
The difference between the R_{CO2} and sensor CO_2 removal values is statistically significant ($P<0.0001$) in all time periods. Therefore, it can be deduced that Eq. 1 under-estimates the CO_2 capture potential of microalgal cultures. This is further corroborated by the information shown in Table 1. Where real-time monitoring has been used [7,67] there are significantly higher CO_2 removal capacities by the cultures even though the species and experimental conditions are similar to those presented in the rest of the literature.

4.4. Techno-economic assessment

The experimental results for *Chlorella* sp. grown with 5 % CO_2 were extrapolated for use in a theoretical facility TEA. A financial and operational baseline scenario was produced based on current information and pilot scale/new technology contingency planning. A cost breakdown for each major section was produced as well as the overall cost of CO_2 capture (CoC, £ $tonne_{CO2}^{-1}$), £1,527.89 $tonne_{CO2}^{-1}$. A variety of different financial and operational scenarios

were then run through the model to help determine which parameters would produce the highest cost-reduction and are therefore, where optimisation should be focused. Figure 8 shows the cost breakdown and CoC values for each of the seven scenarios studied. From the figure, it can be seen that Scenarios 3-7 all lead to a reduction in CoC values compared to the baseline (Scenario 2) and Scenario 7 gives the largest decrease in value of 52 % to £769.05 $tonne_{CO2}^{-1}$. The next lowest CoC value was obtained for Scenario 5 where both CapEx and OpEx are reduced significantly. A combination of species optimisation for increased efficiency and reduced capital and operational expenditure are therefore key areas for cost reduction.

The cost-breakdown for each scenario shows that major equipment cost (PBR, pumps, harvesting tanks, heating, lighting *etc.*) is the largest expense in all cases, contributing ~ 50 % of the cost in all scenarios. Labour is the next most expensive parameter for all cases, followed by electricity demand and indirect CapEx. The indirect CapEx includes 15 % of the direct CapEx for contingency, a large value used for new and developing technologies, with advancements in the field this is likely to drop alongside the direct CapEx costs. Land and buildings contribute the least to cost and this is partially due to the lack of depreciation applied to these items. It is assumed in the model that land does not lose any value over the project lifetime and can be sold at the end of the project lifetime for the purchase value. This being said, industrial land value in the UK has increased over 30 % between 2014 and 2017 [68] and therefore it can be assumed that if land appreciation is taken into account CoC can be lowered further.



454

455 ***Figure 8:*** *Results for the techno-economic analysis carried out under different financial and*
456 *operational scenarios. The bars represent the cost breakdown for each scenario (left y-axis).*
457 *The black points in each bar represent where the cost of capture (CoC) lies for each scenario*
458 *(right y-axis).*

459 As the scenario with strain development and optimisation for improved CO_2 capture efficiency
460 gave the most dramatic cost reductions, a sensitivity analysis of all scenarios to this parameter
461 was conducted. Each scenario was tested with different CO_2 capture efficiencies from 0 – 100
462 %, shown in Figure 9. In each scenario, the CoC value drops with increasing capture efficiency
463 with a minimum value of £ 176.58 tonne$_{CO2}^{-1}$ obtained for Scenario 5. The graph also highlights
464 where the baseline TEA and experimental data currently sits. This single-parameter analysis
465 only considers the improvement of CO_2 uptake by the cultures and not the increased biomass

production which would accompany it. Sales of the additional biomass for HVPs, feed or fertiliser with higher sale prices than energy and fuel biomass would further reduce the overall CoC value, making the algal CCUS more competitive with mature CCS technologies such as amine scrubbing (€ 55-77 $tonne_{CO2}^{-1}$ [69,70]).

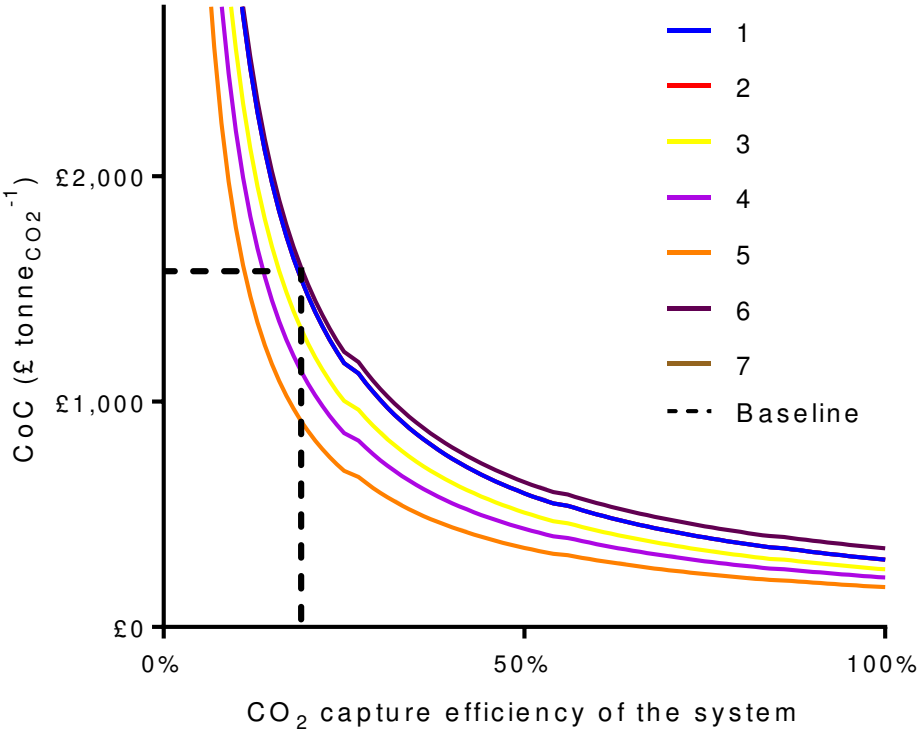


Figure 9: *Sensitivity analysis of the CoC for each scenario when the CO_2 removal efficiency is increased towards 100 %. The black intersecting line shows the current experimental data regarding efficiency and estimated CoC value.*

Beal *et al.* [31] stated in their algae bioenergy CCS (ABECCS) TEA that as the system is specifically designed to take up CO_2 it would be "unrealistic to consider scenarios without a significant carbon credit". Therefore, as this process is designed for the same purpose, a further analysis based on both the CO_2 removal efficiency and an increasing CO_2 credit was conducted. The baseline scenario was used for this and the CO_2 credit was varied from £0 –

£300 $tonne_{CO2}^{-1}$ and the efficiency from baseline (17%) to 100%, shown in Figure 10. The heat map shows how the CoC value changes with the two parameters. To achieve an overall negative CoC the efficiency needs to be above 98% and the credit around £300 $tonne_{CO2}^{-1}$.

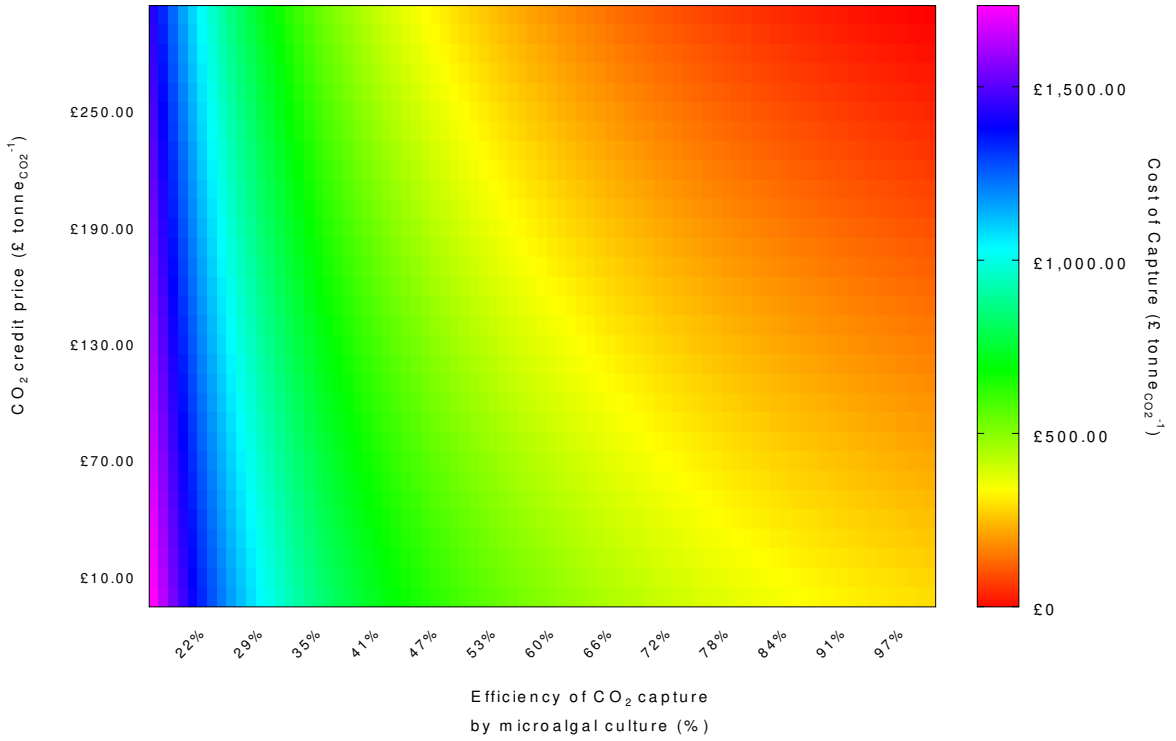


Figure 10: *Two-parameter sensitivity of the CoC for the baseline scenario against increasing CO_2 credit price and variable efficiency in CO_2 capture.*

5. Discussion:

5.1. Optimal CO_2 concentrations for *Chlorella* sp. growth

The *Chlorella* sp. used within this work grew the best under conditions of 5 % CO_2, similar to the concentrations found in closed-cycle gas turbine (CCGT) power plants [71,72], the second largest power generation type in the UK after petroleum oil [73]. These results show similar trends in microalgal growth under elevated CO_2 concentrations to that seen within the literature. Yang *et al.* 2020 *Desmodesmus* sp. and *Scenedesmus* sp. grew the best at 5 – 10

% CO_2 but that increasing the concentration to 15 % CO_2 caused a negative effect on the cultures [74]. The likely reasoning for inhibited growth at higher concentrations is not due to carbon limitation but in fact due to the dissolved CO_2 within the media causing the pH to drop below the optimal for this species.

On the other hand, Chiu *et al.* (2009) found that *Nannochloropsis oculata* was extremely sensitive to elevated CO_2 concentrations above 2 %. At all conditions above 2 % the algae did not grow, while 2 % vastly improved the growth rate compared to aeration with only air [75]. This highlights that CO_2 sensitivity is extremely species specific and will be a key consideration when looking to move into industrial applications.

This work was conducted using a 24-hour light cycle, as many other publications have also done [76,77]. The change in CO_2 capture efficiency when different light cycles, including pulsed, flashing and traditional 12-hour light: dark, are used should be considered in future analyses and experimental work. The difference between the CO_2 uptake of cultures and the CO_2 emissions from providing artificial lighting during night hours will be an important ratio for the consideration of algae as a CCUS option.

5.2. Measuring CO_2 capture in real time versus theoretical estimations

The experimental results expressed in Table 4 for CO_2 removal based on real-time monitoring and based on the assumption used vastly within the literature, compared to the figures presented in Table 1, highlights the key issues with current research techniques. The microalgal cultures can capture a larger proportion of the carbon presented to them than is estimated in Equation 1. Jacob-Lopez *et al.* 2008, Neves *et al.* 2018, and Gonzales Lopez *et al.* 2009 all give examples of exopolysaccharides and additional metabolites which are produced by microalgae which will contribute to the carbon capture but not be evaluated when only the cell density/productivity is assumed to be capable [78–80]. Continued development with real-time monitoring of CO_2 in flow and out flow from bioreactors will allow for a much better understanding of how the cells utilise the resource when it is not a limiting factor. Much

research has been published on how cells adapt to carbon limited environments with the carbon concentrating mechanism but little has considered how to adapt cells to higher concentrations and allow for a better capture rate.

Alongside the requirement for more direct measurements of CO_2 uptake, optimisation of the PBR for CO_2 capture should be addressed. The low CO_2 capture (10-20 %) seen throughout the literature and this work [28,74] highlights that this is a key area for improvement. Yang *et al.* 2020 showed that the use of sequential reactors can improve the CO_2 capture efficiency of *Chlorella pyrenoidosa* cultures from 10 % up to 90 % at a CO_2 concentration of 10 % CO_2 [81].

Moving forward, efforts should be made towards using real and simulated flue gases from various applications such as Kao *et al.* and Doucha *et al.*, to highlight how other components will affect the growth of the microalgae [82,83].

5.3. Techno-economics

As this is an initial assessment, based on laboratory data and a theoretical 'first of its kind' facility there are limitations to the results gained. All the scenarios tested are realistic but changes as singular as those shown are unlikely. For example, improvement of the capture efficiency in Scenario 7 does not consider that, as a result, more biomass will be produced. This could be sold on to increase revenue, but also requires more nutrients feeding into the PBRs, which will incur additional charges. It is also important to note that while the analysis highlighted that the introduction of a government policy for carbon credits could aid the feasibility of this process, these credits will likely not exist for the entire project lifetime (20 years) and their value may fluctuate overtime. Improvements in the scenario management and analyses based on government policy for previous, similar, technologies (first and second-generation biofuel production) would help further improve the accuracy of this assessment.

6. Conclusions:

The *Chlorella* sp. used within this work grew the best under conditions of 5 % CO_2 with a 17 % CO_2 capture efficiency over 14 days of growth. It is important to note that the process used in this work was not optimised and therefore improvements could readily be made including the use of sequential reactors and reduced flow rates [7,28,74]. Adaptive evolution of the species, increasing inoculation concentration of the microalgae and optimised nutrient and light feeding could also further increase the capture efficiency and are where future work will be focused.

The TEA performed in this work highlights that improvements in the efficiency of capture by the microalgae and cost reduction in both the capital and operational aspects of the process would greatly benefit the economics of the proposed facility. The conclusions drawn from it can be used to direct further research to focus in these key areas. Combining the carbon capture process with other waste treatment technologies (e.g. domestic or industrial wastewater remediation) should also be considered for improving the feasibility of the process.

While the premise of using microalgae for CO_2 capture from waste streams has been around for decades now there is little information on the economics of the process. The analysis performed here highlight that strain development, reduction in capital expenditure and government policy advocating for emissions reductions technologies will all be key for microalgal-CCUS at large scale. The production of a stable market for microalgal products will help drive down the cost of production [31] and allow microalgal CO_2 biofixation to become competitive [63] with other CCS technologies.

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846