Carbon Dioxide Enrichment in a Decarbonised Future

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Referaat

CO_2-dosering is belangrijk voor de glastuinbouw, om productiviteit te verhogen en daarmee ook de efficiëntie waarmee andere grondstoffen worden gebruikt. In een toekomst zonder fossiele brandstoffen en met elektrische kassen, waar zal de sector zijn CO_2 vandaan halen? In dit verslag bekijken we deze vraag vanuit vier aspecten: (1) hoeveelheid, (2) concentratie eisen en technieken, (3) schadelijke stoffen en hoe deze verwijderd kunnen worden, en (4) distributie. Met literatuur en berekeningen bekijken we de potentie en uitdagingen van technologieën, zowel gangbaar als experimenteel, in allevier aspecten van CO_2 voorziening.

Abstract

CO_2 enrichment is an important way for greenhouse horticulture to increase its yield and efficiency for other resources. In a future where fossil fuels are to be phased out in favour of electrified greenhouses, where will the the industry get its CO_2 from? This report aims to examine this question through four perspectives: (1) quantity, (2) concentration requirements and how to concentrate CO_2, (3) purity and how to remove contaminants, and (4) distribution options. Based on literature and calculations, we assess the potential and challenges related to technologies, both mainstream and novel, in all four of these aspects of CO_2 provision.

Reportinfo

Disclaimer

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Summary

To reduce CO_2 emissions, great efforts are being made to phase out fossil fuels in favour of carbon-neutral alternatives. For greenhouse horticulture, which currently uses natural gas as an energy source, electrified and 'fossil free' greenhouses are also expected to be the way forward. Currently, extra CO_2 is introduced into greenhouses to increase yields considerably. In a decarbonised future, where will greenhouses get this supplemental CO_2 from?

The objective of this project was to investigate fossil-free CO_2 sources for the Dutch greenhouse horticulture sector and how they can be safely and effectively delivered to the crop. We look at both mainstream and experimental alternatives using literature and simple calculations.

This report looks at CO_2 supply from four perspectives:
1. **Quantity**: how much CO_2 does the sector need and how much is available?
2. Quality in terms of **concentration**: what is required and how can the desired concentration be achieved?
3. Quality in terms of **purity**: which contaminants are likely, harmful, and how can they be removed?
4. **Distribution** to the greenhouse.

Ignoring direct air capture (DAC), we expect enough non-fossil CO_2 from other sources to be available to meet the sector's projected demand of 1.8-3.0 Mt in 2030. This has the advantage of already being concentrated compared to ambient concentrations (see Figure 3). By 2030, biogas upgrading and ethanol fermentation alone are projected to produce enough CO_2 for the entire sector.

For concentration, just 4% CO_2 is enough in theory, with typical ventilation rates – but such low concentrations require larger volumes making them impractical. Even on-site low-concentration DAC has been found to be less practical than concentrating to 100%. Biogas upgrading and ethanol fermentation produce the most concentrated CO_2 flow. This is an advantage, as the cost of concentrating CO_2 decreases nonlinearly with an increasing starting concentration. Various technologies are discussed for the other sources, including electroreduction to synthetic fuels. Besides being energy-intensive, these have the disadvantage of coupling CO_2 release to energy release, a disadvantage currently seen in today's natural gas boilers/CHPs. A combination of membrane separation and cryogenic distillation is currently the most feasible concentration method.

We list contaminants currently known to be harmful, and for the most common, their threshold concentrations and exposure times before harm to the crop can be expected. CO_2 from composting and biomethane combustion is likely to contain harmful contaminants, however no more than from burning natural gas, for which growers have the capacity to remove contaminants. Biomass combustion and composting produce the most contaminants. This report outlines the advantages and disadvantages of various purification technologies for NO_x, SO_2 and ethylene, recommending a combination of wet scrubbing and adsorption.

A number of mainstream solutions are discussed, as well as the advantages and methods for CO_2 buffering. Lastly, three potential solutions are covered quantitatively: local DAC, biogas upgrading with seasonal buffering, and local CO_2 storage based on lime.

1   Introduction

In a decarbonised future, where will greenhouses get their supplemental CO_2 from? This report aims to give insight into this topic.

Great efforts are being made to reduce carbon dioxide (CO_2) emissions. Governments and other organisations have set targets for carbon neutrality, which involve the phasing out of fossil fuels in favour of carbon-neutral alternatives.

Despite the need to reduce CO_2 emissions, CO_2 is an important resource for greenhouse horticulture. It is supplemented in many crops, pushing indoor concentrations to 600-1000 ppm, compared to the current 410 ppm outdoor average. This can increase yields by 20-30% (Nederhoff, 1994; Pan *et al.* 2019). Most of this supplied CO_2 comes from the combustion of natural gas on-site, in boilers or combined-heat-and-power (CHP) systems. Another important source of CO_2 is industry, where exhaust gases are purified and concentrated, and either piped or bottled and delivered by truck to greenhouses.

Though the contents of this report apply globally, this report focuses on the case of Dutch greenhouse horticulture. Over the past decades, huge efforts have been made to reduce the sector's dependence on natural gas. Recent price hikes and supply chain volatility have increased the urgency of these efforts. Electrified and 'fossil-free' greenhouses are expected to be the way forward, within the context of a decarbonised economy (Feije De Zwart, Vanthoor, & Koreneef, 2019). Since renewable energy sources generally do not release CO_2, demand for CO_2 from other sources is expected to increase (van Dijk, Meinen, & Dueck, 2014).

To alleviate this issue, the efficiency with which CO_2 is supplied could be improved. Currently, most supplemented CO_2 is lost though ventilation, about 85% (Vermeulen & van der Lans, 2011). Better greenhouse design and optimised dosage can help reduce this loss (P. De Visser, De Gelder, Warmenhoven, & Petropoulou, 2019). However, even with 100% efficiency, CO_2 would still have to be supplied from somewhere.

1.1   Outline

This report will look at CO_2 supply in four sections:

1. **Quantity** — How much CO_2 does the greenhouse horticulture sector need and how much is available?
2. Quality in terms of **concentration** — How concentrated does the CO_2 need to be, how concentrated are most sources, and how can the desired concentration be achieved?
3. Quality in terms of **purity** — Which contaminants are common, need to be removed, and how can they be removed?
4. **Distribution** — Which options are available for distribution?

The first section covers the expected CO_2 demand of the greenhouse horticulture industry and looks at the quantities of fossil-free CO_2 available. The following two sections cover two aspects of CO_2 quality: concentration (i.e. the volume percentage of the CO_2-containing gas) and purity (i.e. the concentration of toxic impurities). The difference is visualised in Figure 1 For both aspects, we look at requirements and technologies available to meet these requirements. Lastly, distribution options will be discussed, including storage.

These four steps affect system design. Depending on the solutions chosen, some of these steps may happen inside or outside the greenhouse. For example, boiler/CHP flue gases do not require concentration but are purified on-site. For CO_2 supplied by pipeline, all steps are done before transport.
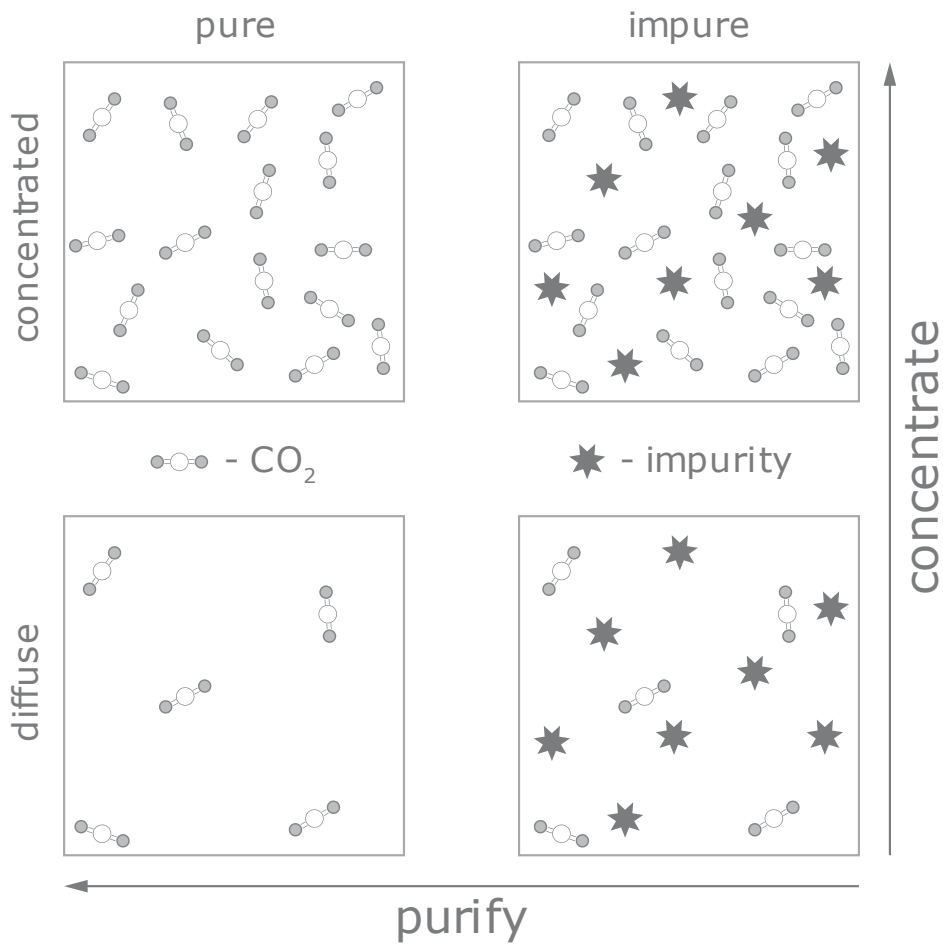
Figure 1 *A visualisation of the difference between purification and concentrations.*

Lastly, several novel scenarios for the near future will be selected. For each, we will give some order-of-magnitude calculations.

2   CO$_2$ Quantity

2.1   Demand

The entire sector uses an estimated 2.6 Mt of CO$_2$ annually (P. De Visser *et al.* 2019), with estimates ranging from 2.0 Mt (C. Blok, Boedijn, A., Brunsting, M., Verkerke, W., 2020) to the more generally-accepted figure of 2.5-2.6 Mt (Medema, 2022; Vermeulen & van der Lans, 2011).

Demand fluctuates throughout the year. Supply rates range between 100 and 300 kg CO$_2$ ha^{-1} h^{-1} (0.01 to 0.03 kg m^{-2} h^{-1}), though CO$_2$ is not continuously supplied so average supply rates are lower. The summer months require the most supplemental CO$_2$ (Mikunda, Neele, Wilschut, & Hanegraaf, 2015), since:

1. The abundance of sunlight leads to a higher crop growth rate, making CO$_2$ supplementation more profitable.
2. Higher temperatures mean high ventilation rates are required, leading greater losses of CO$_2$ and to a less efficient use.

Higher temperatures also mean the boiler system is used far less often during the summer and heat from the CHP being of no use, leading to less CO$_2$ being generated as a by-product and therefore requiring a dedicated CO$_2$ source instead. Some growers will burn natural gas during the summer just for its CO$_2$ (called 'zomerstook' in Dutch, which translates to 'summer heating'). The optimal dosage strategy will depend on crop growth factors as well as the price of CO$_2$, a balance of the marginal cost and marginal return of extra supplementation (Swinkels & de Zwart, 2002).

Different crops have different CO$_2$ dosage requirements, with some crops commonly not receiving any supplemental CO$_2$ at all. Crops typically take up between 13 and 15 kg of CO$_2$ per square metre per year (Vermeulen & van der Lans, 2011). Depending on the desired concentration and dosage efficiency, between 15 and 85 kg CO$_2$ m^{-2} is introduced into the greenhouse in total. In Table 1, an overview of the amount of CO$_2$ used in Dutch greenhouse horticulture in 2017 is given (Velden & Smit, 2019). Table 2 gives the dosages from experiments with specific crops.

Table 1
An overview of the average annual CO$_2$ dosage in 2017, in kg m^{-2}, for each of the subsectors in Dutch greenhouse horticulture, as well as the total annual quantity needed, in Mt (Velden & Smit, 2019).

Subsector	Area (ha)	Annual dosage (kg m^{-2})	Total per annum (Mt)
Vegetables	4585	36	1.6
Flowers	1815	29	0.5
Ornamental plants	2030	15	0.3
Seedlings	650	14	0.1
Total	9080	-	2.6

Table 2

An overview of the average annual CO_2 dosage in 2017, in kg m^{-2}, for each of the subsectors in Dutch greenhouse horticulture, as well as the total annual quantity needed, in Mt (Velden & Smit, 2019).

Crop	Annual dosage (kg m^{-2})	Source
Tomato	45	P. De Visser *et al.* (2019)
	23 (optimised)	De Gelder *et al.* (2012)
	46 (non-optimised)	
Pepper	14 (optimised)	Raaphorst (2018)
	39 (non-optimised)	
Cucumber	21	Kempkes *et al.* (2018)
Rose	70	De Gelder *et al.* (2015)

Total CO_2 demand will depend on the sector's scale, growers' supplementing habits, greenhouse design, and economic factors. Projections by Velden and Smit (2019) for 2030 are given in Table 3, where an annual demand of 1.8 to 3.0 Mt is expected.

Table 3

An overview of the average annual CO_2 dosage in 2017, in kg m^{-2}, for each of the subsectors in Dutch greenhouse horticulture, as well as the total annual quantity needed, in Mt (Velden & Smit, 2019).

| Subsector (Mt CO_2)	2017	2030		
		Low	Moderate	High
Vegetables	1.6	1.2	1.5	1.8
Flowers	0.5	0.4	0.6	0.8
Ornamental plants	0.3	0.2	0.3	0.4
Seedlings	0.1	0.1	0.1	0.1
Total	2.6	1.8	2.5	3.0

2.2   Non-Fossil Sources

Despite crops taking up CO_2, the carbon in crop biomass is eventually released into the atmosphere through decomposition. Therefore, greenhouse horticulture cannot be seen as a form of carbon sequestration, and all CO_2 going to greenhouses must be seen as emitted CO_2 (Mikunda *et al.* 2015). This is why eventually, non-fossil CO_2, from the short carbon cycle (as opposed to fossil CO_2, from the long carbon cycle) will have to be used, no matter the uptake efficiency.

60% of supplied CO_2 is currently generated by boilers and combined-heat-and-power (CHP) installations, with the remaining 40% being supplied externally as a by-product of industry. 80% of industrial CO_2 is supplied by OCAP (Organic CO_2 for Assimilation of Plants), a company that supplies CO_2 as a by-product of industrial processes (mostly fossil-based) to greenhouses though a pipeline (Mikunda *et al.* 2015). Although fossil fuels are to be phased out eventually, these sources of CO_2 will still carry on playing an important role for years to come.

This section aims to present non-fossil sources of CO_2 in the Netherlands and determine whether there is enough. Two alternative sources do not lead to net emissions: biogenic- (via biomass) and atmospheric CO_2. These two sources are indirectly the same, since biogenic carbon was once captured from the atmosphere by photosynthesis. Waste incineration is another potential source, but since it contains fossil-based materials, it will not be examined.

Unlike biogenic CO_2, the supply of atmospheric CO_2 is virtually limitless. Therefore, it will not be examined in this quantitative section. We will discuss anaerobic digestion, composting, aerobic digestion and combustion as CO_2 sources. Atmospheric CO_2 is far less concentrated than biogenic CO_2. Carbon capture methods will be presented later in this report, to concentrate both atmospheric and biogenic CO_2.
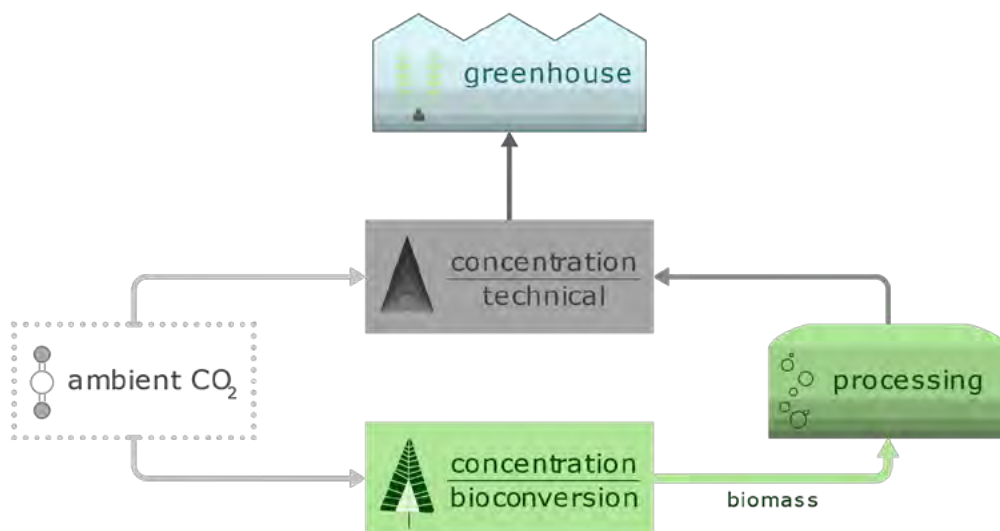
Figure 2 *A diagram showing how biogenic CO_2 comes from ambient CO_2, and how biomass is the result of a biological concentration step that is otherwise similar to technical concentration.*

There is more than enough biomass that could be directly oxidised to CO_2. However, when looking at the R-strategies for the circular economy (Kirchherr, Reike, & Hekkert, 2017), combustion should be seen as a last resort. Before combustion, biomass should ideally be cascaded though other processes first, to make use of its chemical energy and complexity elsewhere.

2.2.1   Anaerobic digestion

Anaerobic digestion involves the production of simpler organic compounds under anaerobic conditions by microbes, using an organic feedstock. Two common forms of anaerobic digestion will be covered in this section: methanogenic biodigestion and ethanol fermentation.

In methanogenic biodigestion, organic matter and its chemical energy are converted to biogas. Biogas is roughly 60% methane and 40% CO_2, along with contaminants such as H_2S (Rodin, Lindorfer, Böhm, & Vieira, 2020; Vermeulen & van der Lans, 2011). Though it can be directly combusted, biogas contains more CO_2 than the 6% legal limit for transport (Van Dijk, Dueck, & Burgers, 2009). Therefore, it is usually upgraded to natural gas standards by removing the CO_2 already present in biogas to produce biomethane, which is more energy-dense and has a wider range of applications. Many established techniques exist for biogas upgrading (Mikunda *et al.* 2015). Similar techniques will be discussed in Section 3.3.

Biomethane production is often expressed in petajoules (PJ). Assuming biogas is 40% CO_2, and methane's energy density is 52 MJ kg^{-1}, this means that per PJ of methane produced, 0.04 Mt of CO_2 is available from upgrading. This number can be used to calculate the amount of biogenic CO_2 available under various energy transition scenarios. The Dutch government has committed to a capacity of 70 PJ per year by 2030 (Delft, 2020). This indicates that biomethane production through anaerobic digestion has an annual capacity of 2.46 Mt CO_2[1].

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1   These ratios reflect those calculated by Mikunda *et al.* (2015), who say that per m^3 biomethane upgraded, 0.001 Mt CO_2 would be released.

Ethanol fermentation, also known as alcoholic fermentation, produces ethanol as a liquid, or is used in beverages. One mole of CO_2 is produced as a by-product per mole of ethanol, meaning that per kg ethanol produced, 955 g of CO_2 is released. Since ethanol is a liquid unlike methane, it is easier to separate the CO_2 from it (Rodin *et al.* 2020). Assuming 22 Mt of beer is produced in the Netherlands annually (Nederlandse Brouwers, 2022) at an alcohol percentage of 4%, ethanol fermentation amounts to only 0.08 Mt of CO_2 per year. CO_2 produced by the beverages industry is also often already used on-site for the carbonation of drinks, or to prevent spoilage (Rodin *et al.* 2020). Outside of the beverages industry, the OCAP network already uses CO_2 from Alco, a bio-ethanol plant in Rotterdam's Europoort (Mikunda *et al.* 2015). This bio-ethanol plant has an annual capacity of 0.3 Mt CO_2 (Khandelwal & van Dril, 2020).

2.2.2   Composting

Composting is the decomposition of solid organic matter under aerobic conditions. Many types exist with different organisms facilitating the composting process, including worms, fungi, and microbes. The by-products of composting are CO_2, ammonia (NH_3) and water. Composting is used to process food and garden waste, but also in industries such as the mushroom industry.

Inside the greenhouse, composting is can be used as a direct source of CO_2, as residual biomass decomposes during and after the end of the crop cycle. This can contribute between 0.9 and 1.7 kg CO_2 per m^2 greenhouse (Vermeulen & van der Lans, 2011). However, it is an internal flow, not a source of CO_2. After all, the carbon in this residual biomass had to first be supplied to the greenhouse. Instead, it should be seen as an improvement in efficiency, comparable to the optimal control of ventilation.

Only if the biomass composted inside the greenhouse is external could it be seen as an alternative CO_2 source. Vermeulen and van der Lans (2011) calculate that between 1.7 and 2.6 kg CO_2 m^{-2} would be released on an organic farm applying composting, depending on the biomass used. This would not be enough, and most of this would be released at the beginning of the crop cycle, when it is needed the least (Vermeulen & van der Lans, 2011).

In the Netherlands, 1.8 Mt of compost is produced annually (Winsen, 2018). Assuming 40-70% (wet weight) of the incoming feedstock is turned into compost, and 250-390 kg of CO_2 is emitted per tonne of feedstock (Boldrin, Andersen, Møller, Christensen, & Favoino, 2009). This indicates that composting has an annual capacity of between 0.6 and 1.8 Mt of CO_2.

2.2.3   Aerobic digestion

Like anaerobic digestion, aerobic digestion uses microbes in a controlled bioreactor, but in the presence of oxygen, similar to composting. Oxidative bioreactors are currently being investigated to produce liquid nutrients from organic matter, producing CO_2 as a by-product. The quality of the output depends on the feedstock. A consistent quality is feasible with a consistent feedstock, but with a more variable feedstock, there may be challenges. Oxidative biodigestion is unlikely to produce NO_x or ethylene, but may produce SO_2 and H_2S (C. Blok, van Winkel, & Boedijn, 2022). There are currently no oxidative biodigesters in the Netherlands. The placement of these will be a logistical consideration depending on the origin of the feedstock and destination of the CO_2 and other products.

With all organic by-products being produced in the Netherlands, aerobic digestion has an annual capacity of 6.2 Mt CO_2. However, many of these streams are already being valorised elsewhere, as animal feed or soil amendments, for example. When looking at unused streams, aerobic digestion only has an annual capacity of 0.7 Mt of CO_2 (C. Blok *et al.* 2022).

2.2.4   Combustion

As discussed at the beginning of this section, deliberate combustion should be seen as a last resort for biomass. The combustion of biomass (and biofuels) is often done in small operations scattered around the country, such as cars running on biomethane, or wood being burned in domestic stoves. This makes its CO_2 difficult to capture.

Despite this, biofuels and even biomass are expected to play a role in the future carbon-neutral energy mix, as a means to stabilise the grid when wind- and solar power are lacking (Arasto *et al.* 2017). In this way, biogenic CO_2 could be supplied from large-scale plants to greenhouse horticulture. Also, if used as an energy source on-site, CO_2 from the flue gases could be used as is done today (Van Dijk *et al.* 2009). Biomass and biofuels are currently the largest source of renewable energy in the Netherlands, at 54%. 27% of this goes to electricity, 53% to heat, and 20% to transport (StatLine, 2021b).

The production of biomethane and bio-ethanol is discussed in Section 2.2.1. 1 kg of methane and ethanol produce 2.7 and 1.9 kg of CO_2 respectively, though bio-ethanol is usually mixed with petrol and combusted in cars. Other feedstocks include wood, pellets and charcoal. For illustration, wood releases 1.6kg CO_2 kg^{-1} (NHK, 2018). Total CO_2 emissions from biomass – including from biofuels – were 19.4 Mt in 2020 (StatLine, 2021a).

2.3   Summary

Figures from the above paragraphs show that there is more than enough biogenic CO_2 for the greenhouse horticulture industry, and by 2030 there should be even more. By 2030, biogas upgrading and ethanol fermentation alone are expected to release enough CO_2 for the sector. We visualise this in Figure 3 and summarise the relevant figures in Table 4.
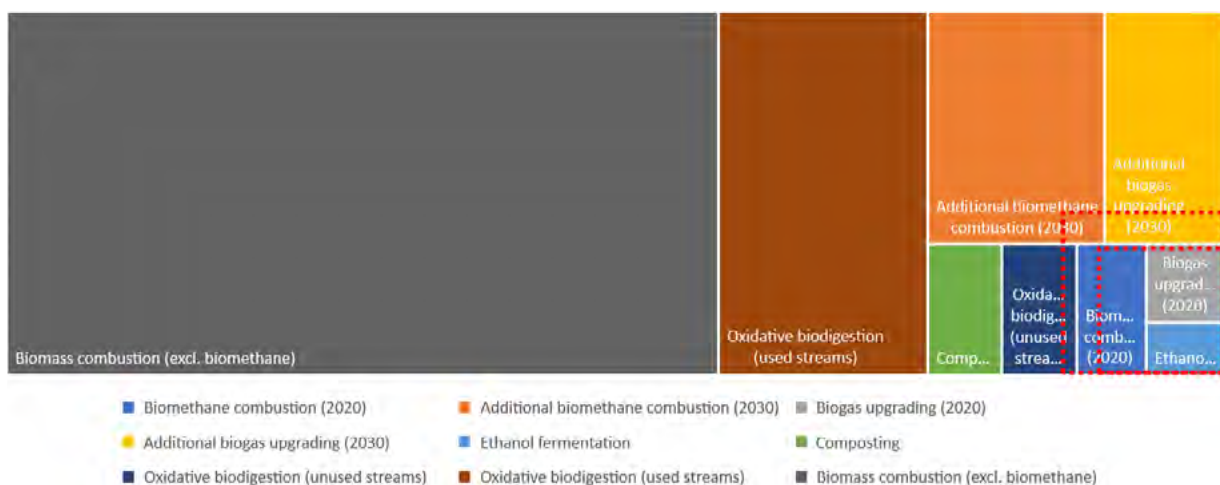
Figure 3 *A visualisation of the quantities of CO_2 mentioned in Table 4, below. The dotted red squares represent the predicted range of CO_2 demand from the industry by 2030, from Vermeulen and van der Lans (2011).*

Table 4

The amount of available CO_2 from various sources discussed in Section 2, in Mt-CO_2 y-1.

| Source	Amount (Mt-CO_2 y-1)
Biomethane combustion	0.66 (2020) 3.63 (2030 goal)
Biogas upgrading	0.44 (2020) 2.46 (2030 goal)
Ethanol fermentation	0.3 (Alco)
Composting	0.6 – 0.8
Oxidative biodigestion	0.7 (unused streams) 6.2 (all streams)
Biomass combustion (excluding biomethane)	18.7 (2020)
Industry requirement	1.8 – 3.0

3   Concentration

3.1   Requirements

Any concentration of CO_2 can be used for dosing, if it exceeds the desired indoor concentration (Vermeulen & van der Lans, 2011). However, as the CO_2 is diluted, the supply volume will need to increase, which increases supply and storage costs (Figure 4). This will be covered in Distibution & Storage (Section 5). An increase in volume will also result in stricter requirements for contaminants (Section 4.1.2).

OCAP requires CO_2 to be at least 98% pure for supply by pipes (Mikunda *et al.* 2015). Bottled CO_2 for greenhouse horticulture is sold at concentrations above 99.99% (Linde Gas, 2022a, 2022b), which is known as 'food-grade' CO_2 (Mikunda *et al.* 2015). There are exceptions, however: RoCa-3, a natural gas power plant near Rotterdam, also supplies CO_2 by pipe, after increasing its concentration from 10% through 'enrichment', though the precise concentration is not specified (De Wolf, 2009). In comparison, flue gases from boilers and CHP systems are typically 8% CO_2 by volume (Vermeulen & van der Lans, 2011).

3.2   Starting concentration

The starting concentration of the CO_2 source determines the ease of further concentration. There is a non-linear decrease in capture costs as starting CO_2 concentration decreases – with a much faster drop between 2.5% and 10% than between 10% and 20%, at a 90% carbon capture rate resulting in a concentration of over 98.5% (Husebye, Brunsvold, Roussanaly, & Zhang, 2012). This is shown for each source in Table 5.
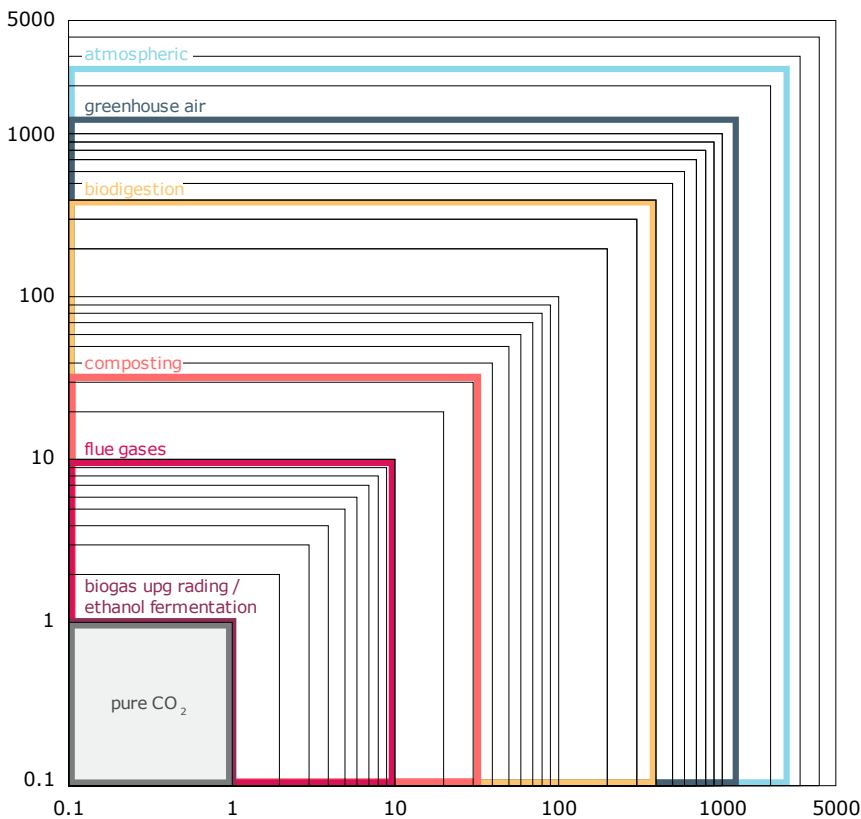
Figure 4 *A visualisation of the concentrations given in Table 5. The area of the squares, on a logarithmic scale, represent the volume required for the equivalent amount of CO_2 as in the 1x1 square in the bottom-left corner.*

Table 5

An overview of the minimum and maximum CO$_2$ concentrations (% volume) in various gas flows.

Source	Minimum	Maximum	Reference
Biomethane combustion	7	12	Vermeulen and van der Lans (2011), Esmeijer (1999)
Biomass combustion	3	8, 14	Rodin *et al.* (2020), Sánchez-Molina, Reinoso, Acién, Rodríguez, and López (2014)
Biogas upgrading	-	99	Rodin *et al.* (2020)
Ethanol fermentation	99	100	Rodin *et al.* (2020)
Composting	2	5	Nakasaki, Yaguchi, Sasaki, and Kubota (1990)
Oxidative biodigestion	0.17	0.37 (estimate) 0.74 (measured)	C. Blok *et al.* (2022)

* At 99% CO$_2$, biogas upgrading looks very attractive, but this is CO$_2$ that has already been through a capture process from biogas (40% CO$_2$) which gives it an 'unfair' advantage.

3.3 Concentration Technologies

In this section, a number of CO$_2$ concentration technologies are discussed. This includes their working principles, technology maturity, and factors related to cost and efficiency. Not all existing technologies are discussed here, but those found to be most relevant. Bioconversion of CO$_2$ to biomass (i.e. photosynthesis) is not examined here, since biogenic carbon has already been discussed as a source in this report. Many of these technologies are known as carbon-capture-and-utilisation (CCU) technologies, as opposed to carbon-capture-and-storage (CCS), where CO$_2$ is only stored but not meant to be re-deployed later.

3.3.1 Physical/chemical absorption

Physical or chemical absorption is currently the most mature CO$_2$ concentration technology. Depending on the concentration of incoming gas, its absorption efficiency is over 90% (Leung, Caramanna, & Maroto-Valer, 2014). The working principle involves using a sorbent (e.g. lime) to separate CO$_2$ from the rest of the gas. The CO$_2$ is then released as a purer stream. Physical absorption uses water scrubbers, as water is cheap and has no harmful environmental impact. Under high pressure and temperature, the CO$_2$ is dissolved (Leung *et al.* 2014). This requires high amounts of energy, a significant disadvantage. Chemical absorption uses specific scrubber liquids such as amines or selexol, optimised for CO$_2$ absorption (De Keulenaere, Tessens, Willeghems, & Buysse, 2017). A disadvantage to chemical absorption is that it also requires high amounts of heat, for sorbent regeneration. The environmental impacts associated with sorbent degradation also need to be investigated (Leung *et al.* 2014). In Section 6.2.3, we cover an example with lime-based absorption.

3.3.2 Membrane technology

Using selective membranes, CO$_2$ can be separated and concentrated, much like how water is purified using reverse osmosis. This can achieve a CO$_2$ concentration of up to 80-90% (Leung *et al.* 2014; Sander Peeters, 2013). Multi-stage systems with multiple membranes also exist, to increase CO$_2$ concentrations further. Membrane technology is currently recommended for small-scale applications (De Keulenaere *et al.* 2017). The disadvantages to membrane technology are low flux rates and membranes being prone to fouling (Leung *et al.* 2014).

3.3.3 Pressure swing adsorption

In pressure swing adsorption (PSA), CO_2 binds to a molecular sieve, made of a material like activated carbon or zeolite. Once enough CO_2 has bound to the material, the sieve is brought under low pressure, the CO_2 is released and the sieve is regenerated. A disadvantage to PSA is that H_2S binds irreversibly to the sieve, so it should first be removed separately (De Keulenaere *et al.* 2017).

3.3.4 Cryogenic distillation

Cryogenic distillation involves cooling and compressing the CO_2-containing gas to a liquid. The fractions are then separated at their various condensation points. The advantage of cryogenic distillation is the high purity of CO_2 that can be achieved (De Keulenaere *et al.* 2017). The disadvantages are that it is energy-intensive (Leung *et al.* 2014) and best used for high starting concentrations. Therefore it may be used after concentrating CO_2 with a different technology first (Mikunda *et al.* 2015).

3.3.5 Electrochemical reduction

Organic compounds can be produced from CO_2 using reduction (the inverse of oxidation), which requires an input of energy. Many reduction processes exist, including hydrogenation, reverse water gas shift reaction, Fischer-Tropsch synthesis (Rodin *et al.* 2020), and Sabatier synthesis (Jarvis & Samsatli, 2018).

In electrochemical reduction, an electrolytic cell uses electricity to convert dissolved CO_2 via a catalyst (Agarwal, Zhai, Hill, & Sridhar, 2011) at ambient temperatures and pressures (Agarwal *et al.* 2011; Jarvis & Samsatli, 2018). The typical CO_2 conversion rate is 85%. Electro-reduction can be turned into many products – methanol, methane, and carbon monoxide – but formic acid currently has the highest likelihood of commercialisation and a favourable energy consumption. Despite these advantages, electrochemical reduction's technology readiness level is low (Jarvis & Samsatli, 2018).

Though electrochemical reduction is far more energy-intensive than most other techniques, its product is not concentrated CO_2 but a fuel with stored chemical energy that can be used later. Therefore its energy consumption is not directly comparable to other techniques. Much like natural gas is both a source of energy and CO_2 in greenhouses today, formic acid or other organic fuels may be able to play a similar role in the future using a fuel cell. Greenhouses that use electricity instead of natural gas have the advantage of decoupling energy consumption from CO_2 release. Using synthetic fuels from electrochemical reduction may be a step backwards in this regard, but these fuels are easier to transport and store than gaseous CO_2.

3.4　Summary

Table 6

An overview of the energy consumption (MWh/kt-CO$_2$) and operating pressures (bar) and temperatures (°C) for the various carbon concentration technologies. All figures are calculated from Mikunda et al. (2015) (where they were used to describe biogas upgrading technologies) except for methanol- and formic acid reduction, which are from Rumayor, Dominguez-Ramos, Perez, and Irabien (2019) and (Thonemann & Schulte, 2019) respectively. A figure for membrane technology on its own was obtained from Fujikawa, Selyanchyn, and Kunitake (2021).

	Energy consumption MWh/kt-CO$_2$	Pressure bar	Temperature °C
Absorption (PWS)	158-198	5-10	10-25
Absorption (amine)	< 71	0.05-0.5	10-40
Membrane technology	158-238	5-16	Ambient
Membrane technology*	4500-17000	5	10-20
Pressure swing adsorption	150-206	4-7	10-20
Cryogenic distillation**	198-293*	17	< -80
Reduction (methanol)	7250-7970	≥ Ambient	Ambient
Reduction (formic acid)	129-19226	≥ Ambient	Ambient

*　from ambient concentrations to 10% concentration (Fujikawa, Selyanchyn, & Kunitake, 2021)

** for hybrid technology with membrane

3.5　Direct Air Capture

The previously-discussed technologies could be used to concentrate biogenic CO$_2$, but also atmospheric CO$_2$. Despite the lower CO$_2$ concentration in the atmosphere, one potential advantage is a steady and predictable supply (Mikunda *et al.* 2015).

Most studies covering direct air capture look at converting the 410 ppm of CO$_2$ currently in the atmosphere to a stream that is over 99% concentrated. However, this is not necessarily required for greenhouse horticulture, since greenhouses are controlled to 800-1000 ppm, or 0.08-0.1%. With a high enough ventilation rate, this could be achieved with far lower concentrations of CO$_2$. A (semi-) closed greenhouse design to reduce leakage would reduce the ventilation rate required as well.

The higher the ingoing concentration, the lower the required ventilation rate, and vice versa. This can be calculated by rearranging the following equation:

$$\phi_{CO_2} = \phi_v \cdot (C_{source} - C_{in})$$

In which ϕ_{CO_2} is the mass flow of CO$_2$ (kg ha^{-1} h^{-1}), ϕ_v the volumetric flow of air due to ventilation (m^3 ha^{-1} h^{-1}), and C the concentration of CO$_2$, from the source and inside the greenhouse. Assuming a dosage rate of between 100 and 300 kg ha^{-1} h^{-1}, we can calculate possible combinations of concentrations and ventilation rates. These are visualised in Figure 5, and can be used for scenarios for future research into DAC.
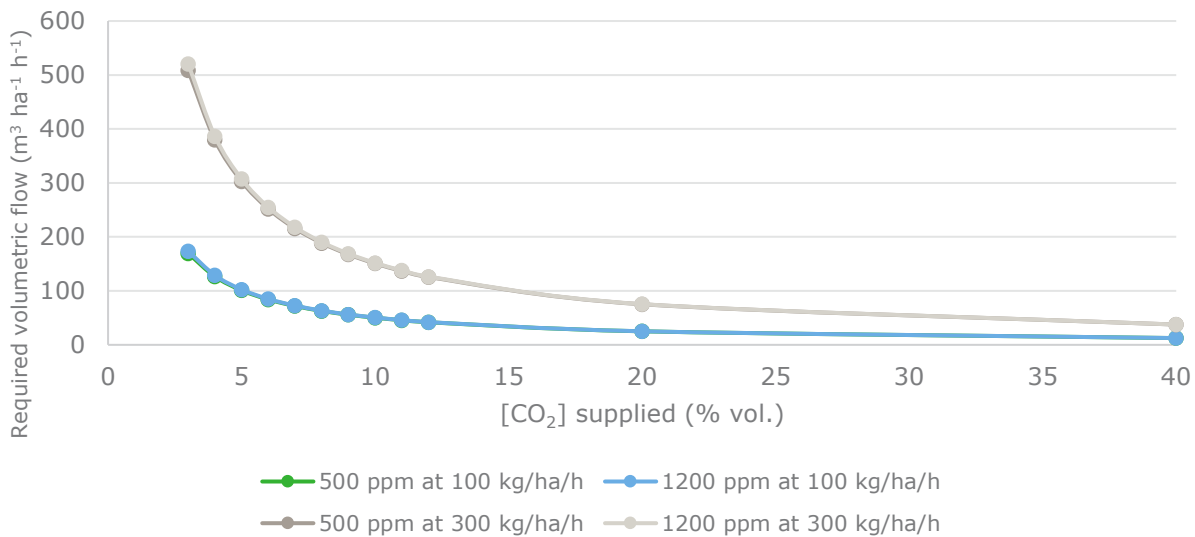
Figure 5 *A visualisation of the required volumetric flow to achieve two dosage rates – 100 and 300 kg ha^{-1} h^{-1} – for two desired indoor concentrations – 500 and 1200 ppm – as a function of the concentration of CO_2 in the flow supplied (% volume). Anything below 175 m^3 ha^{-1} h^{-1} is a realistic volumetric flow.*

Given that typical volumetric flows are between 150 and 175 m^3 ha^{-1} h^{-1}, a low-concentration DAC unit may be feasible starting at around just 4% CO_2. Figure 5 shows us that the dosage rate has a far larger effect on required volumetric flow than the desired concentration: The difference between 500 and 1200 ppm, two extremes, is imperceptibly small compared to the difference between 100 and 300 kg ha^{-1} h^{-1}. This also shows the importance of reducing leakage and increasing efficiency. We go into a potential novel scenario using low-concentration local DAC in Section 6.2.1.

At the time of writing, the Kas als Energiebron ('Greenhouse as an Energy Source') programme has a research project investigating the use of DAC in greenhouse horticulture (Kas als Energiebron, 2020). They conclude that pure CO_2 from DAC is more efficient than using low concentrations in enriched air, for three reasons (Ros, 2022):
1. Both approaches have a similar energy consumption, but a greater volume of air has to be heated in the enriched air approach.
2. CO_2 storage is much more difficult with enriched air, due to the lower CO_2 concentration. Since greenhouse demand fluctuates, an enriched air system ends up having being of a larger size and capital expenditure.
3. Releasing large volumes of air is not convenient, according to growers, and its effect on the rest of the greenhouse operation has yet to be seen.

The first two points do not rule out the idea of using DAC during the summer, when demand is high and incoming air does not need to be heated. Furthermore, though the concentration of CO_2 under their enrichment scenario is not mentioned, these considerations are important, and suggest that even though 100% CO_2 is still not required, the enriched air should not be of a too low concentration.

4 Purification

4.1 Requirements

4.1.1 Indoor air

Plants are susceptible to chemicals in the air that can reduce growth or interfere with development. These contaminants can also be damaging to the greenhouse itself, as is the case with ozone (Dueck, van Dijk, Kempkes, & van der Zalm, 2008). For contaminants, both the concentration and duration of exposure influence phytotoxicity. Typical threshold levels are given in Table 7.

Table 7

An overview of the energy consumption (MWh/kt-CO_2) and operating pressures (bar) and temperatures (°C) for the various carbon concentration technologies. All figures are calculated from Mikunda et al. (2015) (where they were used to describe biogas upgrading technologies) except for methanol- and formic acid reduction, which are from Rumayor, Dominguez-Ramos, Perez, and Irabien (2019) and (Thonemann & Schulte, 2019) respectively. A figure for membrane technology on its own was obtained from Fujikawa, Selyanchyn, and Kunitake (2021).

Contaminant	Theshold (ppb)	Duration	Source
NO_X	600	Acute	Esmeijer (1999)
	40	24 hours	Dueck *et al.* (2008), Vermeulen and van der Lans (2011)
	16	1 year	
	100	Chronic	Esmeijer (1999)
C_2H_4	50	Acute	Esmeijer (1999)
	11	8 hours	Dueck *et al.* (2008), Vermeulen and van der Lans (2011)
	5	4 weeks	
	8	Chronic	Esmeijer (1999)
SO_2	70	Acute	Esmeijer (1999)
	100	24 hours	Vermeulen and van der Lans (2011)
	30	1 year	
	15	Chronic	Esmeijer (1999)
O_3	100	Acute	Esmeijer (1999)
	30	Chronic	

Several compounds have known phytotoxicity, but unknown threshold concentrations and durations: C_2HCl_3, C_2Cl_4, C_6H_6 (benzene), HCl, and HF (Vermeulen & van der Lans, 2011). Compounds with unknown phytotoxicity are: sulfides, mercaptans, ether compounds, NH_3, methane compounds, toluene, and silicon oxide (Vermeulen & van der Lans, 2011). van Dijk *et al.* (2014) recommend investigating the phytotoxicity of ethanol, 1-propanethiol and 1-propanol. Benzene and dimethylsulfide (DMS) were tested up to concentrations of 800 and 600 ppb respectively and no negative effects were found (van Dijk *et al.* 2014).

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[2] Ethylene's effect depends on the crop and the duration of exposure, but in a different way to other contaminants. Ethylene is a plant hormone that stimulates ageing and affects processes such as ripening (Esmeijer, 1999). Unlike with other contaminants, ethylene is harmful at low concentrations but not at higher ones (Dueck *et al.* 2008). Pepper plants have also been found to be able to adapt to a constant concentration, but react poorly to a single peak in ethylene (van Dijk, Meinen, & Dueck, 2011).

4.1.2   In gas supplied

The higher the concentration of CO_2 desired in the greenhouse, and the lower the concentration of CO_2 in the flue gas, the lower the allowed maximum concentrations for contaminants in the flue gas are. Vermeulen and van der Lans (2011) give equations for the threshold concentrations of NO, NO_2, C_2H_4, NH_3 and O_3 in flue gases from CHPs. Figures 6 and 7 illustrate how the two aforementioned factors affect the requirements of flue gases.
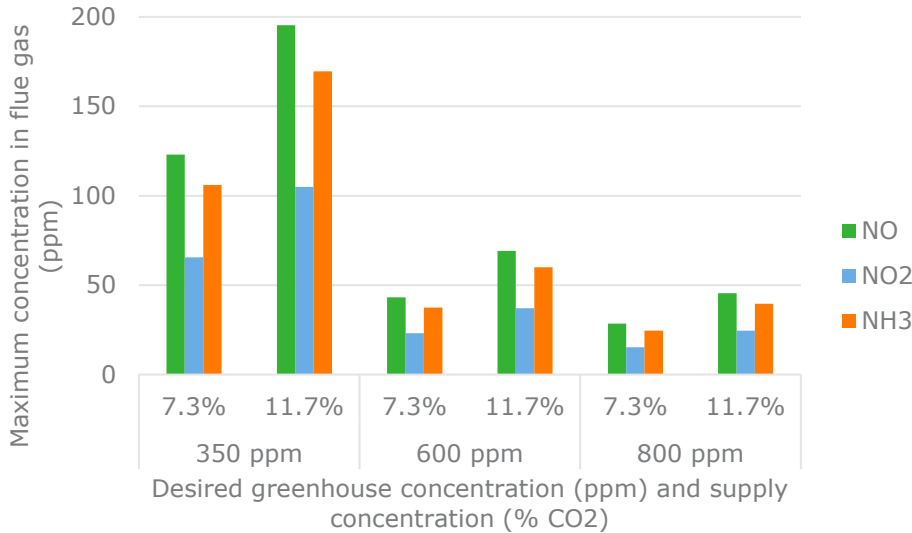
Figure 6 *A visualisation of the required volumetric flow to achieve two dosage rates – 100 and 300 kg ha^{-1} h^{-1} – for two desired indoor concentrations – 500 and 1200 ppm – as a function of the concentration of CO_2 in the flow supplied (% volume). Anything below 175 m^3 ha^{-1} h^{-1} is a realistic volumetric flow.*
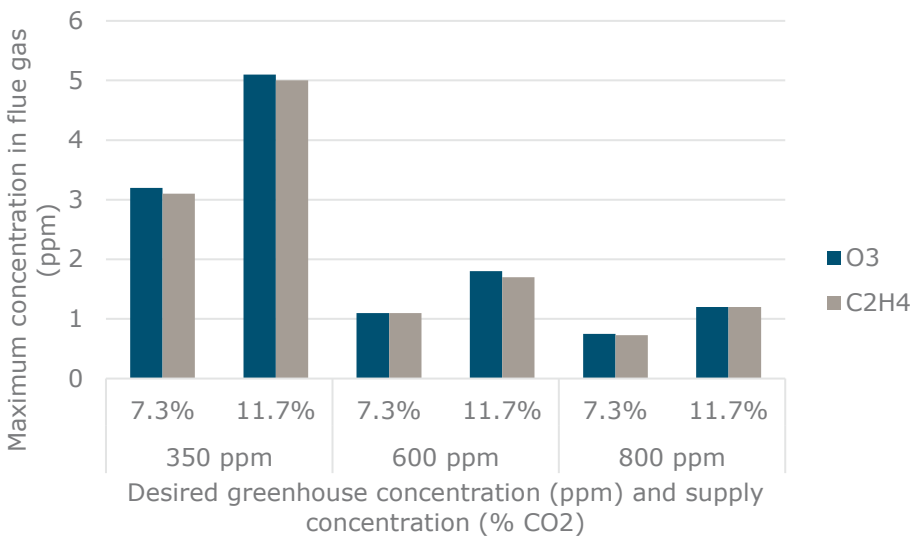
Figure 7 *A visualisation of the maximum concentrations in flue gas for ozone (O_3) and ethylene (C_2H_4) (ppm), depending on the percentage of CO_2 in the flue gas and the desired CO_2 concentration (ppm).*

4.2   Contaminants in non-fossil sources

A few studies have investigated contamination in alternative sources of CO_2 – for example, van Dijk *et al.* (2014) conclude that the contaminants in biogas are unlikely to be of a sufficient level to affect horticultural crops after combustion. Concentrations of contaminants in flue gases also depend on the kind of motor used for combustion. Discontinuous combustion, used in CHPs, releases far more contaminants than continuous combustion processes such as boilers. Also, for C_2H_4, the risks from biogas are the same as for natural gas (Van Dijk *et al.* 2009).

Table 8 gives an overview of the concentrations of various contaminants per source.

Table 8

An overview of the likelihood of occurrence of contaminants found in CO_2 from the sources identified in Section 2.2. The main three contaminants; NOx, ethylene and SO_2; are given. Other significant contaminants are listed in the 'Others' column.

Source	NO$_x$	C$_2$H$_4$	SO2	Others	References
Biomethane combustion	likely	likely	likely	HF, C$_6$H$_6$	Van Dijk *et al.* (2009)
Biogas upgrading	unlikely	unlikely	unlikely	H$_2$S (often pre-separated), NH$_3$, VOCs	Rodin *et al.* (2020)
Ethanol fermentation	unlikely	unlikely	unlikely	unlikely	Rodin *et al.* (2020)
Ethanol combustion	unlikely	unlikely	unlikely	unlikely	Ethanol Fuel - EIBIP
Composting	likely	likely	likely	H$_2$S, NH$_3$	Dhamodharan, Varma, Veluchamy, Pugazhendhi, and Rajendran (2019), Dsouza, Price, Dixon, and Graham (2021), Al-Rumaihi, McKay, Mackey, and Al-Ansari (2020)
Oxidative biodigestion	unlikely	unlikely	likely	H$_2$S, depending on feedstock	C. Blok *et al.* (2022)
Biomass combustion	yes	likely	yes	VOCs, particulate matter	Singh and Shukla (2014)

4.3 Purification technologies

In this section, several purification technologies will be discussed. A more complete summary is given in Table 9 (Section 4.4). For brevity, we will only discuss technologies that can remove one of three main contaminants described above: NO$_x$, SO$_2$ and C$_2$H$_4$. Depending on whether the CO$_2$ stream was concentrated beforehand, and the outcome of this process, purification technologies may not be necessary if whilst concentrating the CO$_2$, it was separated from impurities. It is also important to note that many of the aforementioned sources may be purified anyway, e.g. biomass combustion plants may already remove their NO$_x$ and SO$_2$. Some technologies can remove both NO$_x$ and SO$_2$ simultaneously. These are therefore covered together in their own section.

4.3.1 NO$_x$

There are two main approaches to removing NO$_x$: ad/absorption, or destruction of NO$_x$ by transforming it into benign compounds (Gholami, Tomas, Gholami, & Vakili, 2020).

Selective catalytic reduction (SCR) is the most common approach to removing NO$_x$ (Gholami *et al.* 2020). There are multiple variants, of which the most commonly-used in greenhouse horticulture is urea injection, for cleaning CHP flue gases on site (Vermeulen & van der Lans, 2011). SCR is simple, efficient, and has low running costs. However it has high investment costs due to the catalyst and requires high operating temperatures, of at least 180 °C (Guan, Zhan, Lin, & Huang, 2014). The same advantages apply to selective non-catalytic reduction, except it costs less to install due to lack of catalyst. However, because of this, its efficiency is lower and it requires higher temperatures, at least 850 °C. Other approaches to transform NO$_x$ into more benign compounds include electron beams, non-thermal plasma and electrochemical reduction. The first two have high energy costs but produce no by-products. Electron beam technology has a low NO$_x$-removal efficiency, but it does produce useful by-products: ammonium nitrate and -sulfate (if SO$_2$ is present). The last technique, electrochemical reduction, is still in its infancy (Gholami *et al.* 2020).

Two commonly-used techniques in the category of ad/absorption are wet scrubbing and adsorption. Wet scrubbing works under ambient temperature and has a low cost. However, it has a low efficiency, requiring large multi-stage scrubbers, and produces a liquid by-product. The products of scrubbing could, however, be used as fertilisers. Adsorption does not produce a by-product, has a high efficiency, and has simple equipment. The disadvantages is that this equipment is large and has a high investment cost (Gholami *et al.* 2020).

4.3.2 SO$_2$

Multiple technologies exist for removing SO$_2$ – one of the sulfur oxide ('SO$_x$') gases, the other being SO$_3$, that can be found in flue gases (Asghar *et al.* 2021). Systems for SO$_2$ removal include physical cleaning, chemical/biological cleaning, and dry sorbent injection. Wet scrubbing, also known as wet flue gas desulfurisation (WFGD) is the most widely-used (Asghar *et al.* 2021). This is discussed in Section 4.3.3, where we outline technologies that remove both SO$_2$ and NO$_x$.

4.3.3 NO$_x$ and SO$_2$

Various technologies are good at removing NO$_x$ and SO$_2$ simultaneously: wet scrubbing techniques, non-thermal plasma, and adsorption. Wet scrubbing techniques can produce fertiliser as a by-product, whilst still effectively removing both contaminants (unlike electron beam technology, which also produces fertiliser but is inefficient at removing NO$_x$). In other studies, ozone injection and ammonia have been used to remove both NO$_x$ and SO$_2$, whilst also generating fertiliser (Asghar *et al.* 2021). All in all however, adsorption is recommended for systems that have to remove both NO$_x$ and SO$_2$ (Gholami *et al.* 2020).

4.3.4 Ethylene (C$_2$H$_4$)

Like the aforementioned contaminants, scrubbers can also be used to remove ethylene, using oxidising agents like potassium permanganate (KMnO$_4$), which convert ethylene into CO$_2$ and water whilst being reduced to KOH and MnO, under ambient temperatures (Mabusela *et al.* 2021). KMnO$_4$ is also used in some NO$_x$ and SO$_2$ scrubbers (Gholami *et al.* 2020). Photocatalytic oxidation (PCO) is another technology. Like scrubbers with oxidising agents, it works under ambient temperatures, but it has the extra advantage of producing only CO$_2$ and water. However, the technology is still in its infancy. Vacuum ultraviolet radiation has better potential to remove ethylene, but it does generate ozone (O$_3$) (Mabusela *et al.* 2021), which may be a problem in greenhouse horticulture at high enough concentrations (see Sections 4.1.1 and 4.1.2).

4.4 Summary

In Table 9, an overview of technologies for contaminant removal – for NO$_x$, SO$_2$ and ethylene – is given. Based on it, the most effective solution is a combination of wet scrubbing and adsorption. However, which solution is used in practice will depend on where the contaminants are removed: at the greenhouse or on an industrial scale.

Table 9

An overview of various technologies for contaminant removal, with the number of '+' signs corresponding to efficacy. The main advantages and disadvantages are given for each as well.

Technology	NO_x	SO_2	C_2H_4	Advantages	Disadvantages
Selective catalytic reduction	+++			Low operational cost Simple installation	Catalyst costs/replacement Waste High equipment cost High temperature required
Selective non-catalytic reduction	+			Low operational costs Low equipment cost	Inappropriate for low-NO_x High temperature required Ammonia in by-product
Wet scrubbing	+	+++	+++	Ambient temperature Low equipment cost	Liquid by-product Large multi-stage scrubbers
Electron beam	++	+++		No waste Fertiliser by-product	High energy consumption
Adsorption	+++	+++		No liquid by-product Simple equipment	High equipment cost Large
Electrochemical reduction	++			No reductant consumed	Early development stage
Non-thermal plasma	+			Low equipment cost No waste Simple operation Fertiliser by-product	High energy consumption Low operating pressure
Physical cleaning		++			
Chemical/biological cleaning		++			
Dry sorbent injection		++			
Biofiltration			+++	Environmentally-friendly Cost-effective	Slow Large required area
Photocatalytic oxidation			++		
Vacuum ultraviolet radiation			+++		Generates O_3

5 Distribution & Storage

In this section, two distribution possibilities are discussed: by truck and by pipe. Both are currently used in greenhouse horticulture. The use of CO_2 storage as a buffer is then discussed. For both transport and storage, scale is important. To achieve economies of scale, hubs or clusters are recommended (Mikunda *et al.* 2015).

5.1 Distribution

5.1.1 By truck

Especially for areas without pipe infrastructure, bottled CO_2 transported by truck is the most feasible option short-term. Trucks have a capacity of between 20 and 25 t CO_2 and cost €650-800 per delivery, equating to €26-40 t^{-1} CO_2 (Mikunda *et al.* 2015). Assuming all CO_2 is delivered by truck, a typical 5 ha greenhouse requiring 460 t CO_2 ha^{-1} annually (De Gelder *et al.* 2012) would receive 92 to 115 trucks in a year. Bottled CO_2 transported by truck is usually over 99% pure (Linde Gas, 2022a, 2022b).

5.1.2 By pipe

The OCAP network already supplies CO_2 to the greenhouse horticulture industry by pipe, currently supplying 0.4 Mt annually to 20% of the industry's surface area. The main OCAP pipeline stretches from the Europoort in Rotterdam to Amsterdam, and expansions have been planned (Mikunda *et al.* 2015).

Though setting up such an infrastructure involves costs, the variable cost is lower than CO_2 delivered by truck. Van der Linden (2019) estimates that a CO_2 network with a capacity of 30 Mt coming from industrial clusters around the Netherlands would cost between €1.9-3.5 billion before 2030. The cost of transporting CO_2 would amount to €9 t^{-1}.

Assuming greenhouses become electrified and do not use biomethane instead of natural gas, existing pipelines could be converted to transport CO_2. Estimates say repurposing existing pipelines for CO_2 would cost 1-10% of the cost of building new CO_2 pipelines from scratch, whilst utilising an existing asset, though these pipelines are currently used to supply natural gas to more than just greenhouse horticulture. The main challenge is that CO_2 must be transported under a higher pressure. However, such conversions have been proven to be feasible, with OCAP itself being an example of a pipeline that used to transport oil (Kenton, 2022). The difference is that the oil pipeline used for OCAP had a very specific use with only one destination, compared to current gas pipelines.

Though contaminant requirements for plants have been examined in Section 4.1, transport by pipeline has its own contaminant requirements too. These come from two sources: DYNAMIS, funded by the EU 6th Framework Programme (E. De Visser *et al.* 2008); and the CO_2Europipe project, funded by the EU 7th Framework Programme. These recommendations are likely to become relevant to EU countries, and can be found in a source by Sun *et al.* (2015).

5.2 Storage

Much like a rainwater buffer allows a grower to use more rainwater (and rely less on other sources), CO_2 storage can help the greenhouse horticulture industry meet its requirements year-round by allowing surplus supply to be stored for months with higher demand. This can be done on a local level, but also on an industry level. A diagram visualising the effects of storage on an industry level for the sector is given below, in Figure 8 (Mikunda *et al.* 2015).
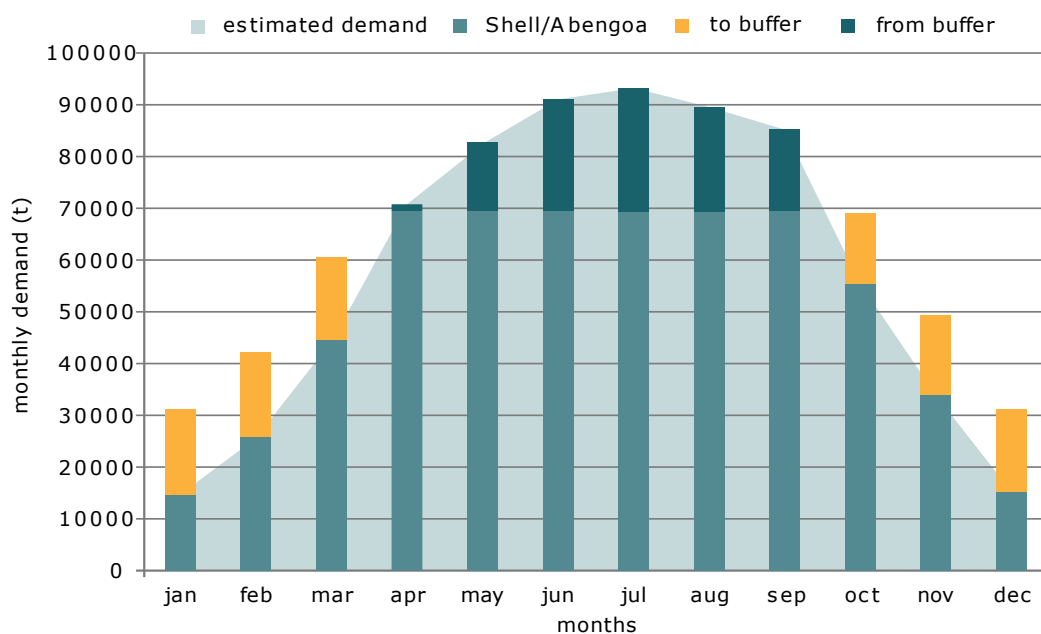
Figure 8 *A monthly visualisation of how CO_2 buffers can help the sector meet its requirements year-round, adapted from Mikunda et al. (2015).*

Storage can be done in tanks, known as surface buffering, or geologically. In a study from 2010, surface buffering was not found to be economically feasible (Smit, 2010). Limiting factors include energy consumption and compression costs (Vermeulen & van der Lans, 2011). Mikunda *et al.* (2015) compared surface buffering to geological buffering. Though cost and supply security information was unavailable, they concluded that geological buffering would have better coverage, whereas surface buffers could be installed more quickly. Surface buffering has an excellent technical feasibility as it is an established technology with a low innovation potential. Geological buffering is a less-developed technology, being less feasible but showing a medium-level innovation potential. It also presents an uncertain CO_2 quality as contaminants may end up in the stream.

At the greenhouse, CO_2 can be buffered more short-term – for example, to store CO_2 generated at night for use during the day or on a weekly level. The higher the concentration of CO_2 in the gas being stored, the less storage space required and therefore the lower the cost. This relationship is linear (Vermeulen & van der Lans, 2011). A recent example of this in practice is an aubergine grower in Zeeland, who uses CO_2 released and stored from biomass combustion (Redactie Duurzaambedrijfsleven.nl, 2019).
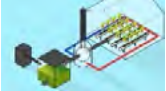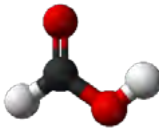
Liquid organic compounds from electrochemical reduction (such as formic acid or methanol) would be even cheaper to store, but come with the disadvantage of coupling CO_2 release with energy release. Looking at the demand pattern in Figure 8, this would happen during the summer, when heat is not required. However, this energy could be used for other processes on-site, or be sold to the grid.

6   Scenarios

6.1   Overview

Table 10

An overview of alternative CO_2 options for greenhouse horticulture, with the steps outlined in this report and how (and whether) they are carried out. Section 6.2 gives quantitative information on three of these: local DAC, regional biodigestion, and the lime-based absorption system.

	Local DAC	Regional biodigestion	Lime-based absorption	On-site combustion	Formic acid fuel cell
Source	Ambient air	Biomass fermentation	Biomass	Biomethane	Formic acid from various C sources
Concentrate	Membrane technology (to 10%)	Already done (to 99%)	Absorption (lime)	No need (8%)	Electroreduction
Purify	No need	Already done	Absorption (lime)	Mainstream technologies	No need
Distribute (as CO_2)	No need	OCAP pipes Bottled	No need	No need	No need
Store	Tank	Tank	Pellets	As methane only	As formic acid only

6.2   Calculations

6.2.1   Local Direct Air Capture

In this scenario, the greenhouse would obtain all of its CO_2 directly from ambient air. This eliminates the need for transport and the removal of contaminants, assuming no contaminants are concentrated during this process. The only step is the concentration of ambient CO_2 to useful levels. Because the CO_2 does not need to be transported, and greenhouses only require 800-1000 ppm (i.e. 0.08–0.1%), this level need not be high, as calculated in Section 3.5.

Existing greenhouses use a network of internal pipes to distribute CO_2 from flue gases to the crops. The size of these pipes, pictured in Figure 9, corresponds to the CO_2 concentration being distributed. Therefore, in this example, we assume the CHP is being replaced with a DAC unit and CO_2 is concentrated to 8%, the same level as in flue gases.

Figure 9 *An example of the pipes (light green) used for CO_2 distribution from flue gases, obtained through personal communication with Frank Kempkes. The size of these pipes corresponds to the concentration of CO_2 being distributed, which is 8% in the case of CHP flue gases.*

We choose a system based on membrane technology because is well-suited to small-scale applications and a high CO_2 concentration is not required. Fujikawa *et al.* (2021) describe a membrane DAC system where 3-4 membrane stages would be required to achieve concentrations of over 10%, and would work using modules of $0.01\,m^3$ for 1 kg capture per day. To achieve peak supply capacity of 300 kg ha^{-1} h^{-1}, (equating to 2400 kg ha^{-1} d^{-1} assuming 8 hours of peak demand), a system of 24 m^3 ha^{-1} would be required. Therefore to reduce its size, such a system would best be combined with buffering whilst running at night, when no CO_2 is required. Alternatively, external CO_2 could be supplied to meet peak demand. Assuming enough to supply only 100 kg ha^{-1} h^{-1} for 8 hours (800 kg ha^{-1} d^{-1}) is captured over 24 hours, a system of 8 m^3 ha^{-1} would be required, excluding the buffer. Using the range given by Fujikawa *et al.* (2021) in Table 6, this system would consume between 3600 and 13 600 kWh daily.

For context, between 40 000 and 119 000 m^3 gas is burned per hectare in the summer for its CO_2, producing unnecessary heat (Raaphorst & Kempkes, 2019). At 10.55 kWh m^{-3}, this equates to between 422 000 and 1 255 450 kWh per year, or, assuming a summer of 100 days, between 4220 and 12 555 kWh d^{-1} on average. Still, this would supply more CO_2 than our proposed DAC system. However, the best-case membrane technology (4500 MWh $kt\text{-}CO_2^{-1}$) does use less energy than is released by burning natural gas (5400 MWh $kt\text{-}CO_2^{-1}$).

A better application of DAC may be to only use it in the summer months, to meet peak demand in addition to another source such as OCAP CO_2. This would require a smaller DAC unit and be more energy efficient. However, in this case, this begs the question of whether simply using a CO_2 buffer would not be a more efficient and cost-effective way to achieve the same thing.

6.2.2 Regional Biodigestion

In this example, we assume a regional biodigestor processing 250 000 t of pig manure, equivalent to 2.5% of the annual amount produced in the Netherlands. Pig manure has an organic matter content of 79 g kg^{-1}, equating to 19 750 t organic matter. By multiplying this with the biochemical methane potential (BMP, 0.36) and methane conversion factor (MCF, 0.94), we estimate a methane production of 9 975 224 m^3. As biogas is 60% methane and 40% CO_2, 6 650 149 m^3 CO_2 is produced as a by-product, equating to just over 13 kt (Boedijn, Casu, & Booijen, 2022). Depending on dosage efficiency, a tomato greenhouse uses between 26 and 65 kg CO_2 m^{-2} annually, meaning between 50 and 20 hectares could be supplied respectively.

Biogas upgrading, which would be done on-site, produces a CO_2 flow concentrated enough to be distributed either by pipe or in bottles. This flow would be unlikely to contain SO_2, NO_x or ethylene. H_2S is a likely contaminant which must be removed, something that most but not all biogas upgrading facilities currently do. As CO_2 demand fluctuates throughout the year but biomethane production is constant, a buffer would be required. In this example, 1.08 kt CO_2 is produced per month. Data from the KASPRO model (F. De Zwart, 1996), run for a tomato greenhouse, gives the difference in peak demand in the summer compared to the winter.

Applying this pattern to the annual 13 kt, starting in October from an empty buffer, shows that a peak capacity of roughly 3.5 kt storage would be required.
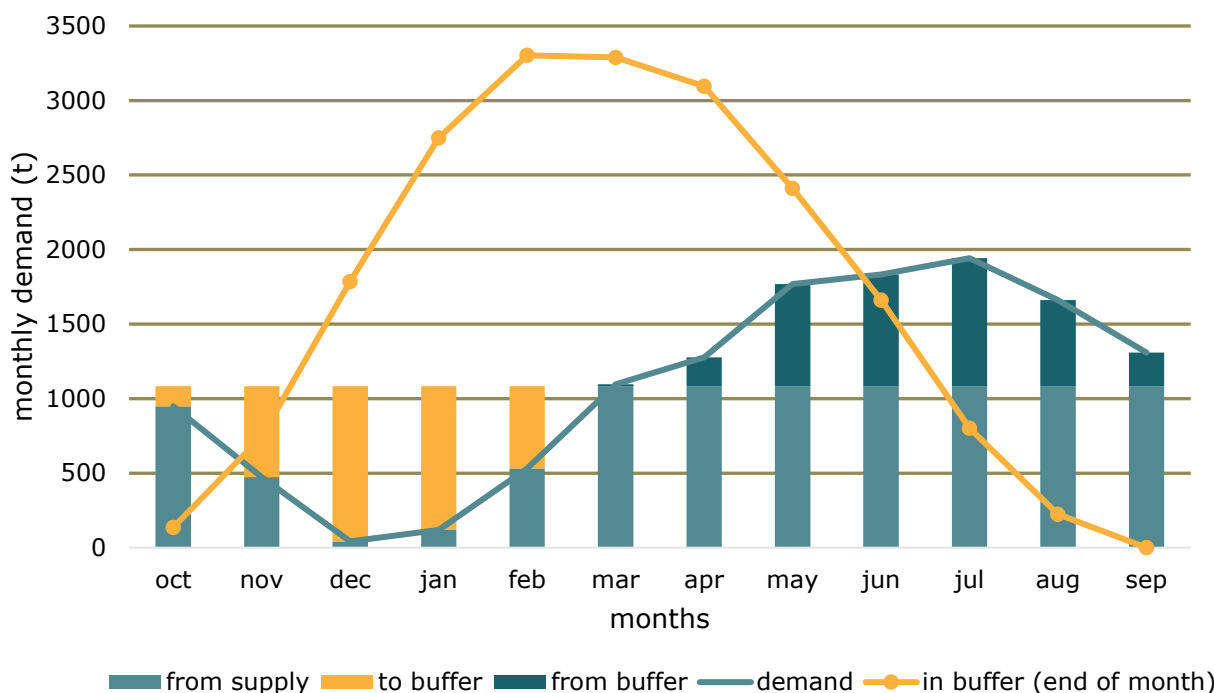
Figure 10 *A visualisation of CO_2 demand over the year, starting in October, the first month where CO_2 is stored in the buffer, with an annual supply of 13 kt evenly spread over 12 months.*

More information (in Dutch) on the potential of a cross-over between pig husbandry and greenhouse horticulture and other cross-overs, for CO_2 but also nutrients and other flows, can be found in the white papers at wur.nl/circulaire-glastuinbouw.

6.2.3   Lime-Based Adsorption

Lime-based materials are able to absorb CO_2, allowing for purification, concentration and storage. A system using lime pellets to do this has been developed by Hot Lime Labs, a company based in New Zealand. The system starts by partially oxidising biomass into a CO_2-rich reducing flue gas. These gases are sent through a high-temperature chamber containing lime pellets which absorb the CO_2 and let all other gases through, thereby purifying the CO_2. When the CO_2 is needed in the greenhouse, it is released from the pellets by blowing air over them. The CO_2 leaves the pellet chamber at a 15-25% concentration (by volume), with the remainder being nitrogen.

To supply a approximately 3.5 t CO_2 per day, or 438 kg h^{-1} for 8 hours, the system uses 2.5 t of dry biomass (10-15 m^3) of biomass daily, in the form of woodchips, residual wood or green waste. This amount of feedstock also produces 8 MWh of heat, which can be delivered in the form of hot water or hot air, at 90-150 °C for either immediate use or to be sent to a heat buffer. A single Hot Lime system supplying 3.5 t CO_2 per day requires an area of around 400 m^2. In order to meet the demand for larger glasshouses, Hot Lime Labs installs multiple containerised systems while combining the feedstock processing section. Whilst such a system ignores the principles of cascading by directly oxidising biomass, it may be useful in rural areas where access to CO_2 sources is limited but biomass is plentiful. However, to not contribute to net increases of CO_2 in the atmosphere, the biomass should come from the long carbon cycle. Therefore, if woodchips must be used, the trees from which they come must be replanted.

7 Conclusion

This report has examined fossil-free CO_2 supplementation for greenhouse horticulture. The main findings are summarised in this section, for each of the questions set out in the beginning.

1. **Quantity**   How much CO_2 does the greenhouse horticulture sector need and how much is available?

There is expected to be enough CO_2 in 2030 to cover the projected demand from the sector, which is between 1.8 and 3.0 Mt per year. Although there are significant differences in concentration and purity of these sources, biogas upgrading and ethanol fermentation together would already be able to supply the whole sector.

2. Quality in terms of **concentration**   How concentrated does the CO_2 need to be, how concentrated are most sources, and how can the desired concentration be achieved?

In theory, with typical ventilation rates, anything above 4% CO_2 could already be useful for greenhouse horticulture, though the lower the concentration, the higher the transport and storage costs. Biogas upgrading and ethanol fermentation produce flows of at least 99% CO_2, whereas others are below 10%.
For concentrating CO_2 sources, a combination of membrane filtration and cryogenic distillation seems to be the most feasible approach. Using synthetic fuels such as formic acid may result in a concentrated stream more practical to transport than gaseous CO_2, but involve large amounts of energy and lead to waste, since CO_2 release is coupled with energy release, as is the case with CHPs and boilers. Low-concentration direct air capture (DAC), as opposed to DAC producing pure CO_2, is in theory feasible with typical greenhouse ventilation rates, though current research shows it to be less practical due to the inconveniences of dealing with larger volumes of air.

3. Quality in terms of **purity**   Which contaminants are common, need to be removed, and how can they be removed?

Biogas upgrading and ethanol fermentation produce by far the purest CO_2. As for other sources, CO_2 from composting and biomethane combustion is likely to come with harmful contaminants, however no more than from burning natural gas, for which growers already have the capacity to remove contaminants (if done on-site). Biomass combustion produces by far the most contaminants. This report outlined the benefits and drawbacks of several purification technologies for the three most common contaminants, none of which are without their own drawbacks, though wet scrubbing and adsorption seem to be the most effective.

4. **Distribution**   Which options are available for distribution?

A challenge in the distribution of non-fossil CO_2 is the fact that many sources are spread out and produce relatively small amounts. For such cases, bottled CO_2 is expected to be the best solution. Pipelines require a higher investment but have a lower variable cost, and it is feasible to convert hydrocarbon pipelines to supply CO_2, as was done for OCAP, even if this pipeline had only a specific use and so could more easily be replaced. CO_2 buffers are likely to play an important role in helping supply match grower demand. For a constant supply, a buffer able to store 30% of the annual demand should suffice. Lime-based absorption is a novel a way to store CO_2 and is described in this report.

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