Carbon dioxide releases from wastewater treatment: potential use in the UK

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Power consumption by the UK water industry has increased as a result of the introduction of new quality standards; the annual (2008/2009) carbon dioxide output was reported at 5.1 Mt. Biogenic output of carbon dioxide for the sector was calculated to be about 2 Mt. The strategies available to the water industry for reducing carbon footprint are increased use of renewable energy, principally anaerobic digestion, using less power and methods for reducing carbon dioxide emissions. This paper reports on work sponsored by UK Water Industry Research to examine methods for capturing and utilising carbon dioxide from wastewater treatment. The review has concluded that bioconversion and biofixation using algae and hydrogenotrophic methanogenesis are the most promising methods for utilising carbon dioxide. These technologies would readily integrate into existing industry flow sheets and both increase biogas production and reduce carbon dioxide emissions.

1. Introduction

The carbon dioxide (CO₂) produced during wastewater treatment (WWT) is defined as biogenic and is not counted as emissions under the current carbon accounting rules. There are, therefore, no mandatory greenhouse gas (GHG) targets for the water industry, but the English and Welsh water utilities are government supervised and regulated on price, investment, service levels, water quality and environmental standards (OFWAT, 2006). In the future it is anticipated that these regulatory mechanisms will be used to direct investment into reducing carbon dioxide emissions by the water utilities. The government’s strategy document, *Future Water* (DEFRA, 2008) set out a vision for the sustainable delivery of secure water supplies with an improved and protected environment. This document highlighted the key role to be played by the water sector in mitigating climate change and these environmental, corporate and social responsibilities have led many water companies to publish carbon reduction targets in their strategic plans.

The water industry represents about 1% of the UK’s current carbon dioxide emissions, but this could rise in response to the government’s carbon reduction commitment. Water UK has published annual sustainability indicators since 2007 and these are summarised in Figure 1. The data show the water industry has become more energy intensive in order to meet the improvements in water quality. New technology for nutrient and persistent pollutant removal, such as activated carbon, membranes and activated sludge, are large energy users. In 2009 there were 5.07 Mt of GHG emissions as carbon dioxide equivalent (Water UK, 2009). In 2009 about 60% of the water utility running costs were for power and rising disproportionately compared to other costs (UKWIR, 2009). It was reported that WWT accounted for 56% and water supply 39%, with administration and transport contributing the remaining 5% of the total GHG emissions. Pumping was the major power use in water supply, with aeration and pumping the main consumption in WWT.

The water industry is also a large generator of renewable energy, with anaerobic digestion (AD) being the most common sewage sludge treatment in the UK (Table 1). The UK water industry AD facilities account for 4% of the UK total annual...

Table 1. Digester and power generation 2005 (adapted from Rogalla et al. (2008))
electrical renewable energy generation (DECC, 2011). The generation of methane (CH\textsubscript{4}) as a by-product and the potential fertilizer value of digestate makes AD an attractive waste treatment process for most organic wastes. It has been estimated that there are now about 1000 anaerobic digesters for food effluents in Europe and 5000 for sewage sludge (Franklin, 2001). In the USA there are 650 sewage sludge digesters, 106 of which had gas utilisation (Kalogo and Montieth, 2008). The European Commission has reported that the AD of domestic waste (sewage sludge and the organic fraction of municipal solid waste) could in theory provide 3% of Europe’s energy needs (Tilche and Galatola, 2008) and the UK government has announced a strategy to encourage the uptake of AD as a source of renewable energy (DEFRA, 2011).

Increased use of AD is an opportunity for the water industry because of their experience and integration within existing WWT. Co-digestion with other organic wastes, for example, may have been inadvertently discouraged by the regulatory framework (OFT, 2011).

Calculations and estimates are presented in this paper on the proportions and quantities of carbon dioxide produced during WWT in relation to the available techniques and potential for capturing, separating and utilising this carbon dioxide.

2. Estimates of carbon dioxide from AD biogas

Biogas produced in AD plants is primarily composed of methane and carbon dioxide, with smaller amounts of hydrogen sulfide (H\textsubscript{2}S) and ammonia (NH\textsubscript{3}). Trace amounts of hydrogen (H\textsubscript{2}), nitrogen (N\textsubscript{2}), carbon monoxide (CO), volatile organic compounds (VOCs) and oxygen (O\textsubscript{2}) are present in the biogas. Usually, the biogas is saturated with water vapour and may contain dust particles and siloxanes.

The Water UK statistics for 2008/2009 (Water UK, 2009) reported a total sludge production as 1·76 Mt as dry solids. It has been reported that 66% of this sludge was processed through AD (UKWIR, 2009). If it is assumed that the sludge had volatile solids (VS) content of 75% and that an average of 45% of the VS was converted during single-stage AD, then this yields 342 million m\textsuperscript{3} of biogas. Barber (2009) estimated that 60% of this biogas was utilised in combined heat and power (CHP). Using an assumed biogas composition of 65% v/v methane and 35% v/v carbon dioxide, and their molar ratios, gives the mass composition of dry biogas at standard temperature and pressure as 464·4 g methane and 687·5 g carbon dioxide/m\textsuperscript{3}, or 40·3% methane and 59·7% carbon dioxide respectively. The annual biogas production (342 million m\textsuperscript{3}) therefore contains 0·27 Mt of carbon dioxide. This is an underestimate because of fugitive losses through roof seals and open secondary digesters, calculated to be around 11% of biogas production by Barber (2009).

2.1 Post-combustion carbon dioxide from biogas

The stiochiometry of methane combustion in either CHP or flaring results in 2·75 kg of carbon dioxide for each kg of methane used. If trace contaminants like hydrogen sulfide, ammonia and siloxanes are ignored or removed then the products of combustion can be assumed to be carbon dioxide, water and oxides of nitrogen (NO\textsubscript{x}) (from atmospheric nitrogen). Thus, from the annual total biogas production noted above and using the mole balance for combustion, the annual quantity of carbon dioxide that could potentially be captured and separated from the flue gases would be 0·5 Mt carbon dioxide/year. Flue gases from combustion plants typically contain between 8 and 15% carbon dioxide v/v (Song, 2006; Wang et al., 2008).

2.2 Carbon dioxide emissions from sludge incineration

Around 13·6% of the total sludge production is incinerated with energy recovery (Water UK, 2009). The calorific value favours raw sludge, although there are examples of the incineration of digested sludge. Using a conversion factor of 1·95 kg CO\textsubscript{2}/kg biomass, based on the assumption that the VS content of the sludge is biomass with an empirical equation of C\textsubscript{12}H\textsubscript{22}O\textsubscript{11}N yields an estimated carbon dioxide emission of 0·26 Mt/year.

3. Carbon dioxide emissions from aerobic treatment

Aerobic WWT releases metabolic carbon dioxide, methane and nitrous oxide (N\textsubscript{2}O) in addition to the carbon dioxide emitted from electrical generation for pumping and aeration systems. The different types of aerobic treatment processes used could be simplified as variants of either suspended or attached growth systems, typified by the activated sludge process and biological filters. Cakir and Stenstrom (2005) derived a simple model and equations to estimate the carbon dioxide emissions from aerobic treatment processes and these were used here to calculate emissions (the calculations can be reviewed as supplementary data). The model was based on stoichiometric equations to describe microbial growth, biomass decay and oxygen utilisation. The model indicates a net release of 0·95 kg carbon dioxide per kg ultimate biochemical oxygen demand (BOD\textsubscript{u}) treated for carbon respiration (including endogenous respiration) and 0·67 kg carbon dioxide per kg BOD\textsubscript{u} with energy recovery from denitrification. This latter figure has been converted to a unit emission of carbon dioxide for secondary treatment per population equivalent (p.e.), giving a value of 0·05 kg CO\textsubscript{2}/p.e. day. Using values for the sewered population of England and Wales as 54 million and that for the UK as 60 million gives an annual estimate for carbon dioxide emissions from aerobic WWT in the range 1·1–1·1 Mt carbon dioxide/year. This is three to four times the quantity of the carbon dioxide from anaerobic biogas, but the concentration of carbon dioxide in the aerobic off-gases is much lower. The carbon...
dioxide concentration from a model diffused air activated sludge plant in the air flow as it leaves the surface of the aeration tank was estimated to be 0.8% v/v. This concentration is around 40 times lower than the concentration of carbon dioxide in biogas and therefore more energy would be needed in processing the off-gases from aerobic treatment to separate and capture the carbon dioxide. Moreover, the aerobic treatment stage would also need to be enclosed and, although there are a number of examples of enclosed plant, this is expected to double costs.

4. Total biogas and wastewater biogenic carbon dioxide emissions

Figure 2 summarises these calculations and shows that the total biogenic carbon dioxide is about 40% of the carbon dioxide emitted from grid electricity generation used by the sector. This analysis points to the overriding benefits of improving fine solids capture in primary settlement and increasing the use of anoxic and anaerobic zones to remove carbon.

The proportion of carbon dioxide from biogas and its use was estimated at 15% (5.3% + 9.9%) of the fossil-fuel-derived carbon dioxide produced by the sector. The higher concentrations of carbon dioxide in biogas, however, make it the most accessible and potentially usable source of carbon dioxide.

5. Techniques for separating and capturing carbon dioxide

For convenience, separation technologies can be divided into physical, chemical or biological methods. Examples include cryogenic, membrane, high-pressure water scrubbing, amine and alkaline absorption and biofixation. Work reported by de Hullu et al. (2008) reviews most of the available biogas purification technologies, which are summarised in Table 2.

![Figure 2. Relative magnitude of carbon dioxide emissions from WWT compared to those from power consumption for the sector](image)

<table>
<thead>
<tr>
<th>Technology</th>
<th>Yield: %</th>
<th>Purity: %</th>
<th>Energy demand: kWh/N m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cryogenic</td>
<td>99</td>
<td>99</td>
<td>1.64</td>
</tr>
<tr>
<td>Membrane</td>
<td>78</td>
<td>89</td>
<td>0.28</td>
</tr>
<tr>
<td>Chemical absorption</td>
<td>90</td>
<td>99</td>
<td>0.19</td>
</tr>
<tr>
<td>High-pressure water scrubbing</td>
<td>94</td>
<td>97</td>
<td>0.49</td>
</tr>
<tr>
<td>Pressure swing adsorption</td>
<td>91</td>
<td>98</td>
<td>0.28</td>
</tr>
<tr>
<td>MT BioMethan (absorption)</td>
<td>99</td>
<td>99</td>
<td>0.11</td>
</tr>
</tbody>
</table>

Data sources de Hullu et al. (2008), Johannson (2008).

Table 2. Expected methane quality (yield and purity) with estimated energy demand

The table is based on three independent sources but represents an estimate, and more information will emerge as more operating data are generated – such as from the Thames Water Didcot (2010) and Severn Trent Minworth WWT projects to produce grid quality gas from sewage sludge and liquefied gas from landfills reported by Gasrec at Albury. Table 2 summarises the expected output of methane in terms of yield and purity along with estimates for the energy demand for some of the commonly used systems.

In order to reuse the carbon dioxide off site, compression and on-site storage would be required, increasing the costs and energy use compared to an on-site application. Therefore, the proposed use of carbon dioxide would affect the choice of technology. The estimated potential value of ‘raw’ carbon dioxide reclaimed from wastewater was between £0 and £9/tonne, whereas the cost of energy for collecting, separating and storage was calculated to be £40/tonne. The calorific value of the biogas is dependent on its quality and the performance of CHP will be improved by treatment to reduce the carbon dioxide content. CHP using internal combustion engines is the most common use of anaerobic biogas, but there is the prospect of other more valuable uses if the gas quality can be improved. These include other engines (turbines and fuel cells), as a transport fuel and to add to the gas grid.

6. Potential uses for carbon dioxide

Song (2006) has reviewed the technologies available for the catalytic conversion of carbon dioxide into other energy sources and chemicals. Carbon dioxide is used in a wide range of processes and Table 3 summarises these, adapted from Song (2006) and Aresta and Dibenedetto (2007). Supercritical carbon dioxide also has benefits as a solvent, especially for
food production, because of its low toxicity and lower GHG potential compared with other solvents.

Carbon dioxide is relatively stable with a large negative value for the Gibbs free energy of formation (−394 kJ/mole) compared to other simple carbon compounds such as methane, methanol and acetic acid (Lide, 2001). Therefore, most chemicals produced from carbon dioxide would require energy input, which would have to be sourced from a renewable or non-carbon-producing energy to avoid or minimise net carbon dioxide production.

A few compounds like the inorganic carbonates and organic dimethyl carbonate have lower (more negative) free energies of formation and the formation of these compounds from carbon dioxide would release energy. Improved catalysis could reduce these energy requirements; for example, photochemical reduction of carbon dioxide has been suggested. Aresta and Dibenedetto (2007) reported on developments using transition metal complexes to reduce carbon dioxide with water to form acetate, formate, methanol and methane. The irradiation of titanium and strontium complexes with sunlight has also been used to reduce carbon dioxide to produce acetate, formate and
methanol. The yield from the photochemical conversion of carbon dioxide using these catalysts is still too low for commercial exploitation but is likely to be cheaper in future as research in this area progresses.

Thus although many of the uses for carbon dioxide shown in Table 3 are technically feasible, the overall energy, balances, economics and practical application within the relatively small scale of a WWT will make them uneconomic. For example, using the carbon dioxide balance from the calculations suggested earlier, a 500 000 population equivalent WWT plant would produce a total of around 26 t carbon dioxide per day from biogas, but a large coal-fired power station produces around 25 000 t of carbon dioxide per day.

7. Utilising carbon dioxide within WWT

An alternative to uneconomic chemical conversions of carbon dioxide would be to enhance biomass production at WWT plants for energy or other products. The potential of algae for waste treatment has been recognised for a long time; Oswald and Golueke (1960), for example, used a high-rate algal pond for carbon dioxide removal from power plant emissions. Toms et al. (1975) reported on the performance of algal ponds to remove nutrients from UK sewage effluents. The recent research on algal bioreactors has been for biomass/biofuel production from carbon dioxide. Schenk et al. (2008) reported that bioreactors designed for biomass production could achieve 5–30 times the yields of crop plants. Calculations and experiments by Chisti (2008) and Pulz and Gross (2004) indicated that algae could be grown at up to 150–300 t/ha per year.

The relatively high lipid, starch and protein content and the absence of lignin make algae an ideal candidate for efficient biofuel and biomethane production in AD plants (Sialve et al., 2009). Figure 3 shows the inputs necessary for algal growth and their potential products.

The environmental and economic costs for supplying nutrients and carbon dioxide have been reported as the most significant constraints to the economics and carbon balance of algae bioreactors (Clarens et al. (2010) and Benneman and Oswald, cited in Clarens et al. (2010)). Aresta et al. (2005) modelled algal bioreactors and concluded that the overall life cycle burden and economics of the process favoured co-location with wastewater nutrients. The model suggested that there was a net energy benefit from carbon dioxide fixation across the waste-water process but there could also be net atmospheric carbon dioxide reductions from further fossil fuel substitution.

Wang et al. (2008) and Park and Craggs (2011) report on case studies using simple algal lagoons to remove nutrients from final sewage effluent or works return liquors, with carbon dioxide from on-site CHP and/or AD gases. High-performance photobioreactor designs have also been developed for algal cultivation and high-value products, such as pharmaceuticals and food additives (Chisti, 2008). An example of this type of optimised photobioreactor design was proposed by Doucha and Livanský (2006, 2009); it was based on the principle of forming a thin, slurry layer, in open air culture on a series of inclined plates. This arrangement allowed a thick, moving layer (6–8 mm) of microalgae to develop and reach a very high algal density at harvest (~4% d.s.). One of the Doucha and Livanský (2006) test sites was at a similar latitude (37˚N) to results reported by Park and Craggs (2011) from conventional algal ponds for tertiary treatment (37˚S with 4-day hydraulic retention time (HRT)), who in contrast achieved an average 275 mg/l algal concentration. Both groups of authors present data which show algal growth rate was proportional to sunlight intensity. The best summer conditions reported by Doucha and Livanský were a daily total of 12–4 MJ/m² compared to Park and Craggs, whose best summer conditions were 23 MJ/m². Some of these differences in light intensity may be explained from the different years studied, cloud cover or

Figure 3. Potential products from algal bioreactor
light measurement technique. Both reactors used carbon dioxide enrichment (70–80% utilisation in both cases) to increase productivity; Doucha and Livanský report average daily reactor temperatures of 30°C from 10 h of sunlight and Park and Craggs 23°C from 7.5 h of sunlight. The Doucha and Lavinsky reactor gave a growth rate of 1.7 g dry weight (DW) algae per gramme of carbon dioxide utilised equivalent to 32.7 g DW/m² per day; Park and Craggs (2011) reported 20.7 g DW/m² per day in summer from carbon dioxide additions used to control pH to < 8.5. Based on data supplied by Doucha and Livanský (2006) it has been calculated that a plate photobioreactor of around 200 m² would be required to process 1 m³/h of carbon dioxide.

Figure 4 is a conceptual flow chart for the algal utilisation of carbon dioxide within a WWT plant.

Based on this work the predicted energy balance for a 100 000 p.e. WWT plant, with an integrated photobioreactor and processing the algal biomass through AD is shown in Figure 5.

By converting the carbon dioxide content of biogas from an AD plant to algal biomass and then recycling this biomass back to the AD plant, the additional daily methane production was predicted to be 470 m³/day with a gross energy yield of 4656 kWh for the 100 000 p.e. plant. Conversion of this gross energy value to electrical energy in a CHP assuming 35% efficiency yields 1629.6 kWh. This is based on an 18% increase in the methane production (27% increase in biogas) and the conversion of 1.3 t of carbon dioxide per day (70% of the carbon dioxide content in biogas) to biomass.

The energy used for capturing and separating carbon dioxide from the biogas and that used for pumping and lighting (nominal 12 h per day) is shown in Figure 5 and these assumptions suggest the positive energy balance is 195 kWh/t of carbon dioxide converted to algal biomass. The lighting was based upon low-energy light-emitting diode (LED) lighting arrays and the gas separation system based upon the chemical absorption. Other assumptions were that there would be a sufficient supply of nutrients (nitrogen and phosphorus) in the

Figure 4. Nutrient and biomass flows through a conceptual biofixation system.

Figure 5. Energy balance showing the additional energy production from the algal biomass growth system following AD for a 100 000 p.e. WWT.
final effluent or sludge return liquors to support this added biomass growth and that the algae did not affect the biodegradability or disposal of the domestic sludge. Sialve et al. (2009) reviewed the co-digestion of algae with both industrial waste and sewage sludge and reported benefits in biogas yields from the mixtures. Sialve et al. (2009) and Cecchi et al. (1996) reported experiments on the digestion of marine algae and there were no adverse effects up to 30% additions at a HRT of 15 days. These calculations and assumptions need more detailed analysis and demonstration.

8. **Hydrogenotrophic methanogenesis**

Autotrophic methanogenic bacteria utilise hydrogen during the anaerobic reduction of carbon dioxide to methane (Alvarado et al., 2005; Zhang et al., 2008). In almost all methanogenic environments, hydrogen is utilised rapidly even when present at very low concentrations.

Alimahmoodi and Mulligan (2008) demonstrated an increased biogas yield by utilising dissolved carbon dioxide with volatile fatty acids as a source of hydrogen. They used a laboratory-scale upflow anaerobic sludge blanket (UASB) reactor operating at 35°C and showed through mass balance studies that the rate of methane production was approximately doubled following the introduction of carbon dioxide at a fixed organic loading rate. The work by Alimahmoodi and Mulligan (2008) used a separate side stream of volatile fatty acids as additional readily biodegradable carbon (rbCOD). Research on the performance of hydrogenotrophic methanogenesis in conventional municipal digestion was reported by Sato and Ochi (1994). The experimental results from a pilot scale trial by Sato and Ochi (1994) indicated a maximal increase in the methane yield of 30% limited by the fall in pH.

The enriched carbon dioxide stream could be injected into an existing digester. This could utilise either the gas recompression mixing system or separate injection as part of an advanced pretreatment stage. Such a scheme could be incorporated into the standard AD process with relatively few changes, as shown in Figure 6.

![Figure 6. Conceptual route for bioconversion of carbon dioxide using hydrogenotrophic methanogenesis](image)

![Figure 7. Graph of potential energy yields from hydrogenotrophic methanogenesis](image)
Figure 7 shows the equivalent energy balance for hydrogenotrophic methanogenesis as Figure 6 for a 100 000 p.e. works but has had to assume a 50% conversion efficiency because there are no experimental figures in the literature. Alternatively, the carbon dioxide extracted from biogas could be delivered to a side-stream UASB reactor with volatile fatty acids enriched feed from sludge treatment/thickening liquors.

9. Other biomass

There are other autotrophic bacteria already used in WWT which utilise carbon dioxide and offer future possibilities, such as a group of ammonia oxidising bacteria recently characterised (Ahn et al., 2011).

Carbon dioxide is also widely used to enrich the atmosphere of glasshouses to increase crop yields. There is at least one commercial example of using AD gases (Pearson and Sons, Alderley Edge) which is reported to use heat and carbon dioxide from an AD system to enhance tomato production in some of their glasshouses.

At smaller wastewater plants where land is available it may be possible to increase biomass production for AD using polytunnels. Aquatic plants would cope better with lower winter light and temperature than algae but their productivity would be lower. Growing sacrificial biomass on contaminated land for AD is also used (Monnet et al., 2002).

10. Conclusions and recommendations

The biogenic output was found to be equivalent to about 40% of the total carbon dioxide emissions produced by the sector from the use of grid electricity for water supply and wastewater services. The most concentrated and amenable source of carbon dioxide for utilisation or mitigation was biogas, which together with the post-combustion carbon dioxide was equivalent to around 15% of the total carbon dioxide emissions (including fossil fuels). The survey in common with others suggested the use of AD within the industry would increase with a commensurate increase in this biogas-derived carbon dioxide.

The most promising routes for any future utilisation of carbon dioxide were the on-site biological routes. These were the hydrogenotrophic conversion of carbon dioxide to methane within the existing AD process and the biofixation of carbon dioxide in algal photobioreactors.

These biological conversions were also considered to be the most practical routes for utilising carbon dioxide because they have other benefits, such as nutrient removal, improved CHP performance and extra energy. They are innovative and emphasise energy balance, and for some utilities should integrate readily into the AD and nutrient removal processes.

The hydrogenotrophic route needs to be demonstrated at full scale, which should be simple since relatively small additions to existing plant would be needed. There are already several demonstrations of the algal biomass route, but the detailed cost benefits and potential impact on digestibility or digestate are not yet available. There is potential for further increases in biomass yields from artificially illuminated bioreactors. This needs more work using standardised measurements of light intensity.

11. Practical relevance

Carbon dioxide emissions have become a major environmental concern. This paper analyses and discusses controlling and reusing carbon dioxide during WWT by either recovery of carbon dioxide or sequestration as organic matter. It has concluded that conversion to organic matter and therefore biofuel or biogas is the most promising route. Demonstrations of the alternatives are reviewed.

An inference from the work was that nitrogen emissions also need to be examined, as good data could not be found on the analysis of off-gas from either activated sludge or biogas.

REFERENCES


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