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Bioprocesses for the treatment and valorisation of gas emissions, odours, volatile compounds and greenhouse gases

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Different bioprocesses for the treatment as well as the valorisation of waste gases and greenhouse gases are briefly reviewed. Biotreatment technologies are based on the use of bioreactors for waste gas treatment, e.g., biofilter, biotrickling filter, bioscrubber, suspended growth bioreactor with gas diffusion. Aspects related to waste gas (bio)valorisation address the potential of aerobic knallgas bacteria (e.g., *C. necator*), microalgae, as well as anaerobic acetogenic bacteria aiming at obtaining a range of biofuels and bioproducts mainly through bioconversion of one carbon gases (e.g., CO_2, CO). Valorisation of waste gases appears to be a promising innovative, cost-effective, sustainable alternative to conventional treatment technologies.

KEYWORDS

acetogenic bacteria, biofuels, bioproducts, carbon dioxide, knallgas bacteria, microalgae

1 Introduction

Solid waste, wastewater and volatile gas pollutants are the main sources of pollution on Earth (Kennes, 2023). Particularly, pollution related to emissions of volatile compounds and greenhouse gases to the atmosphere has become a major concern in modern societies. Their origin can be either mobile sources (e.g., vehicles) or stationary sources such as direct industrial emissions but also emissions from waste and wastewater treatment plants. In those plants, major volatile pollutants are odours as well as greenhouse gases. Common odours and other gases often emitted from wastewater treatment plants are hydrogen sulphide (H_2S) as well as other sulphur compounds (e.g., mercaptans), ammonia (NH_3), and also different Volatile Organic Compounds (VOCs). Odour nuisance and the presence of toxic and hazardous volatile compounds can affect surrounding areas as well as plant operators. In order to quantify the level of contamination, besides using units such as mg/m^3, odour concentration can also commonly be quantified in terms of Odour Units (OU), i.e., OU/m^3 (Burgués et al., 2022). On the other side, typical greenhouse gases found at wastewater treatment plants, mostly related to biological treatment processes, are carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O).

Odours detected at solid waste treatment plants are highly dependant on the type of wastes, their level of degradation and the operating conditions and it includes compounds such as sulfides, nitrogen compounds, volatile fatty acids, aldehydes or ketones, among others (Bruno et al., 2007). They mainly result from the degradation or biodegradation of waste materials. Greenhouse gases reported to be found at waste management plants, e.g., landfills, are similar to those reported for wastewater treatment facilities, i.e., carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), mainly (Gentil et al., 2009).

Finally, the nature of volatile compounds emitted from different process industries is quite broad considering the large variety of manufacturing activities, but, in industrialised countries, somewhat more than 50% of total emissions of Volatile Organic Compounds from any possible source corresponds to non-combustion processes at industrial sites (Kennes and Veiga, 2013).

Different alternatives can be considered to address the mitigation of odours, greenhouse gases and other volatile compounds, which include both non-biological as well as biological processes relying on the degradation of pollutants or their mass transfer from the gas phase to another phase. On the other side, rather than degrading or merely removing those pollutants, it is also possible to (bio)convert them into new, valuable products, which is an innovative, current, area of research with significant recent developments (Kennes, 2025). Bioprocesses used for such purpose are described in some of the next sections.

2 Gas (bio)treatment technologies

Conventional methods for odour and gas treatment, at waste and wastewater treatment plants as well as other industrial sites are physico-chemical processes, mainly absorption and adsorption. However, the latter are often more costly than bioprocesses, which may then become the best choice whenever feasible and when dealing with biodegradable pollutants. Many volatile compounds are indeed, to some extent, biodegradable in many cases (Kennes and Veiga, 2013).

The most common bioreactors used for waste gas treatment are the biofilter, biotrickling filter, bioscrubber and suspended-growth bioreactor (e.g., activated sludge type reactor) with gas diffusion, described in more detail elsewhere (Kennes and Veiga, 2013). They are mostly used to treat polluted air (containing oxygen), though they can also handle anaerobic gases. In that sense organic pollutants containing carbon atoms and often also hydrogen will produce some carbon dioxide and water in most cases. The stoichiometric equation generating CO_2 and H_2O (and biomass, not accounted for in the reaction below) from organic volatile pollutants is illustrated hereafter for toluene, as representative example:

$$C_7H_8 + 9\,O_2 \rightarrow 7CO_2 + 4H_2O$$

If besides C and H atoms, the pollutant is halogenated, HCl would then be produced. The reaction for a gas molecule containing substituent R and a chlorine atom would then be:

$$R - Cl + O_2 \rightarrow CO_2 + H_2O + HCl$$

Instead, the most common inorganic gas pollutants do not contain carbon atoms. In such case, they will then not release carbon dioxide as end-product. The most typical ones are nitrogen-compounds such as ammonia (NH_3) and sulphur-compounds such as hydrogen sulphide (H_2S) as shown in the equations below. Hydrogen sulphide is one of the most common odourous compounds; it is first converted to elemental sulphur and, if enough oxygen is present, it will then subsequently generate sulphuric acid. This will then result in acidification of the reaction medium, unless acid production is neutralised.

$$H_2S + 0.5\,O_2 \rightarrow S^0 + H_2O$$

$$S^0 + H_2O + 1.5\,O_2 \rightarrow H_2SO_4$$

Similarly as for organic compounds, biodegradation reactions for inorganic pollutants do most often simultaneously produce some biomass which can then also be included in the stoichiometric reaction if needed.

Besides biodegradation processes, another more recent and innovative trend consists in converting (i.e., bioconversion) volatile pollutants into higher value products, allowing both to solve an environmental issue but also generating some economic benefit. This is overviewed below for greenhouse gases such as CO_2 and CH_4.

3 Gas and greenhouse gas valorisation through bioprocesses

In order to address air pollution control, the treatment of gaseous emissions, i.e., odours, greenhouse gases or other volatile compounds, has been the common objective for many decades. However, recent research has shown that, besides their treatment, waste, wastewater and gas valorisation is another possible, viable and highly attractive, alternative to their mere treatment (Kennes, 2023). Then, pollutants are not only removed but they are simultaneously converted into high-value commercial products. Hereafter, two examples of greenhouse gas valorisation are provided, i.e., for carbon dioxide (CO_2) and for methane (CH_4), based on their bioconversion to useful products.

3.1 Biological carbon dioxide valorisation

Different alternatives are available for the biological conversion of CO_2-containing gases. This manuscript presents some of the most common possible bioprocesses being considered nowadays, with somewhat deeper focus on acetogenic (anaerobic) fermentation of such C_1-gases (i.e., gases containing one Carbon atom), of which either CO_2 or mixtures of CO and CO_2 are typical. Besides acetogenic CO_2-metabolizing bacteria, other (micro)-organisms are also able to use carbon dioxide and mitigate their emissions. This is the case of aerobic bacteria such as *Cupriavidus necator* (Knallgas bacterium) or also microalgae.

3.1.1 Knallgas bacteria

Cupriavidus necator grows on CO_2 as carbon source and it simultaneously needs hydrogen as energy source as well as oxygen as electron acceptor in the gas mixture, with strain H16 being the most studied one. Carbon dioxide is assimilated through the Calvin-Benson-Bassham (CBB) pathway (Figure 1). Different optimal $H_2/O_2/CO_2$ ratios have been reported, but the best values are often claimed to be around 7/2/1 (Amer and Kim, 2023). Anaerobic respiration with reduction of nitrate and full denitrification down to molecular nitrogen (N_2) is also possible. Additionally, this

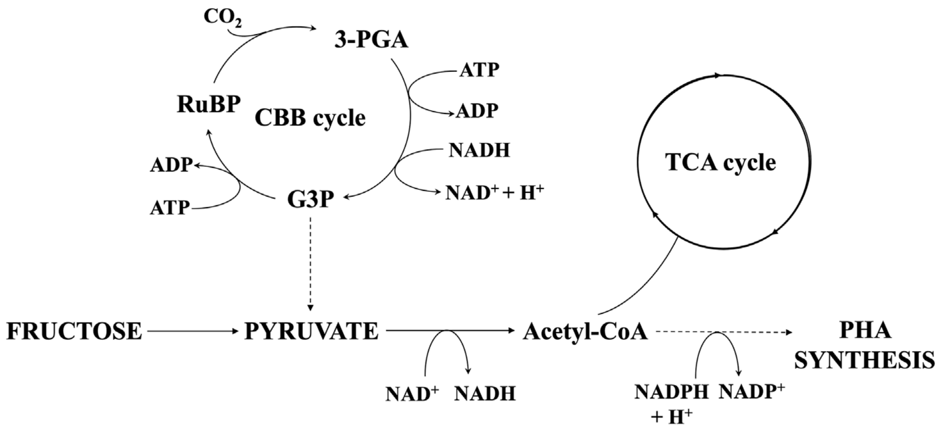

FIGURE 1
Catabolic pathway for fructose and CO_2 metabolism in *C. necator*. Abbreviations: 3-PGA, 3-phosphoglycerate; ADP, Adenosine diphosphate; ATP, Adenosine triphosphate; CBB cycle, Calvin-Benson-Bassham cycle; G3P, Glyceraldehyde 3-phosphate; NAD^+, Nicotinamide adenine dinucleotide (oxidised form); NADH, Nicotinamide adenine dinucleotide (reduced form); $NADP^+$, Nicotinamide adenine dinucleotide phosphate (oxidised form); NADPH, Nicotinamide adenine dinucleotide phosphate (reduced form); PHA, Polyhydroxyalkanoate; RuBP, Ribulose biphosphate; TCA cycle, Tricarboxylic acid cycle.

bacterium can also metabolise other substrates besides CO_2 and heterotrophic growth has been observed with different carbon sources such as fructose (Figure 1). Glucose is not metabolised by native strains of that species though. This organism has been renamed after being originally known as *Alcaligenes eutrophus*, *Hydrogenomonas eutropha* and *Ralstonia eutropha*. Because of the potentially explosive aerobic gas mixture, it is also known as knallgas bacterium. *Cupriavidus necator* has mainly been studied for its ability to accumulate Poly-Hydroxy-Alkanoates (PHA), i.e., biopolymers. In terms of biopolymer content, percentages as high as around 80% have been reached in that species in media with nutrient (e.g., nitrogen) limitation (Lambaue et al., 2023). Besides numerous studies with native strains, the organism has more recently been genetically modified and has also been used in microbial electrosynthesis (MES) systems in order to try to broaden its product spectrum. In that sense, a wide range of products have now recently been reported to be obtainable from *C. necator* cultivation, which include acetoin, 2,3-butanediol, isopropanol, *n*-butanol, iso-butanol, 3-methyl-1-butanol, fatty acids and derivatives, isoprene and terpenes, lycopene, methyl-ketones, alkanes, alkenes, α-humulene, 2-hydroxy isobutyric acid, trehalose, sucrose, lipochitooligosaccharides, among others (Pan et al., 2021). The potential application of such bacteria in full-scale processes is, however, still considered to have several limitations such as the reported low yields, high cost, or significant explosion issues, among others.

3.1.2 Microalgae

Microalgae are able to assimilate carbon dioxide through photosynthesis and use it as carbon source. Under phototrophic conditions, they would only need CO_2 and light as carbon and energy sources, respectively. Interestingly, photosynthetic effciency is often higher than what is commonly observed in plants. Microalgae biomass contains about 50% carbon and some studies claim that around 1.83 kg CO_2 can be fixed for every kilogram of biomass produced (Chisti, 2007). They allow to valorise carbon dioxide through its conversion into high value products. Some of first reports focussed mainly on their potential to accumulate lipids, which is then suitable to produce biofuels such as biodiesel and for which one of the most studied organisms belongs to *Chlorella* spp. (e.g., *Chlorella vulgaris*). In terms of biofuels, some microalgae are also rich in carbohydrates, though their levels are high in only some specific species, and it then represents an interesting feedstock for third generation bioethanol production after biomass harvesting, pretreatment/hydrolysis and subsequent sugar fermentation. Biogas is another interesting biofuel. In that sense, macroalgae (seaweed) are suitable for biogas production through anaerobic digestion of macroalgae feedstock. The same is possible with microalgae characterised by a high biomass productivity together with low ash content.

Additionally, the product spectrum of microalgae has been broadened over the years and they are now studied for their accumulation of compounds such as proteins, carotenoids, extracelular polymers and PHA. *Chlorella* species have been reported to be able to accumulate about 50%–60% proteins in the best cases, though some other species such *Spirulina* may even reach higher values of around 70%, containing all the essential amino acids. Microalgae have proportionally higher protein contents than most plants or crops.

3.1.3 Acetogenic bacteria

Acetogens are mainly comprising *Clostridium* species, although they also include some other ones such as *Acetobacterium woodii*, among others. Native organisms metabolise C_1-gases such as CO_2 and/or CO to produce acetic acid as their main end metabolite, where CO_2 is used as carbon source and hydrogen is needed as an energy source. However, some other products can also be found in some non-engineered strains, though not all of them, such as longer chain fatty acids, e.g., butyrate and hexanoate, ethanol, butanol, hexanol, 2,3-butanediol (Fernández-Naveira et al., 2017). Those metabolites are produced through the so-called Wood-Ljungdahl pathway (WLP) (Figure 2). In the Eastern branch of the WLP, CO_2 is

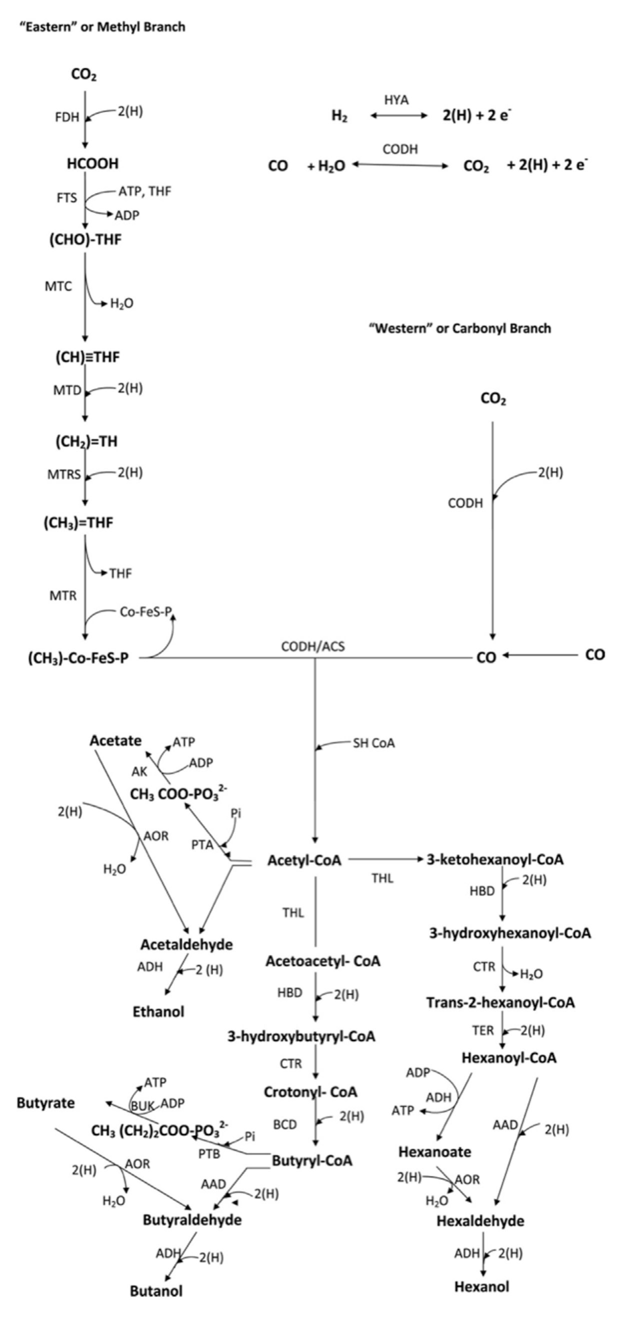

FIGURE 2
Wood-Ljungdhal pathway in acetogenic bacteria for the fermentation of C_1-gases (CO_2, CO) into fatty acids and alcohols in native bacteria [reproduced with permission from Fernández-Naveira et al. (2017)].

first reduced to formate to subsequently lead, stepwise, to acetyl-CoA. On the other side, in the Western branch, CO_2 can be converted into CO or, otherwise, carbon monoxide can be used directly as single substrate for the production of acetyl-CoA. Both in the Eastern as well as the Western branch, acetyl-CoA will then yield fatty acids (i.e., acetate, butyrate, or hexanoate) or alcohols (e.g., ethanol, butanol, hexanol). Although CO can be used as sole carbon and energy source, the presence of hydrogen is recommended, as CO is partly converted to CO_2 and thus subsequent removal of produced CO_2 would benefit from the availability of hydrogen as electron donor. These aspects are shown hereafter in some examples of reactions of bioconversion of CO and CO_2 into acetic acid (Fernández-Naveira et al., 2017).

$$4\,CO + 2\,H_2O \rightarrow CH_3COOH + 2\,CO_2$$

$$2\,CO_2 + 4\,H_2 \rightarrow CH_3COOH + 2\,H_2O$$

$$2\,CO + 2\,H_2 \rightarrow CH_3COOH$$

Fatty acids are the main end metabolites and are generally produced first, while alcohols are mostly obtained from the bioconversion of those fatty acids. Though the production of carboxylic acids is a commun feature of acetogens, only very few of them can produce alcohols and they include only a scarce number of identified species, i.e., mainly *Clostridium aceticum*, *Clostridium autoethanogenum*, *Clostridium carboxidivorans*, *Clostridium ljungdahlii*, *Clostridium ragsdalei* (Kennes-Veiga et al., 2024). Mixotrophic as well as heterotrophic growth is also possible and, similarly as with other bacteria (e.g., *C. necator*), growth under autotrophic conditions is generally more limited or lower than with organic carbon sources. The low solubility and slow mass transfer of C_1 gases and hydrogen in liquid (aqueous) phase contribute to such limitation in all autotrophic bacteria.

Besides the natural production of fatty acids and/or alcohols, more recent studies have been performed aiming at broadening the range of products that can be obtained from C_1 gases with acetogenic bacteria. Basically two approaches can be considered, consisting either in engineering bacteria in order to obtain other new products or, otherwise, use produced fatty acids (e.g., acetate) and alcohols (e.g., ethanol) to ferment and convert them into other, higher value, compounds.

Acetone and iso-propanol are the most typical and probably most studied examples of metabolites produced by genetically modified acetogenic organisms (GMO). In recent research, production of acetone was studied in engineered acetogen, i.e., *A. woodii*, in which the acetone biosynthesis pathway was constructed by combining genes from *Clostridium acetobutylicum* and *C. aceticum* (Arslan et al., 2022). Acetate, as by-product, was however, still detected. In another study, efficient combined production of acetone and isopropanol was also assessed at pilot-scale based on the engineered autotrophic acetogen *C. autoethanogenum* (Liew et al., 2022).

The other alternative consists in using acetate and/or ethanol obtained from gas fermentation for their further bioconversion into other valuable products. Acetate and ethanol produced by acetogenic bacteria from C_1 gases act as electron donor and electron acceptor, respectively, in a chain elongating process, yielding C_6 and, eventually, C_8 medium chain fatty acids. Chain elongation has been studied both with mixed bacterial cultures as well as with pure cultures, mainly focusing on the species *Clostridium kluyveri*. It was shown that co-cultures of an acetogenic strain, fermenting C_1 gases to produce acetate + ethanol, e.g., *C. aceticum*, together with a chain elongator, e.g., *C. kluyveri*, will yield medium chain carboxylic acids (caproate mainly) (Fernández-Blanco et al., 2022).

Acetate, obtained from gas fermentation, is also a suitable, cost-effective alternative to other more conventionl carbon sources such as glucose, for the production of a wide range of biofuels and bioproducts. Some examples include lipids (microbial oils) accumulated by oleaginous yeasts during fermentation of acetate obtained from gas fermentation, or the production of nutraceuticals such as ß-carotene produced under similar conditions with non-conventional yeasts grown on C_1 gas derived acetate (Naveira-Pazos et al., 2023).

3.2 Biological methane valorisation and biogas upgrading

Biological valorisation of methane is generally based on the use of methanotrophs, able to assimilate that compound as sole carbon and energy source. They are divided in two groups. Type I methanotrophs metabolise methane through the ribulose monophosphate (generally abbreviated as RuMP) pathway (or Calvin-Benson-Bassham cycle), in presence of oxygen, low amounts of methane, and relatively high concentrations of nitrogen and copper. Type II methanotrophs select the serin pathway and prefer environments with limited oxygen concentration, high amounts of methane and they tolerate low levels (or absence) of nitrogen and copper. Generally, all methanotrophs can express the enzyme methane-monooxygenase (MMO) allowing to generate energy through CH_4 oxidation. Methanotrophy has more recently also been shown to be possible under anaerobic conditions and with other electron acceptors than oxygen. In terms of methane valorisation, methanotrophs can produce methanol as common metabolite, but they can also accumulate biopolymers, lipids, proteins, or ectoine. Methanol is an interesting product, not only for its use as industrial solvent, but also for its potential for further conversion and use in the synthesis of other products, e.g., methyl tert-butyl ether (MTBE), and dimethyl ether (DME), or olefins.

Biogas can also be upgraded and enriched in methane (CH_4) content through conversion of CO_2 from that biogas into CH_4 catalysed by different possible (micro)organisms. Microalgae as described above can be used to assimilate carbon dioxide. The potential of microalgae to increase the methane content of biogas through CO_2 assimilation, with simultaneous accumulation of some useful products, e.g., lipids, was assessed more than a decade ago already (Tongprawhan et al., 2014). In another study, it was shown that, in co-culture systems, it is possible to enrich biogas from a methane content of less than 70% to as much as about 95%, while eliminating, at the same time, hydrogen sulphide typically found in biogas at low concentrations. Microalgae took care of CO_2 consumption while H_2S was oxidised by oxidizing bacteria using the oxygen genrated photosynthetically (Rodero et al., 2020). In another study, a co-culture of *Methylocystis bryophila* (methanotroph) and *Scenedesmus obliquus* (microalgae) was fed biogas. The methanotroph metabolised CH_4 using at the same time O_2 produced by *S. obliquus*, while the latter assimilate CO generated by *M. bryophila*. Besides, an organic intermediate released by the methanotrophic species allowed to enhance nitrogen absorption while increasing the maximum amount of algal biomass, accumulating more mono-unsaturated fatty acids, and favouring better adaptation to light (Li et al., 2022).

Besides, anaerobic bacteria are also suitable for biogas upgrading, metabolising CO_2 and converting it into additional methane and thus enriched biogas (Kennes-Veiga et al., 2024). The process is similar as described above and is based on CO_2 fermentation, but, in this case using methanogenic strains or, more often, mixed cultures converting carbon dioxide + hydrogen into methane, thus removing CO_2 from biogas and enriching the latter into a gas with increased methane content. The bioconvresion of CO_2 into CH_4 by hydrogenotrophic organisms would be as follow:

$$CO_2 + 4\,H_2 \rightarrow CH_4 + 2\,H_2O$$

4 Discussion and conclusion

(Bio)technologies for air pollution control can be implemented using either treatment processes that will simply remove pollutants. However, recent research and even upscaled studies have shown that an attractive alternative to conventional treatment processes consists in converting volatile pollutants, including greenhouse gases (e.g., CO_2, CH_4) into valuable biofuels and bioproducts with different types of microorganisms, e.g., aerobic bacteria, anaerobic bacteria, microalgae. Among anaerobic bacteria, acetogens are particularly interesting. They can metabolise C_1 gases either through direct conversion of those gases into carboxylic acids or alcohols, mainly with native bacteria or through their bioconversion into other platform chemicals (e.g., acetone, iso-propanol) with GMO. Alternatively, acids and/or alcohols from acetogenic fermentation can be metabolised by other microorganisms to generate other biofuels and bioproducts, broadening that way the range of high value compounds (e.g., microbial oils, nutraceuticals).

Author contributions

CN-P: Data curation, Investigation, Writing – review and editing, Writing – original draft. MV: Supervision, Writing – original draft, Writing – review and editing, Resources, Funding acquisition. CK: Funding acquisition, Writing – original draft, Project administration, Data curation, Supervision, Conceptualization, Writing – review and editing.

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