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Developing a biogas centralised circular bioeconomy using agricultural residues - challenges and opportunities

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Abstract:

Anaerobic digestion (AD) is an efficiency approach to convert organic residues to biogas and digestate. It can be used as a stand-alone process or integrated as part of biorefining process to produce energy, biofuels, biochemicals and fertilizer, which has a potential to play a central role in the emerging circular bioeconomy (CBE). However, the possible adverse environmental impacts related to a biogas centred CBE should not be underestimated. This review comprehensively analyses the benefits and potential adverse effects related to developing biogas centred CBE. In general, digestate is the main source and distributor for most of the potential contaminants, while the environmental risks are highly dependent on the input feedstocks and digestate management. For proper risk assessment, it is essential to understand the fate and toxicological interactions of various contaminants and to establish monitoring routines. Integrated treatment processes should be developed as these could both minimise risks and improve the economic perspective.

Keywords: Anaerobic Digestion, Biogas, Circular Economy, Nutrient recycling, Waste management.

1. Biogas centred circular economy

In recent decades, the concept of circular bioeconomy (CBE) has been promoted as a key ingredient of a sustainable society. Development of CBE is especially important for the agricultural sector as large volumes of agricultural wastes (e.g., lignocellulosic residues, animal slurries, food waste) exist that can be used for generation of energy and fertiliser, reducing environmental pollution and greenhouse gas (GHG) emission, and add extra value for the farmer and industries in the food value chain. Several circular business models are available to recycle agro-based waste, including biogas plants, upcycling entrepreneurship, environmental biorefinery, agricultural cooperative, and agropark (Donner et al., 2020). Among these approaches, developing a biogas centred circular bioeconomy (biogas CBE) to valorise organic residues, including agricultural waste, is very promising as it produces biogas as renewable energy and digestate as biofertilizer. AD could produce biogas and biomethane (biomethane is biogas that has been upgraded to natural gas quality) to replace fossil fuels, reduce CH_4 emissions from manure, store carbon in soils, produce green fertiliser and enable carbon re-use (EBA, 2020). In recent years, the scope of AD is expanding due to the availability of new technologies and the emerging drivers of nutrient conservation and recovery (Batstone & Virdis, 2014). There are several advantages to develop biogas CBE, including various applicable scales (from farm-size to centralised scale), relatively low-cost equipment and maintenance, and it recovers nutrients from the wastes stream as digestate (Kapoor et al., 2020). In addition, it is a process that can be easily adjusted to match the needs of developed or developing economies to optimise agri-waste utilisation, boost energy production, and mitigate GHG emission from their respective agricultural sectors. Moreover, AD has unique advantages to be integrated into various (anaerobic) biorefinery concept, either as the final disposal step or centralized to produce value-added bioproducts other than biogas/methane only (Sawatdeenarunat et al., 2016).

On the other hand, the implementation of a circular economy requires exploring both resource circularity to contribute to sustainable development and minimising the negative impacts (Velenturf et al., 2019). The traditional linear economy primarily focuses on the utilisation of resources for production, utilisation of products and disposal of waste. Comparatively, in the circular economy, resources and raw materials are utilised, recovered and regenerated at each stage of production and service period without disposal. Thereby, it reduces the carbon footprint in economic development (Blades et al., 2017). Nevertheless, recirculation and regenerating residues within the agricultural production system could risk (bio) accumulation of toxic compounds. Therefore, continuous monitoring, bioaccumulation testing and environmental impact assessment is needed within to maintain healthy circular systems.

Recent scientific research has focused on circular bioeconomy development, value-added product synthesis from waste, improved utilisation of resources and/or integration within a biorefinery platform. Nevertheless, no systematic review has explored the challenges of developing a biogas centralised circular economy, especially considering the overall picture and risk of spreading environmental contaminants. In this review, the potential risk of biogas CBE was systematically assessed, and possible solutions proposed. The risk associated with the accumulation of biological contaminants (antibiotics, antibiotics resistance genes), heavy metals and persistent chemicals are thoroughly discussed. The identification of methane loss from the biogas production processes and possible control measures are further elaborated, and other gases emitted either through leakage or as combustion products are considered. Overall, this review points out challenges and the way forward for establishing a sustainable biogas-centred circular bioeconomy.

2. Challenges in developing biogas centred CBE

There has been a long history to re-use agricultural residues/wastes to produce biogas, while the commercial development of biogas industries has boomed since the mid-1970s as a result of the energy crisis in Europe (Rüdiger, 2014). Recently, the role of biogas and biomethane have been emphasized as being among the most important contributors to minimize GHG emission and secure energy supplement on replacing nature gas from Russia (Biomethane production potentials in the EU, a Gas for Climate report, 2022). In terms of circular bioeconomy, the AD and biomethane production processes could be a central point to initiate various CBE activities as the synergy existed between agricultural residues, anaerobic digestion, and circular economy, where biogas processes uses agricultural residues to provide bio-product extraction activities and upcycled materials and energy, enhancing the biomass cycle system within the local community (Lybæk & Kjær, 2021).

(Figure 1)

Under the circular bioeconomy concept, implementing AD is no longer limited to biogas production or waste treatment, but also be considered as a tool to reduce GHG emissions, generate biofertilizer, and nutrients recovery (Fagerström et al., 2018). Additionally, methane from biogas plants can substitute the use of fossil natural gas. The biogas CBE concept illustrates a promising approach to connect each element together as an ecosystem within a closed loop (Fig.1). Unlike cascading use of materials, biogas CBE focuses on circularity and closing the biological nutrient cycle;

an efficient approach to minimising adverse environmental impacts and for enhancing the nutrient recovery from digestate product (Fontaine et al., 2020). The environmental benefits of biogas CBE are often highlighted. However, in order to fully achieve those benefits, it requires a full understanding of feedstock chemical composition, the fate of these compounds during AD, benefits offered by products and associated risks to ensure the safety of the user (Longhurst et al., 2019). For instance, the associated risk of distribution of emerging contaminants, such as antibiotics or antibiotics resistance genes, persistent chemical compounds and microplastics (MP) from sources such as animal slurries, food waste, and industrial organic wastes, have become an emerging risk for the society as a result of intensive livestock production (Pu et al., 2018). In addition, biogas plants tend to achieve acceptably efficient biogas production with low energy input, but this may illustrate a preference for using shorter retention times and lower digestion temperatures (mesophilic AD). Such a strategy will potentially lead to large amounts of undegraded organic residues and pathogens (due to the low temperature) remaining in the digestate. As a consequence, application of such a digestate as fertiliser has a risk of high GHG/odour emission, spreading of pathogens or other toxic organic compounds on agriculture soil (Nkoa, 2014). Additionally, GHG emissions (CH_4, CO_2, N_2O, etc.,) associated with biogas production activities, from storage to field application, should be included when assessing the environmental impact (Buratti et al., 2013). Moreover, although the application of digestate as fertiliser has been well-accepted as an alternative for sustainable agriculture, the effectiveness of digestate as organic amendment and fertiliser is under discussion (Nkoa, 2014). Knowledge about their long-term impact, for instance on soil structure and fertility, are needed. Understanding on any perceived risks on re-using products from biogas-CBE should also be highlighted in order to establish fully confidence in their safety. Both of the benefits and risks on re-using of products from biogas-CBE should be understood to secure the safety on developing biogas-CBE (Muvhiiwa et al., 2017).

2.1 Green-house gas (GHG) emission

Biogas CBE has tremendous potential to reduce GHGs emissions by replacing fossil fuels, reducing methane emissions from manure and recirculating carbon and enabling its re-use (Capros et al., 2019). Meanwhile, it is still a challenge to estimate methane emission precisely form processes, for instance anaerobic digester, biogas upgrading, and digestate. According to Bakkaloglu et al. (2022), methane emission is two times greater than previosuly estimated, while the digestate storage stage contributed to majority of methane emission.

(Figure 2)

In addition to emissions from biogas production process and digestate storage, application of digestate at the field has been reported as another significant source of GHG emission (Møller et al., 2009). Significant efforts have been made for decades to quantify the GHG emission (CH_4, N_2O, or CO_2) along the biogas process. Recently, different methods such as Fourier transform infrared spectroscopy (FTIR), Open path tunable diode laser inverse dispersion mode spectroscopy (OP-TDLAS), Remote sensing (RS), Tunable diode laser absorption spectroscopy (TDLAS), High volume sampler (HVS) and onsite quantification have been tested for quantification of CH_4 loss from biogas production sites (Kvist & Aryal, 2019). However, it is still unclear on CH_4 emission corresponding to each stage along biogas and biomethane production. Fugitive emissions at the AD facility is not negligible and has been assumed to account for 1.8%

of the methane produced in the digester due to leakages from process equipment (Olesen et al., 2004). However, according to Bakkaloglu et al. (2021), the losses at biogas plants vary greatly; the measured emissions between 0.02 and 8.1% of the total methane production, with an average of 3.7%. Digestate storage emits significant amounts of GHGs, which can account for 32.3% of the total emission from the biogas production chain (Buratti et al., 2013). Digestate is produced in large quantities and consists of a mixture of microbial biomass and undigested material that emits ammonia and methane after storage and field application (Monlau et al., 2015). A particular case study concluded that the examined biogas plant failed to meet the GHG reduction target (35% saving in CO_2 emission), mainly due to the open storage of digestate (Buratti et al., 2013). The increasing number of large-scale commercial biogas plants might lead to an oversupply of digestate in a particular area which amounts to significant GHG emissions (Rehl & Müller, 2011).

Besides digestate, animal manure storage prior to digestion, especially liquid manure, is another hotspot of GHG emission (dominated by CH_4 and nitrous oxide (N_2O)). The animal slurry contains volatile solids (VS), which will be converted to CH_4 by methanogens when it is stored anaerobically, while N_2O is associated with nitrogen transformations via nitrification and denitrification under oxygen-limited conditions (Habtewold et al., 2017; Kristensen et al., 2011; Maharjan & Venterea, 2013). The animal slurry is usually stored from 10 to150 days in tanks before feeding to biogas plants and can undergo CH_4 losses which were quantified to be 1-2% of the total CH_4 production potential from manure (Feng et al., 2018b; Shin et al., 2019).

Biogas from anaerobic digestion contains mainly CH_4 but also a significant concentration of CO_2 as well as other components such as H_2S, $H_2O(g)$, and ammonia, in much lower concentrations. Even for end uses such as Combined Heat and Power (CHP), the non-CO_2 gases need to be brought down to a level to suit the equipment, based on the supplier's recommendations (Thomas & Wyndorps, 2012). If biomethane is the desired gaseous end product, which has natural gas quality (typically >97% methane content), the CO_2 also needs to be removed as well as higher standards of removal of the trace gases (Aryal & Kvist, 2018; Aryal et al., 2018). The upgraded methane can be utilised in different applications, such as transportation fuel and a renewable substitute for natural gas (Raboni & Urbini, 2014). CH_4 loss during gas upgrading generally ranges from 1-4%, depending on upgrading technologies (Adnan et al., 2019). It has been reported that 1.97% of methane slips from water scrubber biogas upgrading plants (Kvist & Aryal, 2019). Interestingly, the water scrubber is the dominant biogas upgrading technology globally. Comparatively, amine-based technology has reported only 0.04% methane slips while upgrading biogas. This illustrates methane slips from biogas upgrading plants should not be ignored (Kvist & Aryal, 2019). Similarly, another study reported that GHG emissions related to biogas upgrading amounted to 7.4% of the total GHG emission of the biomethane production chain (Buratti et al., 2013).

The development of biogas CBE indicates that almost all digestate should be used as fertiliser to recycle nutrients from agricultural residues. As an efficient and sustainable biogas CBE, it must be ensured that emissions are minimised, whilst pre-process biomass storage and digestate management are the critical steps. Consequently, dedicated measures, such as process monitoring, digestate/slurry management, should be taken to minimise the GHG budget. In addition, sustainable re-utilisation of CO_2 from biogas, for instance, to produce food-grade CO_2 or used for carbon capture and storage (bio-CCS), might be an alternative strategy to mitigate the GHG emission (Esposito et al., 2019). Besides CH_4, N-related emissions, *e.g* N_2O and NH_3(g) formed a large proportion of GHG. However, the implementation of digestate (as fertiliser) in general has lower total emissions of N compared to mineral fertilizer (Verdi et al., 2019). Additionally, it is necessary to adopt methane slip monitoring at biogas production plants, which will allow for mitigation after identifying the emission loopholes. One study recommended applying a regenerative thermal oxidiser (RTO) that may lower the emission from biogas upgrading facilities by 99.5% (Kvist & Aryal, 2019).

2.3 Antibiotics and antibiotics resistance gene (ARG)

Nowadays, intensive livestock production has led to higher demands on animal health, resulting in excess utilization of antibiotics to prevent sickness or enhance growth in animals (Levy, 2013; Widyasari-Mehta et al., 2016). Most of the active antibiotics cannot be metabolised entirely and, as a result, will enter downstream processes such as direct field application or biogas production in an unchanged form (Hirsch et al., 1999). Consequently, using antibiotics contaminated slurry to produce biogas will potentially make the digestate a carrier medium to transport antibiotics to environment (Feng et al., 2017). Spreading of antibiotics contaminated digestate provides a selective advantage for resistant bacterial populations and risks of transferring antibiotics resistance genes (ARGs) to soil microbes and plants, which could disable the efficacy of antibiotics against pathogens so that infection is not effectively inhibited in clinical treatment (Zhang et al., 2021a). Currently, the spreading of ARGs has attracted widespread concern due to their considerable threat to public health and environmental ecosystems (Barancheshme & Munir, 2018). During the AD process, some antibiotics are remediated within a short process retention time while others are very persistent (Feng et al., 2017). Indeed, it has been widely reported that AD is inefficient in removing antibiotics and that AD effluents may contain a relatively high amounts of un-degraded antibiotics. Álvarez et al. (2010), for instance, reported that most of the antibiotics could be effectively removed during AD of livestock manure but the removal efficiency varied among 40-90%. Arikan et al. (2006) assessed the removal of oxytetracycline during the AD of manure and only achieved a removal efficiency of 59%. Such presence of low levels of antibiotics in AD processes could support the selection and proliferation of microbes carrying ARGs (Yang et al., 2016).

ARGs have been considered as one of the new emerging types of contaminants, while the fate and mechanism of ARGs reduction in AD process are not fully recognised (Wang et al., 2019). Spreading of ARGs follows not only digestate application but can also be emitted *via* biogas digestate during storage through aerosol dispersion, including bacteria possessing NDM-1 and vanB ARGs (Zhang et al., 2021b). Compared to the antibiotics themselves, it is even more complicated to predict the fate of ARGs. In general, AD is considered a process capable of ARG reduction from animal manure (Flores-Orozco et al., 2020). However, even if the absolute abundance of ARGs is reduced, there may still be ARGs found in the AD effluent (Mao et al., 2015). Tian et al. (2019) reported 55-87% of ARGs reduction during thermophilic AD process, while Pu et al. (2018) observed increased ARGs' abundance during AD of pig manures. AD of antibiotic-containing animal slurry creates favourable conditions for ARG proliferation as it applies persistent selective pressure from antibiotics at sub-inhibitory concentrations on a dense and diverse microbial population in a nutrient rich environment (Rizzo et al., 2013).

Besides spreading antibiotics and resistance genes to the environment due to incomplete degradation, the existence of antibiotics could influence the anaerobic digestion process itself (Xiao et al., 2021). Theoretically, the AD efficiency is inhibited with the presence of antibiotics and residues, leading to lower efficiency of the entire AD system (Kovalakova et al., 2020). However, experiments with antibiotic mixtures have shown both synergistic and antagonistic effects on biogas production, depending on the type of antibiotics (Xiao et al., 2021). Long-term evaluations of the biogas-based circular economy and its impact on the environment and the food production system are still insufficient. It is necessary to monitor antibiotics and antibiotics resistance genes in the digestate and control measures could be applied. It has been suggested that contaminated digestate from wastewater treatment plants should be either landfilled or utilised in gasification processes in order to harvest the energy as biogas or syngas (Werle & Sobek, 2019). The syngas is intermediated gaseous feedstock that is rich in carbon monoxide (CO), CO_2, CH_4 and H_2 (Rasmussen & Aryal, 2020). The syngas derived from the gasification of contaminated digestate can be further utilised to produce chemicals or fuel (Aryal et al., 2021a).

2.4 Micro and macro nutrients application and their impact

AD process releases large quantities of nitrogen and phosphorus into the liquid phase (Ma et al., 2018). Thus, AD digestates have high content of nutrients, which do not generally meet the requirements of local regulations to be discharged directly. Indeed, as well as being a resource rich in macro-nutrients (nitrogen, phosphorous, and potassium), digestates also contain micro-nutrients (*e.g.* Ca, Mg, Zn, Mn, etc.). Both macro and micronutrients are vital for crop cultivation and therefore the common approach is to apply digestate directly as fertiliser (Möller & Müller, 2012). Currently, 95% of the digestate produced in EU is used as an organic fertiliser for field crops on agricultural land as an alternative to mineral fertiliser (Saveyn & Eder, 2014; Vaneeckhaute et al., 2013). Under the concept of CBE, application of mineral fertiliser should be avoided and instead directly use animal manure or digestate. Compared to manure, utilisation of digestate as fertiliser has some advantages as it has higher proportions of mineralised plant-available nutrients than untreated manure and a significant reduction in odour (Insam et al., 2015). In addition, digestate has higher alkalinity/pH than untreated manure, which is considered a benefit for preventing soil acidification.

Nonetheless, some uncertain issues and challenges exist. As the end product of anaerobic digestion, the fertiliser values are linked to the input feedstock and parameters, for instance temperature or retention time (Guilayn et al., 2019; Tambone et al., 2013). A large variation in total nitrogen from digestate was previously reported in the literature and was related to the N content in the feedstock (Risberg et al., 2017). Pokój et al. (2015) evaluated the digestate's composition from annual and perennial crops and found a big variation in fertilising values. The availability and turnover of micronutrients from digestate are rarely reported. Because of the complexity of the AD processes and formation of complexes in solution with intermediates and product compounds produced during AD (Callander & Barford, 1983; Chen et al., 2008). The compositional variability of digestates is a challenge for precise fertilisation and has been indicated as one of the major bottlenecks for digestate fertilizer (Dahlin et al., 2015).

As a fertiliser, it is important to assess the efficiency of nutrient uptake between raw manure and digestate. In general, digestate has both higher total nitrogen (TON) and a higher ratio of NH_4^+/TON, which is desirable as fertiliser (Risberg et al., 2017). However, long-term use of fertilisers rich in NH_4^+may negatively affect primary plant root growth (Liu et al., 2013). High NH_4-N concentration in residues also creates a risk of N losses through ammonia volatilisation and N leaching (Chu et al., 2007; Riva et al., 2016). As another important plant macronutrient, it is often stated that AD will enhance the availability of phosphorus (Massé et al., 2011). However, results from most of the field experiments indicated that AD of animal manure has almost no effect on P availability but even has negative influence on crop P availability (Bachmann et al., 2011). Moreover, as a nutrient 'closed' system, development of biogas CBE points to a future where more animal slurry, food waste and agricultural waste will be recycled for producing energy and fertiliser. This will enhance the total nutrients available for agriculture and ensure less nutrient losses within the food production system (Stinner et al., 2008). However, the demand for fertiliser and nutrients is seasonal, which does not usually match the generation of digestate, especially from areas with centralised biogas plants (Baral et al., 2018). In addition, nutrients extracted via anaerobic digestion are supposed to be circulated within the agricultural system. However, these nutrients may not necessarily match the need for regeneration of the system, which could eventually destroy the cycles (Skene, 2018). It is therefore recommended to implement adequate measures to ensure nutrient cycling and avoid nutrient shortage or excess in the ecosystems. Improved quality assurance of digestates to be used as fertiliser is required, which can mean quantification either of the inputs to an AD plant or of the produced digestate, documented proof of the process parameters (e.g., temperature, retention time) and certification of the treatment plant to declare and label their products. To some extent, such quality assurance already exists for aerobically composted organic wastes, but not for digestates (Saveyn & Eder, 2014). Some national regulations exist, such as in the UK (Al Seadi & Lukehurst, 2012), but these are typically to quantify maximum permissible concentrations of pathogens and heavy metals rather than the levels of macro and micronutrients.

Soil microorganisms are critical to soil functions, for instance formation of stable aggregates and cycling of plant nutrients, while microbial activities normally response faster than physical and chemical properties of the soil (Sapp et al., 2015). Thus, monitoring of the microbial response after application of fertilizer can give early indication of potential negative impact (Stenberg, 1999). Nyberg et al. (2006) reported that soil microbial activities, *i.e.* potential ammonia oxidation rate (PAO) and soil respiration, were not significantly changed when amended with digestate compared to manure. In some cases, direct implementation of animal manure could have negative impact on soil microorganisms, for example, activities of PAO could be inhibited due to existence of organic acids. In contrast, it was reported that application of digestate to soil led to lower microbial activity in the soil and decreased biomass of earthworms compared to conventional cattle slurry, since the digestate contains less readily available nutrients but higher barely decomposable organic matter (Ernst et al., 2008). In general, the quality of biofertilzer (digestate) is largely dependent on input biomass which has large variations within or between biogas plants, which might be the main reason of the contradictory results reported, in combination with the limited number of samples studied (Risberg et al., 2017). It should be noted that some novel technologies offer the possibility to manipulate the quality of digestates, like improving the N content by using plasma technology (Mousavi et al., 2022).

2.5 Emerging contaminants sourced distributed along the value chain

There is a long list of residual contaminants that are continuing to be identified in a wide spectrum of biological wastes (as detailed in section 2.2) including emerging contaminants discharged through aqueous or solid waste streams (Longhurst et al., 2019). However, most of these contaminants are associated with recirculating of the biosolids from sewage sludge treatment, while that from agriculture-based biogas plants are lower as the input are mainly animal manure and crop residues. In practice, most biowastes have contaminants, including plastics, regardless of if they have been pre-treated or not (Weithmann et al., 2018). Utilisation of more biowaste for biogas will inevitably introduce more microplastic particles (MPPs) into the biogas stream, which will be further transferred to the environment after field application. Thus, the potential risk of applying organic fertilisers may be a source of environmental MPPs that should not be underestimated. Moreover, accumulating those contaminants affect not only the subsequent utilisation of digestate, but also the process itself. Previous studies noted that certain types of (nano) microplastics, such as polyethylene, polyvinyl chloride, etc., could adversely affect anaerobic digestion under both short and long-term exposures (Feng et al., 2018a; Wei et al., 2020). Accumulating specific types of MPPs could highly inhibit the AD process and reduce the biogas yield. Wei et al. (2019) demonstrated a decline in hydrolysis rates (up to 15%) and methane production (up to 27.5%) at high polyethylene levels (100–200 particles/g-TS). The potential inhibition mechanisms were summarised by Mohammad Mirsoleimani Azizi et al. (2021), including: i) leaching of toxic chemicals or additives from microplastics can affect microbial activities in a digester by directly damaging microbial cells and by inhibiting activities of key enzymes associated with biochemical reactions and defence against oxidative stress; ii) microplastics can catalyse the generation of reactive oxygen species

(ROS), such as H_2O_2 (the mechanisms of ROS generation under anaerobic conditions are still ambiguous); iii) microplastics may directly damage the microbial cells and inhibit metabolic functions (Fu et al., 2018). It is notable that the (nano) plastics could penetrate microbial cell membranes through the gaps between the biopolymer chains, leading to damages on proteins and phospholipids (Zhang & Chen, 2020).

Heavy metals (HM) in manure-based residue/slurry attracts concern as they could be transferred from digestate to farmland and subsequently to crops (Zhang et al., 2019). Implementing digestate containing excessive HMs as fertiliser can increase the bioaccumulation rate of HMs in the crops leading to increased risk to human health (Shamsollahi et al., 2019). Several studies showed that, as one of the main substrates for agricultural-based biogas plants, swine manure contains high concentrations of HM due to the feed additives (Ji et al., 2012). HMs in the digestate are known to vary significantly as a result of changing the input materials or the process control. Zheng et al. (2020) reported the majority of HMs identified from manure-based biogas residue, which were found in the order of Zn > Cu > Ni > As > Pb > Cd (decreasing concentration). Nemati et al. (2009) found that Fe and Mn were the highest concentrated metals in aquaculture sludge, followed with Cu, Cr, Cd and Pb. Fish sludge was reported to contain large amounts of As, Pb, Hg, Cd (Uotila, 1991). It is widely accepted that trace amounts of certain HMs are necessary for the activity of some enzymes involved in the biogas process (Guo et al., 2019). However, in terms of biogas-CBE, occurrence of HMs could both decrease the efficiency of biogas production and prohibit the field application of digestate if the concentration is above the limitations, such as those provided in the UK (Al Seadi & Lukehurst, 2012). Heavy metals, for instance Cu and Zn, or mixtures, could have inhibitory impact on production of intermediates in the AD process, such as total volatile fatty acids (VFAs) (Lin, 1993). As a biological process, the anaerobic digestion process itself will not reduce the HM concentrations. In contrast, it was reported that anaerobic digestion could enhance the bioaccumulation of heavy metals in the crops after using the digestate as fertiliser (Shamsollahi et al., 2019). Meanwhile, it was also reported that the HMs are more prevalent in the solid fraction of digestate than liquid, regardless of the variety of feeding materials, which offers the opportunity to manage HMs prior to field application (Dąbrowska & Rosińska, 2012). In addition, HMs could interact with other emerging contaminants; for instance, Ji et al. (2012) reported that toxic HMs, such as Hg, Cu, and Zn, exerted a strong selection pressure and acted as complementary factors for ARG abundance. Studies have shown that when digestates are applied to soil leaching of metals like Al and Cr will occur but usually be below regulated threshold values (Dragicevic et al., 2018).

Organic contaminants are unwanted chemical compounds supplied to biogas process in various amounts via digestible materials (Al Seadi & Lukehurst, 2012). Non-degradable organic contaminants, also known as persistent organic contaminants (POPs), are normally directly toxic to biota and could progressively accumulate higher up in the food chain (Aravind kumar et al., 2022). The occurrence of POPs varies geographically, which depends on the incoming feedstock, while the fate of POPs is affected by the process control (Longhurst et al., 2019). In general, the majority of POPs ultimately enter wastewater treatment or sewage sludge while agricultural waste-based biogas process, especially crop-derived feedstock, mainly contain traces of herbicides and fungicide rather than other POPs (Al Seadi & Lukehurst, 2012). It should be noted that the occurrence and concentrations of such POPs are dependent to a large extent on control through legislation. For instance, implementation of the persistent pesticides DDT and HCH, has been banned in most of the EU countries, which therefore eliminated such contaminants from the agricultural AD. However, these pesticides are still being used in most developing countries, leading to much high risk of their occurrence in agricultural products and waste streams (Handl, 2012).

2.5 Airborne trace contaminants

The potential for air quality impact when developing biogas energy source is normally overlooked as biogas is considered as a 'green' gaseous energy. However, the generation of biogas can also spread airborne contaminants, such as airborne microplastics, carbonyl compounds, and ammonia, which are often neglected and could also pollute the environment. Airborne carbonyl compounds have long been chemicals-of-concern in the environment since their reactivity and potential for negative health effects (Alaimo et al., 2021). Like digestate, the toxicity of combusted biogas is normally associated with the substrate used for biogas production. Li et al. (2019) reported the composition of six California biogas streams from different feedstocks (dairy manure, food waste, and municipal solid waste) and the toxic compound in each stream is varied. Utilisation of biogas to produce electricity and heat in a CHP unit also produces airborne emissions such as NOx, CO and SO_2, the presence of the latter prevents the use of exhaust catalytic conversion, therefore the motors are run on excess air to lower the combustion zone temperature, which reduces NOx emissions (Thomas & Wyndorps, 2012). However, the authors found that the studied CHP motor often drifted from the excess air optimisation setting very quickly, leading to increased NOx emissions.

Compared to MPs in digestate, airborne microplastics has received increasing attention as the ubiquitous nature and the potential human health consequences of inhalation (Zhu et al., 2021). The environmental implications of airborne contaminants from biogas will depend on the anticipated end-use; direct on-site electricity generation will likely combust the carbonyls along with the methane in the biogas leading to no subsequent environmental reactivity while upgrading biogas to biomethane by removing CO_2 and other components such as H_2S, siloxanes can lead to emission of these compounds (Aryal et al., 2021b). After CO_2 removal, biogas becomes biomethane, a direct alternative for natural gas, which require careful consideration (Alaimo et al., 2021). Therefore, biogas plants coupled with upgrading facilities should be characterised in order to minimise potential trace contaminants (Aryal et al., 2021b).

3. Approaches to address the potential impact

As aforementioned, the potential risks of developing biogas CBE stands on: 1. GHG emission due to the biogas process and digestate application; 2. Nutrients management (the mismatching (time and/or location) between the generation of digestate and demand of nutrients); 3. Contaminants due to implementation of digestate as fertiliser, and 4. Gaseous carried airborne contaminants. The specific impact of a particular biogas plant differs due to the varied input substrates. In general, farm-based biogas plants that use animal manure and agricultural by-products have a much lower risk of distributing contaminants as the input biomasses are generated within the loop of agricultural production, but it often has a higher risk of spreading antibiotics and ARGs. Sustainable management and utilisation of digestate is the key to minimise the negative impact of biogas CBE, as it is the main source of both GHG and contaminants. Application of specific regulations, monitoring and maintenance and/or the application of specific technologies to reduce GHG or minimise the potential environmental risks, such as restrictions on input materials or concentration limits, obligatory use of specific processes or conditions of use, have been previously well-discussed (Corden et al., 2019; Liebetrau et al., 2017). Moreover, biogas CBE is a biological system that treats a complex input and a variety of contaminants, therefore, physical-chemical features should also be identified to assess the possible distribution of specific risks, while an understanding of the interactions between these contaminants is also the key to assess the potential risk correctly.

(Figure 3)

3.1 Fast identification and monitoring

To build up an efficient and sustainable biogas based circular economy, it is of great importance to identify the occurrence/fate of contaminants and monitor the relevant environmental impact/potential risk to support the risk-informed decision making and selection of feasible subsequent process. For instance, there are in general two different approaches for emission measurement (from biogas plants); 1. identify and quantify every single source on site and sum them; or 2. consider the overall plant as one single emission source and the overall emission of the plant is determined (Liebetrau et al., 2017). There are various technologies available for qualification of the emission, such as direct measurement (for instance, closed chamber measurement) or indirect approaches such as portable imaging infrared cameras or remote sense, for instance laser spectrometers or tracer ratio methods (Vergote et al., 2020). Both methods have their own limitations due to their limited viability, unrepresentative data set (due to a limited data collection period), and also the disagreement between the data reported from laboratory and on-site measurements (Reinelt et al., 2017). Another main challenge of implementing relevant approaches into a biogas-based process is to make the approach cost-effective to support long-term analysis and adequate sampling replication. For instance, the MP contamination from digestate is often quantified with visually examining a pre-filtered and digested sample (Piarulli et al., 2020; Rivier et al., 2019). Spectroscopic-based analyses (e.g Raman or FTIR microscopy and/or spectroscopy) are increasingly used, but they may be time-consuming and require extensive manipulation of the samples. Nevertheless, monitoring substrate contamination has the advantage of controlling the contaminant's spreading, as it is harboured within digestate, and the appearance of specific contaminants is somewhat predictable as it is always associated with the input substrate. Thus, elaborating standards and specifications for digestate management to minimise the environmental impact and increase biosafety should be prioritised. Novel approaches, such as machine learning (ML) algorithms, have received significant interest in AD process optimisation, prediction of uncertain parameters, detection of perturbations, and real-time monitoring (Andrade Cruz et al., 2022). ML based processes uses inductive inference to generalise correlations between input and output data and therefore is an efficient tool to assist in decision-making (Alzubi et al., 2018). With sufficient training and input data, ML might be also capable of predicting the potential environmental impact, not only of the contaminants but probably also the GHG emission.

3.2 Understanding of occurrence, fate, and toxicological interaction of contaminant

Understanding of the occurrence, fate, and toxicological interactions of contaminant is crucial to minimise the environmental impact of biogas CBE. Digestate, especially sewage sludge-based biogas digestate, contains multiple types of contaminants. Co-occurrence of several contaminants could lead to interactive toxicity and have an increased negative impact on the environment. For instance, interaction of MPs with co-contaminants, e.g., HMs, pesticides, pharmaceuticals, can cause significant changes to the surface properties of the MPs which is able to alter the MPs uptake and accumulation of MPs and other contaminants in the organisms exposed to them (Zhou et al., 2020). Changes in the accumulation profile in a variety of microorganisms during a combined exposure have been associated with synergistic, antagonistic, or additive effects (Bhagat et al., 2021). Besides microplastics, significant positive correlations have been observed between ARGs/antibiotics and HMs, indicating the potential selection pressure of HMs on the ARG/antibiotics (Lu et al., 2020). It is therefore crucial to have a deep understanding of potential toxicological interaction of various contaminants for risk assessment and management.

3.3 Approach for impact assessment, identifying new risks and prioritisation

For biogas CBE to succeed in strengthening sustainable development, it is mandatory to have the entire life cycle assessment to avoid negative environmental effects. Prioritising the specific risk associated with the process could enlighten the strategy on selecting the most effective subsequent process to minimise the risk. Rigorous assessment of the environmental performance of biogas CBE requires a detailed assessment from multiple feedstocks and energy conversion scenarios including options for utilisation or disposal of the digestate (Poeschl et al., 2012). New risks should be identified as the project progresses through its life cycle. Life Cycle Assessment (LCA) is considered as a structured, comprehensive, and internationally standardised method, which has, therefore, become one of the most useful tools. LCA quantifies all relevant emissions and resources consumed and the related environmental and health impacts and resource depletion issues that are associated with any goods or services ("products") (Chomkhamsri et al., 2011). LCA has been widely implemented to evaluate both the environmental and public health impacts under various scenarios of biogas production (Lyng et al., 2015). Pivato et al. (2016) proved that LCA could be a potentially useful tool to assess the effect of contaminants on digestate management. Hijazi et al. (2016) concluded that LCA could provide a largely clear picture of the environmental impact of biogas systems and suggest possible measures to minimise the negative effects. However, there are several environmental consequences, such as dispersion of drug residues, bacterial resistance, and spreading of microplastics that are of critical importance but have not yet been considered in LCA (Havukainen et al., 2022).

3.4 Processes integrated into the biogas process

Specific to biogas CBE, digestate is obviously the main source of potentially hazardous substances. Sustainable management and recycling of the nutrient-rich anaerobic digestate is required, in order to close the resource loop and actualise the biogas CBE. Thus, integrated processes which could add value to a biogas process, improve nutrient recovery, and reduce contaminants and/or GHG emission are promising. Indeed, the effect of an integrated process on minimising the environmental impact of biogas-CBE is largely dependent on the types of contaminants. Digestates can be processed downstream to separate nutrients and therefore contaminants. Such methods vary in their efficacy, cost, and readiness for market when used with digestates, but can include separation of solid/liquid fractions, membrane filtration, ammonia stripping, pyrolysis and gasification (Lamolinara et al., 2022). Gurmessa et al. (2021) treated digestate by composting, which removed more than 80% of ARGs from digestate. However, post-composting of digestate had very limited impact on reducing the abundance of MPs or even no impact at all on specific types of MPs (Sun et al., 2021). Besides composting, integrating microalgae cultivation with an AD process also presents an opportunity to establish a circular economy solution that could reduce the excess nutrients and GHG emission from the digestate (Stiles et al., 2018). It was also reported that microalgae are able to achieve 100% removal of nitrogen and phosphorus, 50-90% of COD, and also 35-90% of heavy metals from liquid digestate (Xia & Murphy, 2016). However, the most prominent challenges for algal treatment are turbidity (even for liquid digestate), which directly affects light availability, and ammonia toxicity, resulting from the high ammonia nitrogen concentrations in anaerobic digestate (Chuka-ogwude et al., 2020). Various pre-treatment methods have been applied to remove the solid particles from digestate to reduce the turbidity, for instance filtration, centrifugation, and precipitation (Cheng et al., 2015; Yang et al., 2015). Nevertheless, the impact of digestate pre-treatment methods on microalgal growth rates is rarely investigated (Xia & Murphy, 2016). Solid digestate, on the other hand, could be processed with approaches such as hydrothermal processes, to produce syngas and biochar, or hydrothermal liquefaction (HTL) to yield biocrude (Monlau et al., 2015).

Integrated process could have extra benefits in terms of minimising of the environmental pollution from biogas process. For instance, Chand et al. (2022) reported that the continuous hydrothermal liquefaction (HTL) led to approximately 76% of MP reduction in terms of number and 97% in terms of MP mass. In addition, hydrothermal treatment (HT) was reported as an effective approach to remove ARGs. Compared to pyrolysis, which requires intensive energy input due to the high moisture content of digestate, an integrated system via hydrothermal process is demonstrably more promising, as it has the potential to convert digestate to hydrochar without drying (Wang & Lee, 2021).

4. Perspectives and conclusion

Biogas CBE is an efficient way to improve sustainability. Still, some challenges need to be solved. In this review, the most significant environmental risks are summarised and potential solutions are proposed. As a versatile process, AD can treat broad range of organic residues and re-distribute carbon and nutrients. However, the risk associated with potential accumulation of pollutants originating from the feedstock must be addressed. Some aspects of risks have been studied, but overall, this review has shown clear knowledge gaps that needs to be filled. Moreover, an integrated approach to valorise biogas process could add value and minimise its impact.

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Tables and Figures:

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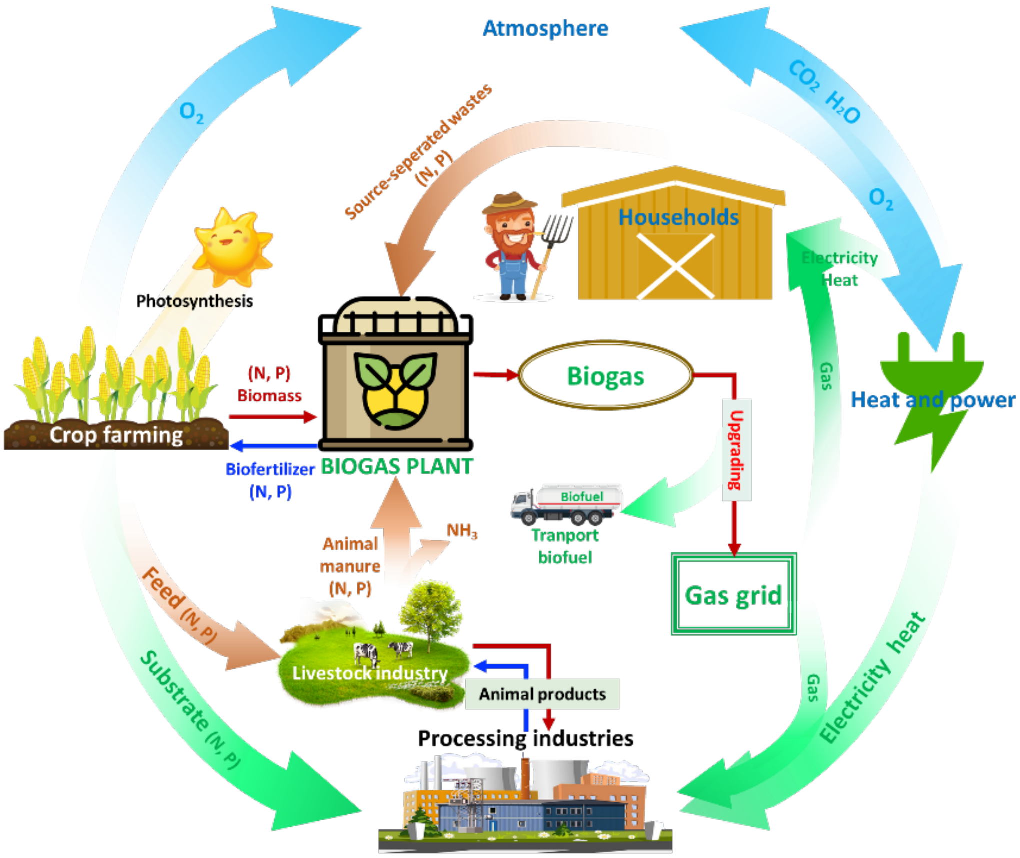
Figure 2. GHG emission from biogas process (Buratti et al., 2013).

Figure 3. The potential impact from biogas-based CBE.



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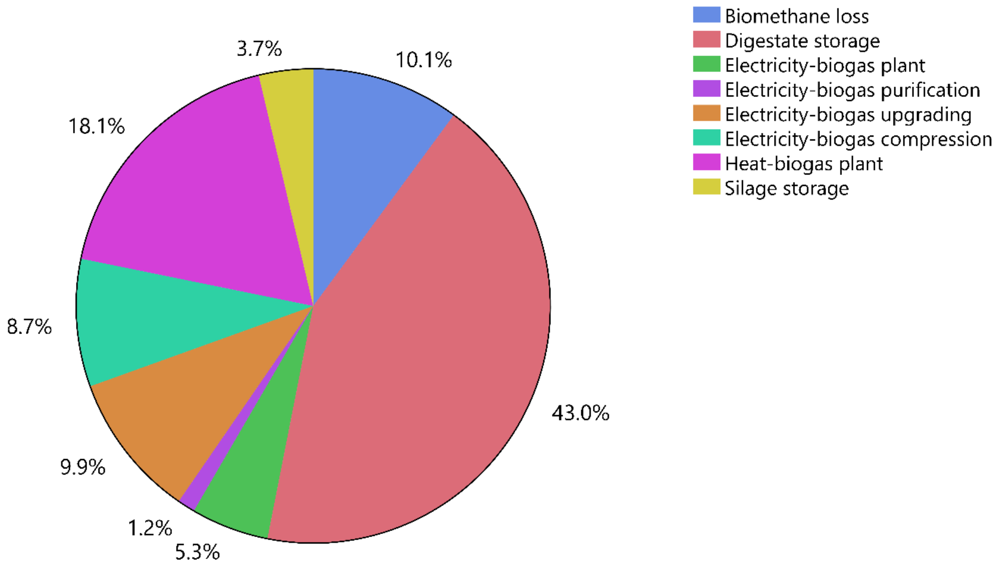

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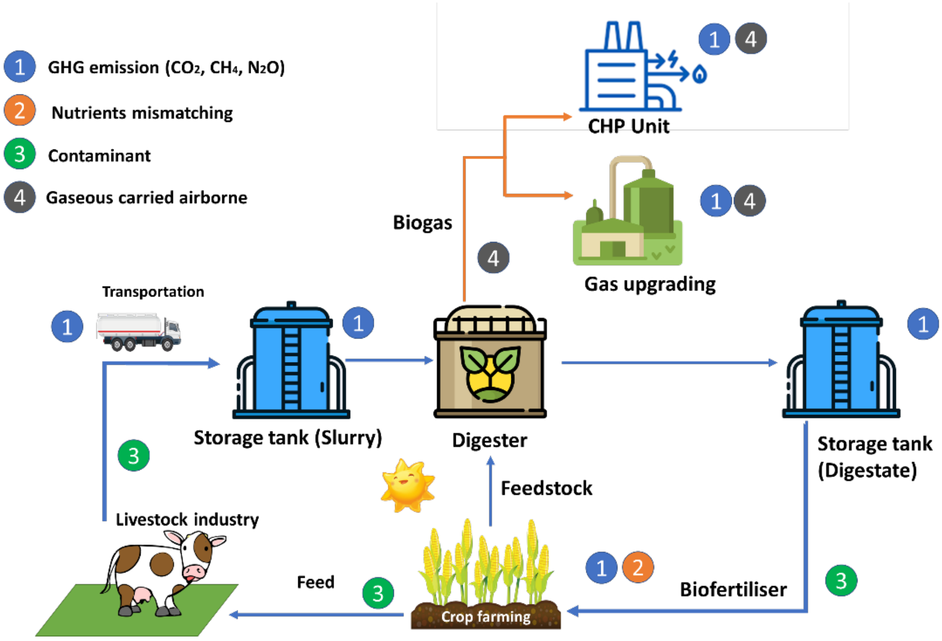

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