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Synergy of anaerobic digestion and pyrolysis processes for sustainable waste management: A critical review and future perspectives.

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Abstract

The worldwide growing population, challenged by an ever-increasing global demand for food production, is also concomitant with increased waste production, particularly organic waste. During the last decades, several waste processing technologies have been developed such as anaerobic digestion and pyrolysis. Recently, there has been an increased interest in creating industrial synergies by combining technologies in order to increase the efficacy of the process and improve waste management in the circular economy. In this review, we report on the importance of coupling anaerobic digestion and pyrolysis while providing evidence on the synergistic effects that may occur within such a combined waste bi-functional process. Specific attention has been paid to multiple symbiosis features that exist when coupling both processes, mainly 1) maximizing energy recovery through pyrolysis of solid digestate or feeding of the aqueous bio-oil phase in anaerobic digestion, 2) biogas purification by biochar or activated biochar, and 3) improving anaerobic digestion process stability by biochar addition to the system. In addition, the effects of coupling anaerobic digestate with biochar on soil biochemical properties and crop production were also presented. Improving the dual symbiosis of coupling anaerobic digestion and pyrolysis is likely to be a sustainable based approach that holds promise for wiser and more eco-efficient processing of organic wastes for versatile applications.

Keywords: Pyrolysis, anaerobic digestion, biochar, digestate, fertilizer, detoxification.

Abbreviations

APL: Aqueous Pyrolysis Liquid

BMP: Biochemical Methane Potential

COD: Chemical Oxygen Demand
DM: Dry Matter
EBC: European Biochar Certificate
GHG: Greenhouse Gases
HR: Heating Rate
HTL: Hydrothermal Liquefaction
IBI: International Biochar Initial
LCA: Life Cycle Assessment
OLR: Organic Loading Rate ($\text{kg VS Nm}^{-3} \text{ day}^{-1}$)
OFMSW: Organic Fraction of Municipal Solid Wastes
PAHs: Polycyclic Aromatic Hydrocarbons.
RT: Residence Time
S_{BET}: Specific surface area ($\text{m}^2 \text{ g}^{-1}$)
TAN: Total Ammonia Nitrogen
TRL: Technology Readiness Level
UASB: Up-flow Anaerobic Sludge Blanket
VFAs: Volatile Fatty Acids
VS: Volatile Solid

1. Introduction

As the world's population grows, food production should constantly increase to meet the increasing global demand. Moreover, considerable food waste accumulation needs to be processed in a sustainable manner to avoid possible environmental burdens associated with unprocessed organic waste. Several waste valorizations routes have been investigated and developed (pyrolysis, anaerobic digestion, composting). Recently, two main valorization routes have emerged through biological (anaerobic digestion) and thermochemical (pyrolysis) processes, allowing a large spectrum of wastes and biomasses [1,2] to be processed. Among the scientific communities, these two types of technologies have often been opposed, but in

recent decades, the combination of pyrolysis and anaerobic digestion processes have attracted increasing attention [3–5]. Anaerobic digestion is defined as a biological-based degradation process anaerobically producing biogas (CH_4 and CO_2) and a residue called digestate. Meanwhile, pyrolysis consists in a thermochemical decomposition of biomass that occurs in the absence of oxygen producing syngas (a mixture of CO , H_2 , and CO_2), bio-oil, and biochar. At the agricultural level, both processes can compete with each other in terms of biodegradation of a large biomass spectrum of substrates that are easily biodegradable and can be optimally degraded by the anaerobic digestion process, whereas treatment of hardly biodegraded substrates may be achieved by pyrolysis [6,7]. During the last decade, “cascading” use of biomass utilization, where the output of one process becomes the input of the following one (targeting the “zero-waste” goal) with biomass progressing through a series of material flows and energy conversions, has attracted attention [8,9]. The integration of such technologies may offer interesting opportunities with the objective of contributing to the circular economy [10]. Coupling anaerobic digestion - pyrolysis processes through an industrial symbiosis approach can address several issues related to agricultural waste management by improving resource use efficiency, energy recovery, GHG emissions reduction, and soil preservation [8,9,11–13]. For example, pyrolysis could be used to convert the dried solid digestate into biochar, which could then be used for different applications and purposes [14,15], including its potential use to improve biogas production and quality [16], to reduce instabilities and inhibition in digesters during the process [17] or to purify or upgrade the biogas [18,19]. The syngas produced during the pyrolysis process can also be converted through biological methanation [20]. Finally, the aqueous pyrolysis liquid could be used as organic feedstock for anaerobic digestion [21], possibly with the addition of biochar for selective adsorption and mitigation of toxic inhibitors.

Based on the science direct database between 2015 and 2021, the following **Table 1** summarizes some of the recent and interesting literature reviews dealing with the benefits of integrated pyrolysis or other thermochemical processes with anaerobic digestion processes and the different synergies between the processes and their products.

Table 1: Recent literature reviews of coupling anaerobic digestion and pyrolysis with the interactions of their products for energy, bioremediation, and agronomic purposes.

Review title	Journal name, year (IF: impact factor)	Main topics	Ref.
New opportunities for agricultural digestate valorization: current situation and perspectives.	Energy & Environmental Science, 2015 (IF: 38.5)	-Discussion of the use of both solid and liquid digestate for several applications (microalgae, biofuels, thermos-chemical processes...). -Investigation of coupling anaerobic digestion and pyrolysis for energy production through the conversion of solid digestate into added-value products (char , bio-oil, syngas)	[22]
Linking pyrolysis and anaerobic digestion (Py-AD) for the conversion of lignocellulosic biomass	Current Opinion in Biotechnology, 2016 (IF: 9.7)	-Investigation of the pyrolysis process applied downstream the anaerobic digestion as pretreatment for recalcitrant biomasses. -Study of converting the aqueous and organic bio-oil phases through anaerobic digestion process. -Biological methanation of the syngas -Investigation of the boundaries and perspectives of coupling pyrolysis and anaerobic digestion.	[8]
Integrated processes of anaerobic digestion and pyrolysis for higher bioenergy recovery from lignocellulosic biomass: A brief review	Renewable and Sustainable Energy Reviews, 2017 (14.9)	-Reviewing recent development, feasibility, and advantages of integrated processes of anaerobic digestion and pyrolysis. -Discussion of the various strategies of combination: anaerobic digestion-pyrolysis, pyrolysis-anaerobic digestion, anaerobic digestion-pyrolysis-anaerobic digestion .	[9]
A review of biochar properties and their roles in mitigating challenges with anaerobic digestion	Renewable and Sustainable Energy Reviews, 2019 (IF:14.9)	-Discussion of the challenges of integrating biochar in anaerobic digestion process. -Summarize of the characteristics of biochar and its physicochemical properties that can simultaneously promote anaerobic digestion process stability, increase biomethane yield rate. - Biochar for digestate quality improvement. - Biochar for CO ₂ adsorption and other impurities in biogas.	[23]
Coupling anaerobic digestion with gasification, pyrolysis, or hydrothermal carbonization: A review	Renewable and Sustainable Energy Reviews, 2019 (14.9)	-Summarizing the literature for the coupling of the biological process of anaerobic digestion with one of three thermal processes: gasification, pyrolysis, and hydrothermal carbonization. -Investigation the thermochemical processes as pre and/or post treatments for anaerobic digestion, and the synergies between the processes and their products. -Overview of the most promising future research investigations.	[4]

Review of biochar role as additive in anaerobic digestion processes	Renewable and Sustainable Energy Reviews, 2020 (IF: 14.9)	<ul style="list-style-type: none"> -Investigation of the influence of biochar properties on anaerobic digestion performances and its stability. - Discussion of microbial attachment and acclimation. - Study of biochar capacity for adsorption of inhibitors of anaerobic digestion. - Effect of biochar for biogas upgrading. -Evaluation of the economic and environmental advantages connected to biochar use in anaerobic digestion processes, compared to conventional solutions.. 	[10]
The role of biochar to enhance anaerobic digestion: a review	Journal of Renewable Materials, 2020 (IF: 8.0)	<ul style="list-style-type: none"> -Discussion the noting physicochemical properties of biochar, and its roles and related mechanisms in anaerobic digestion. -Highlighted the advantages and drawbacks and pointed out the corresponding challenges and prospects for future research and application of biochar amending anaerobic digestion. 	[24]
Potential of coupling anaerobic digestion with thermochemical technologies for waste valorization	Fuel, 2021 (IF: 6.6)	<ul style="list-style-type: none"> -Evaluation on the viability of integrating anaerobic digestion with thermochemical processes. -Comparison of three thermos-chemical processes: pyrolysis, gasification and hydrothermal carbonization. 	[25]
A critical review on biochar for enhancing biogas production from anaerobic digestion of food waste and sludge	Journal of Cleaner Production, 2021 (IF: 9.2)	<ul style="list-style-type: none"> -Discussions of the biochar integration into the anaerobic digestion process of food waste and sludge. -Roles and mechanisms of biochar in anaerobic digestion. -Techno-economic and life cycle analysis. 	[26]

Based on **Table 1**, the study of coupling anaerobic digestion and thermochemical process (especially pyrolysis) has been recently investigated in terms of energy recovery and by using pyrolysis biochar as an anaerobic digestion stabilizer/biomethane enhancer, or for biogas upgrading. Up to date, two main strategies has been investigated by means of anaerobic digestion followed by subsequent solid digestate pyrolysis and pyrolysis of recalcitrant biomasses prior to anaerobic digestion. However, the agronomic interest of coupling biochar and digestate or liquid digestate has been little studied in the different published review (Table 1). For these purposes, the following review proposes to investigate the overall synergy possible between anaerobic digestion and pyrolysis process with a special focus on coupling biochar and digestate for agronomic purposes. A specific paragraph is also dedicated to the economic, environmental, and societal consideration of the dual approach. Finally,

some promising perspectives of coupling anaerobic digestion-pyrolysis process are also discussed in this review.

2. Anaerobic digestion and pyrolysis processes

2.1 Pyrolysis Process

Pyrolysis is defined as the thermal decomposition of the organic matrix under non-oxidizing or very low-oxidizing stoichiometric atmospheres [27], and it occurs between 250 and 1200 °C [28]. During pyrolysis, organic matter such as lignin, protein, cellulose, and hemicelluloses are thermally broken down to form three major products: biochar (carbonaceous solid fraction), bio-oil (mainly composed of 15–30% w/w_{liquid} of wide variety of organic components and 70–85% aqueous pyrolysis liquid (APL)) [29], and syngas composed of non-condensable gases (CO, CO₂, CH₄, and H₂) [30,31]. Depending on the operating pyrolysis conditions (temperature, heating rate, residence time (RT)) the pyrolysis process can be classified as: slow, fast, and flash (**Fig. 1**) [28]. According to the typology of pyrolysis applied and the biomass treated, the distribution of biochar, bio-oil, and syngas will be different.

Due to its high calorific content, pyrolysis gas can represent an interesting supplementary energy source that can be further converted into heat or heat/electricity alone or mixed with biogas in boilers, engines, and other equipment [32]. Furthermore, the product gas can be transformed to methanol, dimethyl ether and other important chemical feedstocks *via* Fischer-Tropsch (FT) synthesis [25]. In parallel, the pyrolysis liquid presents a light- to dark-brown color consisting mainly of a mixture containing hundreds of organic molecules [33]. Often, pyrolysis liquid separates into an organic, light non-aqueous phase (bio-oil), and aqueous phase known as APL. The bio-oil produced is of primary interest as it can replace diesel in internal combustion engines to produce electricity [34]. On the contrary, APL currently has no apparent use and should be carefully managed due to its high organic strength that can cause

environmental burdens [32]. Biochar produced by pyrolysis due to its rich phosphorous content and physicochemical properties (porosity, surface area...), can be use in agronomic applications or for bioremediation [35,36]. Biochar can be also converted into activated carbons through physical and chemical activation processes to increase its physicochemical properties, especially, the accessible surface area and porosity. However, the use of activated biochar related to anaerobic digestion applications have been poorly investigated until yet [37,38].

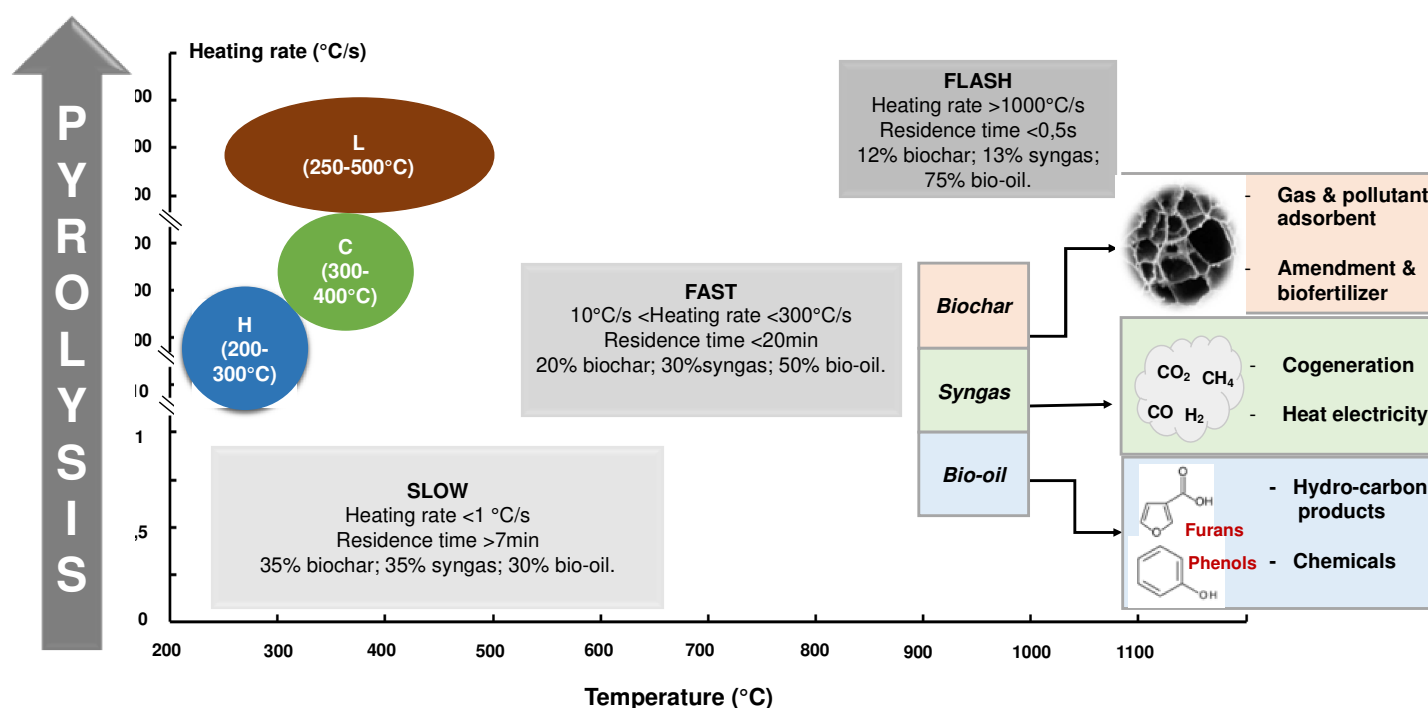


Fig. 1. Operating conditions, distribution of pyrolysis products for each pyrolysis type, and degradation temperature ranges of lignocellulosic compounds (Cellulose (C), Hemicelluloses (H), and lignin (L)). The scheme was inspired from Arnold et al. [39].

2.2 Anaerobic digestion process

Anaerobic digestion represents one of the most promising processes to convert diverse organic substrates (animal manure, food waste, municipal solid waste, and lignocellulosic biomass as agricultural waste) into energy carriers [40,41]. This process corresponds to a microbiological degradation of organic matter under anaerobic conditions leading to

stabilization of organic matter (digestate) and formation of biogas [42]. The anaerobic digestion process could be divided into two types according the feedstock dry matter (DM): wet state [43] and solid state [44], and it involves four successive stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis [45]. The anaerobic digestion biogas composed mainly of CH₄ (55–75%) and CO₂ (25–45%) [42]. The produced biogas is used as a source of renewable energy by combined heat and power (CHP) generation, and/or upgraded to bio-methane to be used as transportation fuels, or injected into natural gas grid [46,47]. In parallel, digestate is also generated through anaerobic digestion process and correspond to recalcitrant fraction or not degraded during the anaerobic digestion process [48]. To improve its transport and handling, digestate is generally separated into a liquid (rich in nutrients N and K) and a solid phase (rich in P and organic matter) through various technologies: screw press, vibrating sieve, and centrifuge [48,49].

3. Complementary of pyrolysis and anaerobic digestion processes

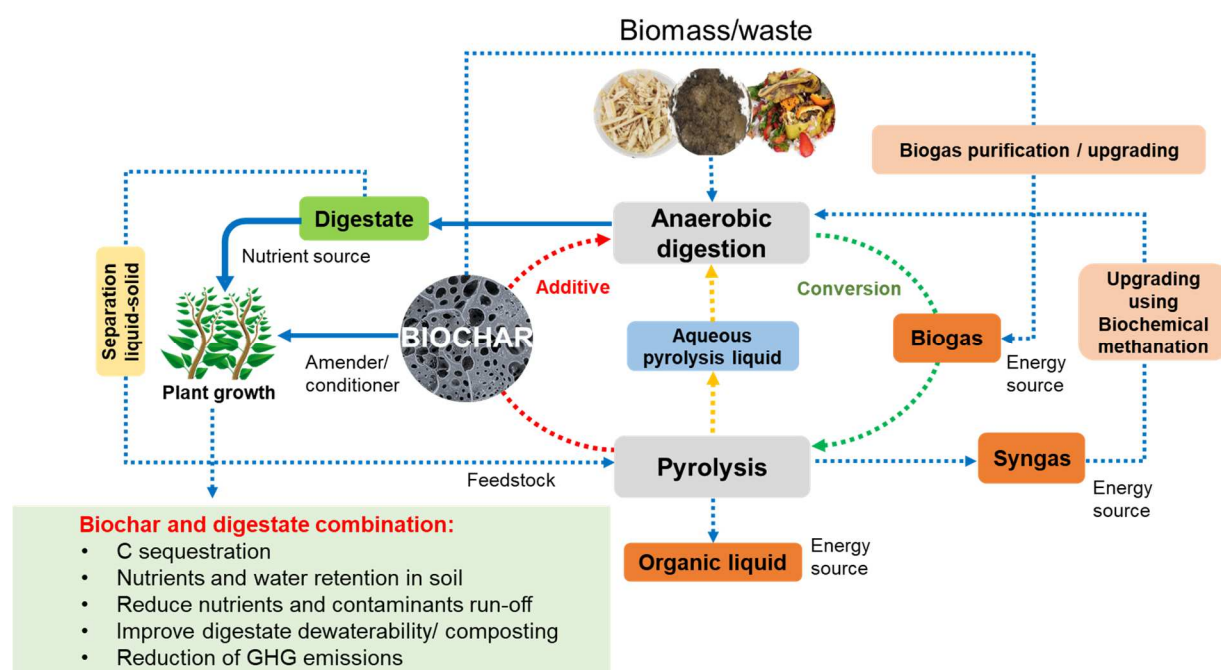


Fig. 2. Synergies between pyrolysis and anaerobic digestion processes by using biochar as central element. The scheme shows biochar interactions with anaerobic digestion by means of stabilizer/enhancer, biogas purification and/or upgrading and also for soil preservation.

Recently, the combination of anaerobic digestion and pyrolysis processes has started attracting attention among the scientific communities (567 publications between 2010 and 2019 in Science Direct using the keywords: coupling anaerobic digestion and pyrolysis). As mentioned by Fabbri and Torri [8], this low number of papers on a potentially interesting topic could be easily explained considering the relatively recent progress in commercialization of anaerobic digestion plants and pyrolysis/gasification technologies. Furthermore, in academia, thermochemical approaches are often parallel or competing with biochemical ones. Nevertheless, it is evident from literature data that these two processes can be associated according to a progressive integration (“industrial symbiosis”) approach, establishing a functional relationship. Recently, two main scenarios have been investigated with the pyrolysis process applied: 1) upstream of the anaerobic digestion process by biological upgrading of the liquid pyrolysis fraction (APL phase) through anaerobic digestion, and 2) downstream by converting the digestate recalcitrant solid phase [8,9,50]. Additionally, to improve energy recovery from initial biomasses, pyrolysis applied upstream or downstream of the anaerobic digestion process has revealed many benefits (**Fig. 2**) including:

- The treatment of a large range of substrates: easily biodegradable substrates are sent to anaerobic digestion whereas hardly biodegradable (wood, straw, solid fraction of digestate) are sent to pyrolysis.
- The dual symbiosis approach of coupling anaerobic digestion and pyrolysis is interesting because a pretreatment unit preceding anaerobic digestion could be avoided, as pyrolysis could handle the dry, poorly biodegradable materials that otherwise would have required hydrolysis before anaerobic digestion.
- The ex-situ or *in-situ* addition of biochar for inhibitors uptake.

- The *in-situ* addition of biochar for biofilms formation and stability performance improvement of the anaerobic digestion process
- The *ex-situ* or *in-situ* addition of biochar for biogas purification or upgrading.
- The aqueous bio-oil treatment by the anaerobic digestion process.
- The syngas upgrading through the biological methanation process (*in-situ* or *ex-situ*).
- The complementary in using digestate with biochar for agronomic issues.

3.1. Biogas purification and/or upgrading using biochar.

Biogas produced can be valorized as transport biofuel, injected in the national gas grid (biomethane after purification), or further converted into heat and electricity through a cogeneration (Combined Heat and Power – CHP) system [51]. Electricity can be sold to the public grid at a fixed rate dependent on the national policy and provide economic benefits to farmers [52]. According to the biogas valorization routes, some elements, such as H₂S and CO₂, should be removed from the biogas before use [53,54]. Biogas contains H₂S at concentrations ranging from 0.1 to 2.0% (v/v) depending on the types of substrate fed into the digester, which is a very corrosive gas [55]. If the valorization through the CHP system required only the removal of H₂S, upgrading biogas into biomethane would also require the removal of CO₂. Due to its physicochemical properties, biochar can be readily used to purify/upgrade biogas [16,18,19,56]. Recently, Xu et al. [16] investigated the performances of two biochars (produced from pig manure and sewage sludge) for their abilities to remove hydrogen sulfide (H₂S) from biogas. Biochar from pig manure exhibited a higher capacity for H₂S sorption than sewage sludge biochar in both dynamic and static systems [16]. Similarly, Kanjanarong et al. [18], introduced biochar (produced at 600 °C from a mix of woodchips/DG) in a continuously stirred tank reactor for treating sulfate-laden wastewater. With an H₂S concentration varied from 105 to 1020 ppm, the biochar was able to remove

more than 98% of H_2S at high pH (7.98) and high moisture content (80%–85%) conditions. Pelaez-Samaniego et al. [57] investigated the effect of pyrolysis temperature (from 300 °C to 600 °C) on the capacity of biochar to adsorb H_2S and demonstrated that the most attractive biochars were those produced at 600 °C for 60 min. Kanjanarong et al. [18] highlighted that the sorption of H_2S was mainly due to the presence of carboxylic and hydroxide radicals groups on the biochar.

In parallel, biochar has also been investigated for biogas upgrading into biomethane. Two main strategies by ex-situ use of biochar [56] or direct in-situ biochar addition during the anaerobic digestion process have been investigated [58]. Sethupathi et al. [56] investigated the effect of four different types of biochars produced from several biomasses (perilla leaf, soybean stover, korean oak, japanese oak) on the H_2S and CO_2 removal from a synthetic biogas composed of 0.3% of H_2S ; 40% of CO_2 , and 59.7% CH_4 . The various biochars exhibited different performances with the optimal one using perilla although all were efficient in reducing H_2S and CO_2 , whereas CH_4 was not adsorbed [56]. Direct addition of biochar in-situ during anaerobic digestion has also been investigated from biogas upgrading. Shen et al. [58] investigated the in-situ addition of biochar from corn stover for biogas upgrading. Besides to improve anaerobic digestion performances, biochar addition also conducted to produce high quality biomethane (> 90% CH_4 , < 5 ppb H_2S), with CO_2 removal until 86.3%. Therefore, the biochar improved process stability and considerably reduced energy/cost biogas upgrading. Similarly, Linville et al. [59] investigated biogas purification by in-situ addition of coarse and fine walnut shell biochar and demonstrated that fine biochar was the most efficient. It was demonstrated that addition of 0.96–3.83 g biochar g^{-1} VS_{added} fine walnut shell biochar into the digester resulted in biogas with 77.5%–98.1% CH_4 content by removing 40%–96% of the CO_2 compared with the control digesters at mesophilic and thermophilic temperature conditions [59]. To improve the purification efficiency and removal

of H_2S and CO_2 , biochar can be also used as a precursor of activated carbon [37,38]. Indeed, activated carbon has been found to be an effective carbonaceous sorbent for H_2S and CO_2 removal due to its high surface area and porosity [57,60,61]. For instance, Pelaez-Samaniego et al. [57] demonstrated that the activation of biochar with Na_2CO_3 impregnation improved the H_2S scrubbing capacity by 37% [57]. Finally, once the biochar is exhausted, it can be regenerated [60,62] or used as a nutrient-rich soil amendment for sulfur deficient soils [47,63].

3.2. Bio-oil and syngas valorization through integrated approach.

Pyrolysis liquid is composed of two phases: an organic phase (bottom phase consisting of 15–30% w/w of bio-oil) and an aqueous phase (85–70% w/w of bio-oil). The aqueous phase (APL) is formed by water, water soluble substances (C_2 – C_6 sugars, volatile fatty acids, oligomers), and slightly soluble substances (mainly phenols and furans) [29,64–66]. It is characterized by low heating value, high pH, high N-NH_3 concentration, and high water content and currently, no apparent use has been defined [32]. For this purpose, the use of APL in anaerobic digestion (Fig.2) has gained attention in recent decades [29,32,67].

Torri and Fabbri [29] investigated the methane potential of APL (from corn stalks pyrolysis at 400 °C for 10 min) in batch tests and reported a methane production corresponding to 34% of the theoretical value. Similarly, Hubner and Mumme [67] investigated the methane potential of various APL of pyrolysis liquid produced from solid digestate at 300 °C, 430°C, and 530°C and added in batch tests at four COD-based concentrations of 3, 6, 12, and 30 g L^{-1} . The lower concentrations (based on COD) of APL were converted during the anaerobic digestion process, while a concentration of 30 g L^{-1} caused process inhibition [67]. The temperature of pyrolysis process also influenced the APL anaerobic digestion. For instance, at a COD concentrations of 12 g L^{-1} , methane yield of 220 mL CH_4 g COD^{-1} was observed for APL produced at 330°C, followed by the 430°C with methane yield of 199 mL CH_4 g COD^{-1} ,

whereas the 530°C sample was inhibited with a methane yield of $38 \pm 10 \text{ mL CH}_4 \text{ g COD}^{-1}$ [67]. Similarly, Seyedi et al. [32] have investigated the potential toxicity of various pyrolysis liquids (derived from catalyzed and non-catalyzed pyrolysis at 800°C of anaerobically digested primary sludge and waste activated sludge) on the methane potential. Using the anaerobic toxicity assay, APL digester loading rates higher than 0.5 g COD L^{-1} for non-catalyzed and $0.10 \text{ g COD L}^{-1}$ for catalyzed APL were not sustainable due to toxicity [32]. $\text{NH}_3\text{-N}$ was not the main inhibitory constituent and other organics including 3,5-dimethoxy-4-hydroxybenzaldehyde, 2,5-dimethoxybenzyl alcohol, benzene, cresol, ethylbenzene, phenols, styrene, and xylenes as well as nitrogenated organics (e.g., benzonitrile, pyridine) generated methane production inhibition [32].

Currently, few studies have reported at continuous pilot scale the anaerobic digestion of APL [29,68]. Torri and Fabbri [29] demonstrated the difficulties to maintain stable semi-continuous anaerobic digestion of APL at daily addition higher than $15 \text{ g COD L}^{-1} \text{ d}^{-1}$. Similarly, Seyedi et al. [68] investigated the co-digestion of synthetic sludge with APL in semi-continuous assay and a daily methane production of 38 mL was reported which was comparable with the control treating only synthetic sludge (38.8 mL). For microbial communities, for both digesters, similar bacteria communities dominated by *Clostridium* genus and similar archaea communities dominated by *Methanosaeta* genus were observed. Nonetheless, a higher concentration of APL can generate instability in the anaerobic digestion process causing microbial communities to shift. Chen et al. [69] investigated the methane potential of hydrothermal liquefaction (HTL) wastewater. Interestingly, different microbial community compositions were observed from wastewater produced at different temperatures probably due to different organic compound concentrations. For low temperature of HTL, *Petrimonas*, which could degrade sugars, was the predominant in the anaerobic digestion of samples, whereas *Syntrophorhabdus sp.*, were more abundant in the anaerobic digestion of samples with higher HTL temperatures probably due to its capacity to degrade phenols

compounds [69]. If the anaerobic digestion process of APL of pyrolytic bio-oil is technically feasible at low concentration (0.06 to 0.3 g COD L⁻¹) [67], at higher concentration APL can be a hardly substrate to be degraded through anaerobic digestion process due to potential anaerobic digestion inhibitors in higher quantity (*i.e* furans, phenols) [32]. Indeed, the APL of bio-oil is rich in furans, polyphenols, and ammonium, which have been previously recognized as potential inhibitors of the anaerobic digestion process [42,70]. To overcome this limitation, APL can be co-digested with other biomasses [68] or biochar can be added in-situ during anaerobic digestion to improve the mitigation of the potential inhibitors such as furans and phenols compounds [29]. Indeed, Monlau et al. [71] investigated the use of biochar produced from anaerobic digestate to treat synthetic medium rich in furans compounds. At a concentration of 40 g L⁻¹ of biochar, more than 94% of 5-HMF and 99% of furfural were removed after 24 h of contact time in a synthetic medium, whereas sugar concentrations remained unchanged. During the trials, furfural adsorption was faster than 5-HMF [71]. Recently, Torri and Fabbri [29] demonstrated in BMP tests within-situ incorporation of biochar increased methane yield (60 % of theoretical value) with respect to pure APL (34 % of theoretical value) [29]. At the semi-continuous digestion scale, they demonstrated the benefits of adding biochar; with a daily input of 5 g d⁻¹ L⁻¹ of APL, where no intoxication signal was revealed and a relatively stable methane production was observed [29].

Besides bio-oil, syngas can also be upgraded into methane through biological methanation [72,73]. Several parameters such as cell concentration and microorganisms origin, reactor typology, pressure, pH, and temperature can influence the performances of the biological methanation process [74]. Biological methanation can be realized in a separate reactor or *in-situ* during anaerobic digestion, [74,75]. Indeed, such a configuration, even if it presents several technological barriers can improve the CH₄ amount produced by adding inorganic electron donors such as H₂ and CO, both which can be produced as syngas from solid digestate or other biomass [75,76]. Recently, some authors have investigated and simulated

the incorporation of syngas into biological methanation process for upgrading into methane [75,76]. Guiot et al. [75] investigated several aspects of syngas BM, including the dominant metabolic pathways used by anaerobic sludge when converting CO into methane and its tolerance level to syngas impurities. They demonstrated that direct methane generation from CO was very low, and CO was converted (CO acetogenesis) through an intermediate step mainly into acetate, which was further converted into methane. Furthermore, acetoclastic methanogens were the most sensitive to CO and syngas impurities, but higher impurity concentrations affected all activities [75].

From a technical point of view, one major limitation is the poor solubility of hydrogen and CO that limit the mass transfer between the gas phase and the microbial cell [72,77]. Indeed, in aqueous medium, CO₂ is around twenty-three times more soluble than hydrogen [74]. For this purpose, Guiot et al. [75] investigated in three bioreactor designs: a fluidized bed reactor (FBR), a closed-loop gas-lift reactor (GLR), and a bubble column reactor (BCR) the CO conversion efficiency. From all reactors investigated, the continuous stirred tank reactor was a sustainable option because it showed high specific bioactivity and gas-liquid mass transfer rates [75]. In parallel, Schwede et al. [72] recently investigated the immobilization of methanogenic archaea onto biochar (from green waste pyrolysis) to improve biological methanation processes. Results demonstrated that biochar was a suitable carrier for methanogenic archaea and that the biochar was successfully inoculated. During the first 24h, the conversion of H₂ with CO₂ and CO to CH₄ was fast and achieved more than 50% of the syngas components were converted to methane. However, CO was in majority utilized for formate/acetate formation rather than for methanogenesis conversion. Then, due to inhibition of methanogenesis by CO, it was demonstrated that methane production declined with increasing CO partial pressure [72]. Finally, Figueras et al. [20] have recently investigated the biomethanation of syngas by enriched mixed anaerobic consortium in a 10 L continuous

stirred tank reactor working at 4 bars and 55 °C [20]. Higher pressure reactor is a pathway to improve the gas-liquid transfer Syngas (40% CO, 40% H₂, 20% CO₂) biomethanation was performed successfully and methane productivity as high as 6.8 mmol_{CH₄}/L_{reactor}/h with almost full conversion of CO (97%) and H₂ (98%) was achieved [20].

3.3 Use of biochar to improve the performances of anaerobic digestion

3.3.1. As methane enhancer

In recent decades, several studies have investigated the supplementation of anaerobic digestion using specific materials to improve stability, performance, and microbial biofilm formation [23,78,79]. Activated carbon [80], zeolites [81], and more recently biochar [23,78] are among the materials tested. Due to their physicochemical properties, high porosity, and accessible surface area, biochar has been largely investigated recently to improve anaerobic digestion performance [23,82,83]. Carbonaceous material addition into anaerobic digestion systems was first investigated by Kumar et al. [80], who studied the effect of charcoal powder addition on mesophilic anaerobic digestion of cow slurry in continuous flow anaerobic digestion and batch anaerobic digestion. They demonstrated that adding 0.4 g g⁻¹ fresh matter of charcoal corresponded to an increase in methane yield of 34.7% and 17.4% for continuous flow anaerobic digestion and batch anaerobic digestion, respectively. However, the high production cost of charcoal limited its use in anaerobic digestion because the observed energy gain cannot cover the price of charcoal [82]. For this purpose, the use of biochar and/or activated biochar supplementation in the anaerobic digestion process have been investigated as described in **Table 2** [17,23,78,84]. Biochar can improve the anaerobic digestion stability and performances by means of: 1) buffering the medium due to alkaline pH of biochar [59]; 2) adsorption potential anaerobic digestion inhibitors (ammonium, phenols, limonene) [17,23]; 3) accelerating the transformation of macromolecular substances to dissolved ones [85]; 4) providing high nutrients (Ca, Mg, P, K) supplies [86]; 5) acting as microbial biofilm for bacteria and methanogens due to its high porosity of biochar [23,87,88]; and 6)

stimulating methanogens by promoting direct interspecies electron transfer among the bacteria and methanogens [88,89]. Acid-buffering issues represent a major reason for why anaerobic digestion is disruptive and generally occurs when the feedstock composition changes by total ammonia accumulation or by high organic loading rate (OLR) [23]. Biochar has proved its ability to adjust the alkalinity of the anaerobic digestion system. Linville et al. [59] found an improvement in alkalinity of CaCO_3 from 2800 mg L^{-1} to $4800 - 6800 \text{ mg L}^{-1}$ providing digestion better anaerobic digestion stability when walnut shell biochar was added inside the digester [59]. Promising results have been highlighted and biochar addition has been demonstrated to accelerate and improve the production of biomethane [17,23]. To date, several assays of biochar or activated biochars supplementation on anaerobic digestion have been tested: in wet anaerobic digestion [59,90], dry anaerobic digestion [88,89], and more recently in two stage hydrogen-methane production [87]. Li et al. [90] studied the effects of manganese oxide-modified biochar (corn straw, 600°C), which is known for its high heavy metal content, on anaerobic digestion of sewage sludge. The results showed that the addition of $0.09 \text{ g g}^{-1} \text{ DM}$ of modified biochar enhanced methane production by increasing the cumulative methane yield to 127.4% compared to the control. However the high biochar dose ($0.20 \text{ g g}^{-1} \text{ DM}$) exhibited the lowest cumulative methane production (**Table 2**), suggesting that microbial activities could be inhibited by high biochar doses [90]. Li et al. [90] demonstrated that manganese oxide-modified biochar incorporation into the anaerobic digestion process could improve the performances by increasing the buffering capacity and promoting the degradation of intermediate acids [90]. Besides the effect of the biochar dose, biochar addition with different sizes also showed a significant impact on the maximum methane production rate (Q_{max}). A study conducted by Cheng et al. [84], showed that the highest increase in Q_{max} was observed by using granular biochar ($2.2 \text{ g g}^{-1} \text{ VS}$), which was $12 \pm 6.1\%$ higher than the control (without biochar addition). It was highlighted that a linear relationship between Q_{max} and biochar surface area did not exist, but in general, Q_{max}

decreased with increasing surface area, suggesting that differences in adsorption capabilities of biochar may be one explanation for these observed discrepancies [84]. Pan et al. [85] investigated the impact of different biochar origins (wheat straw, fruitwood, chicken manure) produced at various pyrolysis temperatures (350, 450, and 550 °C) on the anaerobic digestion of chicken manure [85]. The best results were observed by addition of fruitwood biochar (550 °C), with an increase of 69% of the cumulative methane yield compared to the control, with a methane potential of 294 L CH₄ kg⁻¹ VS_{added} compared to 174 L CH₄ kg⁻¹ VS_{added} for the control. Interestingly, fruitwood biochar produced at 550 °C exhibited the larger surface area (202 m² g⁻¹) and the highest total ammonia nitrogen reduction capacity (TAN concentration of 3.34 g L⁻¹ compared to 4.48 g L⁻¹ for the control), which can explain the improvement in methane potential [85]. Finally, Wu et al. [91] investigated the effect of magnetic biochar addition prepared with different FeCl₃:rice straw ratios on anaerobic digestion organic fraction of municipal solid waste (OFMSW). The results showed that the methane production with magnetic biochar prepared with 3.2 g FeCl₃:100 g rice straw ratio increased by 11.69 % compared to control (without biochar addition); these findings were justified by the selective enrichment of microorganisms on magnetic biochar that participate in anaerobic digestion. However, high Fe content in magnetic biochar (33 g FeCl₃:100 g rice straw ratio) led to a decrease in methane production by 38.34% compared to the control due to iron oxide electron competition [91]. Jang et al. [86] studied the effect of adding biochar (derived from dairy manure) on methane production at different temperatures conditions (psychrophilic; mesophilic; thermophilic). In comparison with the anaerobic digestion without biochar addition, the methane yield with 10 g L⁻¹ of biochar increased by 26.47% in psychrophilic, 24.90% in mesophilic, and 24.69% in thermophilic conditions. Furthermore, the addition of biochar lowered the concentration of total VFAs and propionic acid and shortened the lag phases of anaerobic digestion for all the temperatures investigated [86]. Mumme et al. [92] also investigated the effect of biochar addition on biogas production and ammonia inhibition.

Kinetic and microbiota analyses revealed that biochar can prevent mild ammonia inhibition ($2.1 \text{ g TAN kg}^{-1}$). Stronger inhibitions ($3.1\text{--}6.6 \text{ g TAN kg}^{-1}$) were not mitigated. Biochar addition to the anaerobic digestion process has also been investigated in continuous assays [93–96]. Shen et al. [93] investigated the biochar incorporation in anaerobic digestion process in a semi-continuous assay treating sewage sludge. Interestingly, substrate utilization, methane productivity, and process stability was improved by biochar addition in the anaerobic digestion process [93]. Corn stover biochar addition enhanced the methane production rate by up to 37% compared to the control [93]. Microbial communities were also investigated, and corn stover addition ($3.4\text{--}3.6 \text{ g biochar g}^{-1} \text{ VS added}$) promoted the abundance of Clostridia and Methanosarcina [93]. Similarly, Sun et al. [88] investigated the impact of biochar addition in microbial communities shift during mesophilic and thermophilic anaerobic digestion process. Interestingly, methanogen community distribution was more intensively influenced by the biochar supplement in mesophilic than thermophilic conditions. Segun-Giwa et al. [94] also investigated the addition of biochar (0.25 g day^{-1}) on long-run anaerobic digestion of food waste. At an OLR of $2 \text{ g VS L}^{-1} \text{ day}^{-1}$, the reactor without biochar addition led to VFA accumulation and failed whereas the reactor fed with biochar remained stable up to an OLR of $5.4 \text{ g VS L}^{-1} \text{ day}^{-1}$ [94]. Wanbugu et al. [95] also investigated the addition of biochar (from waste wood pyrolysis at $600 \text{ }^{\circ}\text{C}$) on the anaerobic digestion of food waste in up-flow anaerobic sludge blanket (UASB). The average COD removal efficiencies of the control and the biochar-amended reactor were 47% and 77% at an OLR of $6.9\text{--}7.8 \text{ g COD L}^{-1} \text{ day}^{-1}$, respectively. Cumulative biogas production and methane composition were improved in the UASB fed with biochar and after 34 days the concentration of VFAs were, respectively, 1600 mg L^{-1} and 350 mg L^{-1} for UASB without and with biochar addition [95]. Future research should be led to better understand biochar–microbe interactions and the effects in continuously-fed anaerobic digesters.

Biochar addition to dry anaerobic digestion has also revealed positive results [82,88,89]. Sun et al. [88] studied on dry anaerobic digestion of beer lees the effect of biochar addition (2 g L⁻¹, 6 g L⁻¹, 10 g L⁻¹, and 14 g L⁻¹) at both mesophilic and thermophilic conditions. Cumulative methane production rate and yield with 10 g L⁻¹ of biochar were improved by 82.9% and 82.6% at mesophilic conditions and 47.2% and 46.8% at thermophilic conditions in comparison with the control [88]. Nonetheless, high concentration of biochar (14 g L⁻¹) led to a decrease in methane production rate and yield. Similarly, Meyer-Kohlstock et al. [82] investigated the influence of biochar addition (wood residue at 650 °C) in a mesophilic solid-state (batch) fermentation with percolation. The results showed that methane production increased by 5% with a biochar addition of 5% (based on organic DM biochar to bio-waste), while an addition of 10% showed an increase of 3%, suggesting that high biochar concentration can reduce efficiency. Finally, Capson-Tojo et al. [89] recently investigated the impact of biochar and FeCl₃ addition on dry continuous pilot reactors. It was demonstrated that higher methane production rates (up to 1.75 L L⁻¹ day⁻¹) and lower concentrations of acetate and propionate were achieved when biochar and trace elements (FeCl₃) were added [89]. Finally, biochar addition (biochar from pyrolysis of pine sawdust at 650 °C) was investigated recently in the performances of two stage hydrogen and methane production treating aqueous carbohydrates food wastes at mesophilic conditions (35 °C) using bench-scale bioreactors [87]. Interestingly, biochar addition improved hydrogen yield by 31.0% and methane by 10.0% and reduced the lag in the two phases by 36.0% and 41.0%, respectively. Biochar addition promoted the VFA generation during dark fermentation process and subsequent VFAs degradation in methane production [87]. These different studies have demonstrated the benefits of using biochar in anaerobic digestion even if certain concentrations should not be exceeded to avoid an opposite effect [87,88].

Table 2

Selected reported studies of in-situ addition of biochars in anaerobic digestion systems and their effects on biomethane production.

Anaerobic digestion substrate	Anaerobic digestion process conditions	Biochar feedstock/ carbonaceous material	Pyrolysis conditions/ Activation	Biochar Dose	Cumulative / yield CH ₄ -control	Cumulative /yield CH ₄ -biochar	Improvement rate %	Ref.
Cow slurry	Mesophilic batch at 35 °C	Powder charcoal (Darco G60)	-	0.4 g g ⁻¹ fresh matter	-	-	+ 17.4	[80]
	Semi continuous digesters at 35 °C		-		-	+ 34.7		
Swine sludge and wastewater	Glass serum bottles at 30 °C	Pine wood	Pyrolysis 980 °C	2.2 g g ⁻¹ VS (granular loading)	-	-	+12 ± 6.1	[84]
				9.6 g g ⁻¹ VS (powder loading)	-	-	- 57 ± 3	
Dewatered sewage sludge	Mesophilic batch wet anaerobic digestion at 35 °C	Corn straw	Pyrolysis 600 °C RT of 120 min HR of 25 K min ⁻¹ /Soaking in KMnO ₂ MR of biochar: KMnO ₂ =10:1	0.09 g g ⁻¹ DM	~53.3 L kg ⁻¹ VS	121.2 Lkg ⁻¹ VS	+ 127.4	[90]
				0.20 g g ⁻¹ DM		~37.0 L kg ⁻¹ VS	- 30.6	
Chicken manure	Mesophilic batch wet anaerobic digestion at 35 °C	Fruitwood biochar	Pyrolysis 550 °C RT of 2 h	5 g g ⁻¹ DM	174 L kg ⁻¹ VS	294 L kg ⁻¹ VS	+43.1	[85]
Citrus peel	-	coconut shell, wood, and rice husk	-	1 g g ⁻¹	165.9 mL CH ₄ g ⁻¹ VS	178.2 mL CH ₄ g ⁻¹ VS	+ 12.3	[97]
				3 g g ⁻¹		186.8 mL CH ₄ g ⁻¹ VS	+ 20.9	
Dairy manure	Psychrophilic serum bottles at 20 °C	Dairy manure	Pyrolysis 350 °C RT of 3 h HR of 10 °C min ⁻¹	10 g L ⁻¹	-	368.2 mL CH ₄ g ⁻¹ VS	+ 26.5	[86]
	Mesophilic serum bottles at 35 °C			10 g L ⁻¹	-	474.6 mL CH ₄ g ⁻¹ VS	+24.9	
	Thermophilic serum bottles at 55 °C			10 g L ⁻¹	-	431.3 mL CH ₄ g ⁻¹ VS	+24.7	
Beer lees	Mesophilic batch dry anaerobic digestion at 35 °C	Cow manure	Pyrolysis 500 °C RT of 4 h HR of 100 °C h ⁻¹	10 g L ⁻¹	220.1± 7.7 L kg ⁻¹ VS	401.8 ± 7.7 L kg ⁻¹ VS	+82.6	[88]
				14 g L ⁻¹		358.1±4.2 L.kg ⁻¹ VS	+62.7	
	Thermophilic batch dry anaerobic digestion at 55 °C			10 g L ⁻¹	310.4 ± 9.2 L kg ⁻¹ VS	456.8 ± 7.7 L kg ⁻¹ VS	+47.2	
				14 g L ⁻¹		416.9±8.9 L.kg ⁻¹ VS	+34.3	
Aqueous carbohydrates food waste stimulated from white bread	Bach wet two-phase anaerobic digestion (H ₂ and CH ₄ production) at 35 °C	Pine sawdust	Pyrolysis 650 °C RT of 20 min	8.3 g L ⁻¹	1070.0 ± 3 mL/L	1136.6 ± 7.0 mL/L	+6.2	[87]

Organic fraction of municipal solid waste (OFMSW)	Mesophilic batch wet AD	Rice straw	Pyrolysis 500 °C RT of 2 h / the substrate was suspended in FeCl ₃ solution	0.5 % (w/w) of 3.2g FeCl ₃ :100g rice straw biochar	174.2 L kg ⁻¹ VS	194.6 L kg ⁻¹ VS	+11.7	[91]
				0.5 % (w/w) of 33 g FeCl ₃ :100 g rice straw biochar		107.5 L kg ⁻¹ VS	-38.3	
Bio-waste (OFMSW)	Mesophilic solid-state batch at 40 °C	Clean forestry wood residue (Holm Oak)	Pyrolysis 650 °C	5% VS	245 N L kg ⁻¹ VS	257.4 N L kg ⁻¹ VS	+5.1	[82]
				10% VS		252.3 N L kg ⁻¹ VS	+3	
Food wastes	Continuous pilot scale reactors at 37 °C	Slow-pyrolyzed wood biochar	-	From 10 to 50 g L ⁻¹ with 100 mg Fe L ⁻¹	~ 200 mL CH ₄ g ⁻¹ VS	350 m L CH ₄ g ⁻¹ VS	+ 75	[89]
N-rich substrates	Mesophilic batch, glass syringes at 42 °C	Mixture of paper sludge and wheat husks	Pyrolysis 550 °C RT of 20 min	-	-	-	-8.5	[92]
		Wheat straw DG	Hydrothermal carbonization 230 °C RT of 6 h		-	-	+31.7	
wastewater sludge	Mesophilic Batch glass bottles at 38 °C	waste forest industry wood (a mix of pine, spruce, and cedar)	Pyrolysis 450–550 °C	0.8-3.7 g g ⁻¹ VS	-	-	+ 192–461	[98]
Sewage sludge	Yearlong semi-continuous thermophilic two stage anaerobic digestion at 55 °C	Corn stover	Pyrolysis & gasification 500–850 °C	0.25-1 g day ⁻¹	-	-	+ 36.9	[93]
		Pine	-		-	-	+ 16.6	
Chicken manure	Semi-continuous stirred tank reactors at 35 ± 1 °C	Orchard waste wood	Pyrolysis 550 ± 50 °C	4.97 % TS	-	-	+33 (OLR of 0.625 g VS L ⁻¹ day ⁻¹) + 36 (OLR of 3.125 g VS L ⁻¹ day ⁻¹) + 32 (OLR of 6.25 g VS L ⁻¹ day ⁻¹)	[96]

RT: Residence Time; HR: Heating rate

3.3.2. As a tool for AD inhibitors removal

Biochar represents an exceptional sustainable product for remediation and adsorption; it has been investigated as a low-cost sorbent for various organic molecules and inhibitors as summarized in **Table 3**. Ahmad et al. [99], reported that the adsorption performances of biochar depends mainly on some physicochemical proprieties such as surface area, porosity, alkalinity, ion-exchange capacity, and surface functional groups. To deal with the high adsorption capacity of commercial activated carbon, activation processes represent a promising strategy to improve biochar adsorption performances [99,100].

Adsorption or inhibitors mitigation can be done ex-situ or directly in-situ of the anaerobic digestion process. Phenol and furan derivatives are potential anaerobic digestion inhibitors and has been carried out in many adsorption studies using biochar as sorbent. A study showed that biochar produced from pistachio shells removed 51% of phenols through hydrophobic sorption from an aqueous solution of 200 mg L⁻¹ [101]. Mohammed et al. [102] reported that pine fruit shell biochar removed more than 26.74 mg phenols g⁻¹. Li et al. [103] reported a high adsorption capacity of 169.0 mg phenols g⁻¹ through chemical interactions and pore expansion using chemically activated biochar (with KHCO₃ and urea) produced from bamboo. Monlau et al. [71] studied the adsorption of furfural and 5-HMF from lignocellulosic hydrolysates with a concentration of 1 g L⁻¹ using biochar derived from anaerobic digestion residues.

Table 3.

Selected reported studies of *ex-situ* addition of biochars as adsorbent for inorganic and

Inhibitor group	Name/origin	Biochar feedstock	Inhibitor concentration	Removal performance	Removal mechanisms	Ref.
Polyphenol	Phenol	Pistachio shells	100 mg L ⁻¹	51%	Hydrophobic sorption	[101]
	Phenol	Pine fruit shells	20–100 mg L ⁻¹	26.7 mg g ⁻¹	Physisorption mechanisms	[102]
	Phenol	Oil palm frond chips	40-260 mg L ⁻¹	62.9 mg g ⁻¹	Surface adsorption and pore-filling	[104]
	Phenol	Bamboo/ activated with KHCO ₃ and urea	1000 mg L ⁻¹	169.0 mg g ⁻¹	chemical adsorption and physical adsorption through pore expansion	[103]
Ammonium	NH ₄ ⁺ from biogas slurry	Biogas residue	0–855 mg L ⁻¹	48.9 mg g ⁻¹	Ion exchange mechanism	[105]
	NH ₄ ⁺ from NH ₄ Cl	HNO ₃ ⁻ impregnated corncob	10–100 mg L ⁻¹	22.6 mg NH ₄ ⁺ -N g ⁻¹	Sharing or exchange electrons	[106]
	NH ₄ ⁺ from NH ₄ Cl	Orange peel	10–100 mg L ⁻¹	4.7 mg g ⁻¹	surface complexation, cation exchange and electrostatic attraction	[107]
	NH ₄ ⁺ from NH ₄ Cl	Pineapple peel	10–100 mg L ⁻¹	5.6 mg g ⁻¹	surface complexation, cation exchange and electrostatic attraction	[107]
	NH ₄ ⁺ from NH ₄ Cl	Caragana korshinskii biomass/ ultrasonic activation	0–100 mg L ⁻¹	26.3 mg g ⁻¹	Specific surface area and electrostatic adsorption	[108]
	NH ₄ ⁺ from NH ₄ Cl	Oil palm shells/chemical activation with K ₂ CO ₃	100 mg L ⁻¹	1.49 mg g ⁻¹	surface complexation and electrostatic interaction	[109]
Furan derivatives	Furfural	Solid digestate	1 g L ⁻¹	99%	π - π stacking interaction	[71]
	HMF	Solid digestate	1 g L ⁻¹	94%	π - π stacking interaction	
	HMF	cotton and linen residues/steam activated	1 g L ⁻¹	99%	-	[110]
	Furfural	Bamboo/heat treated (800°C)	10 g L ⁻¹	100 %	Pore filling due to high specific surface area	[111]

organic contaminants.

The authors reported that more than 94% and 99% of 5-HMF and furfural were removed respectively [71]. Same results were founded in the studies conducted by Klasson. K et al. [110] and Li et al. [111]. Biochar has also exhibited great abilities in adsorbing ammonium

[112]. Ammonium (NH_4^+) in groundwater and surface water can caused severe environmental pollution and health issues among people [113] and represents one of the anaerobic digestion inhibitor, which the threshold inhibition level was reported to be varied between 1.5 to 2.5 g/L [114–116]. Recent studies confirmed that the adsorption method using biochar and activated carbon is considered to be an economic and effective strategy for removing NH_4^+ [117–119]. T.M. Vu et al. [106] have used a modified corncob-biochar with HNO_3 and NaOH for removing NH_4^+ from a synthetic water (ammonium concentration from 10 to 100 mg L^{-1}). The highest adsorption capacity of 22.6 mg $\text{NH}_4^+\text{-N g}^{-1}$ was obtained using modified biochar with NaOH 0.3M and HNO_3 6M [106]. The authors reported that the ammonium adsorption depended strongly on the pH [106]. Yu et al. [105] investigated the efficiency of biochar derived from biogas plant digestate as adsorbent for ammonium removal on artificial wastewater solution and the highest adsorption capacity of 48.89 mg $\text{NH}_4^+\text{-N g}^{-1}$ biochar was achieved [105]. It was highlighted that the ash content in biochar played an important role in ammonium adsorption due to the significant role that SiO_2 compounds played as adsorption sites in the process [105]. Moreover, a recent study of Wang et al. [108] evaluated the potential adsorption of an activated biochar by ultrasonic activation produced from *Caragana korshinskii* biomass at different temperatures (450, 500 and 650 °C), for removing ammonium from a synthetic solution (concentration from 0-100 mg L^{-1}). The results showed that the highest adsorption capacity of 26.3 mg $\text{NH}_4^+\text{-N g}^{-1}$ was obtained using the ultrasonic (time was 480 min, the frequency was 45 kHz, and the power was 700W) activated biochar produced at 500 °C. They reported that the ultrasonic activation induced the pore formation, which increase the specific surface area through cavitation corrosion and micro-acoustic flow mechanism [108]. Inhibitor mitigation can also be performed directly in-situ of the anaerobic digestion process. Biochar addition has also been reported as efficient in-situ adsorption of potential inhibitors to the anaerobic digestion process such as D-limonene present citrus peel [97]. In the experiment, three different kinds of biochars were used: coconut shell, wood, and

rice husk. The cumulative methane production in incubations containing biochar and citrus peel ranged from 178.2 (ratio 1:1 citrus peel/biochar) to 186.8 mL CH₄ g⁻¹VS (ratio 1:3 citrus peel/biochar), while citrus peel only produced 165.9 mL CH₄ g⁻¹VS. Furthermore, the authors noted high methanogenic activity within the digester due to the biochar ability to absorb limonene [97]. Finally, Torri and Fabbri [29] demonstrated that biochar addition in situ enables anaerobic digestion of aqueous phase (APL) and mitigation of inhibitors compounds. Batch tests exhibited poor performance in anaerobic digestion of APL, which underlined the inhibition of biological process. Biochar addition increased yield of methane (60 ± 15% of theoretical) with respect to pure APL (34 ± 6% of theoretical) and improved the reaction rate [29]. The adsorption and removal of anaerobic digestion inhibitors such as polyphenol, furans, and ammonium has become a key area of research interest these last decades. It appears that using biochar from renewable biomasses and wastes to adsorb various contaminants and molecules has emerged as a potential cost-effective alternative of conventional activated carbon. Indeed, the price of biochar was estimated at 246 \$ US t⁻¹, corresponding to about 1/6 of commercially activated carbon (~US \$1500 t⁻¹) [99].

4. Agronomic benefits of dual use of biochar and digestate

4.1 Nomination and history of using biochar in agriculture

Biochar origin was first discovered in the Amazon region and known as Terra Preta de Indio, where it was created through the use of slash and burn techniques [99,120,121]. Nowadays, around 10% of the Amazon is occupied by this soil typology, characterized by its high fertility in comparison to others soils [122]. The International Biochar Initiative (IBI) defines biochar as a “solid material obtained from biomass in an oxygen limited environment” [123]. On another hand, the European Biochar Certificate (EBC) defines biochar as “a heterogeneous substance rich in aromatic carbon and minerals produced from sustainable biomass under controlled conditions with clean technology and used for any purpose that does not involve rapid mineralization of biochar to CO₂” [124]. Therefore, the term biochar is

generally associated with high stable carbon content contributing to carbon sequestration [125] and improving soil properties due to its physicochemical properties [126–128]. Until yet, biochar exhibited several advantages, among other 1) improving soil nutrient dynamic and exchange with the soil environment [126], 2) enhancing water retention and soil micro-organisms biofilm formation [126,127], 3) retaining organic and inorganic contaminants in soil [129,130], 4) improving the physicochemical properties of soil [127], 5) reducing GHG especially CO₂ emissions [131,132], 6) improving plant growth and quality [132,133], and enhancing carbon sequestration due to its high stable carbon content [128,133,134].

4.2 Effects of combining biochar and digestate on plant nutrition and productivity.

Co-application of biochar and digestate has been tested on plant growth performances under both controlled (growth chambers and greenhouses) and field conditions on different crops such as maize [135–137], winter rye [138], winter wheat [139], taro [140], geranium [141], Chinese melon [142], and radish [143] (**Table 4**). Rhizosphere biological modifications, notably the microbial communities and functions, also occurred in response to dual application of biochar and digestate and the co-application resulted in reduced microbial respiration along with increased soil carbon sequestration [144–147] and reduced GHG emissions [147,148]. In **Fig. 3**, several positive effects on coupling biochar and digestate illustrate the likely benefits of combining them for agronomic purposes. Biochar could not be considered as a nutrient-supplying source, but its potential in crop fertilization has been demonstrated combined with mineral fertilizers or other types of organo-fertilizers like compost [149,150] and digestate [3,139,145] (**Fig. 3**). Indeed, both digestate and biochar can have significant beneficial effects when co-applied, owing to their complementary agronomic and biostimulant properties [3,139,145] with positive consequences on soil fertility and nutrient availability, which presumably would stimulate both plant and microbial functional traits responsible for enhanced fertilizer use efficiency [151]. In addition, liquid digestates are

characterized as nutrient-rich (ammonium, potassium, etc.), whereas solid digestates usually contain high phosphorus and fibers [142]. Thus, combining both solid and liquid digestate would provide adequate nutrient elements and may, in some cases, improve soil properties and replace/minimize use of mineral nutrients fertilizers [145,146]. For N supply, which is the most important macronutrient for crop productivity, most digestates are brought to the soil at N concentrations varying from 80 to 275 kg N ha⁻¹ [152]. Regardless of the soil nature and plant species, a rate up to 170 kg N ha⁻¹ is generally applied, which corresponds to the maximum amount initially allowed by the EU Nitrate Directive for spreading in vulnerable zones [153].

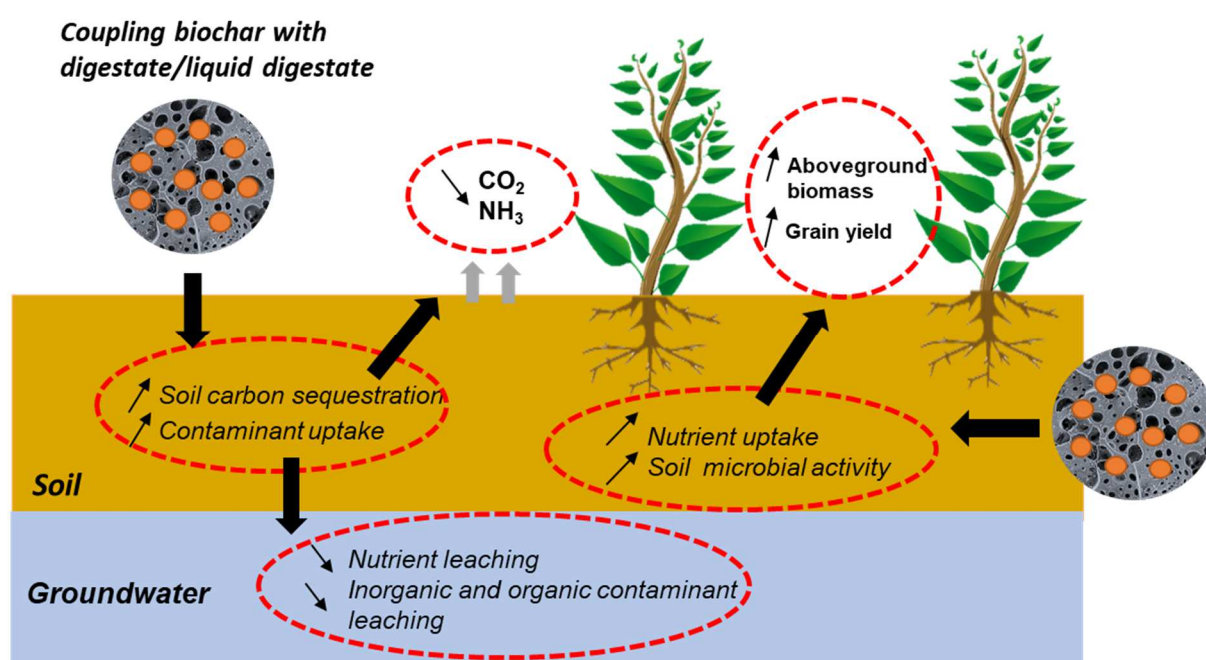


Fig. 3. Illustration of advantages of combining biochar and digestate for agronomic purposes. Downward arrows indicate an increase and upward arrows indicate a decrease.

Although biochar and digestate are known to be agronomically beneficial, effects of coupling biochar and digestate on crop yields are still inconsistent among different studies (Table 4). Positive results of such a coupling were highlighted many times in maize, Taro (*Colocasia esculenta*), chinese Melon (*Cucumis melo* L.), tomato (*Solanum lycopersicum* L.) and

marigold (*Tagetes erecta* L.) plants [135,140,142,154]. For instance, Glaser et al. [135] demonstrated that application of biochar and digestate (at rate of 1 and 40 t ha⁻¹ and digestate at 200 kg N ha⁻¹) on maize produced better yields (yields and plant nutrition) compared to pure digestate. At a concentration of 40 t ha⁻¹ of biochar, the co-application of digestate improved maize yield by 42% compared to untreated plants without biochar addition [135]. Similarly, Bouaravong et al. [140] also demonstrated the positive effect of coupling biochar (2 kg m⁻²) with different digestate doses (0 to 100 kg N ha⁻¹) on taro plant growth [140] with the observation that co-application of digestate at levels up to 100 kg N ha⁻¹ with biochar resulted in linear increase in biomass yield, higher crude leaf protein content, and improved soil fertility as measured by pH, water-holding capacity, and N content. For instance, at a digestate dose of 100 kg N ha⁻¹, biomass yield exhibited 1027 g m⁻² compared to 609 g m⁻² obtained with biochar alone [140]. Similarly, Elbashier et al. [142] make a comparison of industrial fertilizers, biochar and digestate amendments on photosynthesis rate and growth parameters of the Chinese melon (*Cucumis melo* L.) under saline and no saline water conditions. This study found that combination of biochar and digestate resulted in a more productive fertilization specifically under saline irrigation conditions compared to mineral fertilizer. Under saline water irrigation, combination of biochar and digestate improved plant height by 36, 22, and 54% compared to biochar, digestate, and unfertilized soil, respectively [142]. Finally, Tayibi et al. [155] have investigated the effect of coupling biochar produce from solid digestate at 500 °C (50 and 100 t ha⁻¹) with liquid digestate (170 Kg N ha⁻¹) on nutrient leaching (NH₄⁺, NO₃⁻, PO₄³⁻, K⁺, and Na⁺) and wheat plants growth. This study found that the coupling of biochar and liquid digestate have only decrease the leaching of NO₃⁻ by 82% and 91%, at 50 and 100 t ha⁻¹ respectively, compared to soil treated only with liquid digestate [155]. In addition, this combination demonstrated that biochar application at 50 t ha⁻¹ did not exhibit a negative impact on the relative seed germination of wheat plants and improved aerial dry biomass production up to 27.5%, compared to soil with only liquid

digestate addition. In another study, same tendency was noticed on the aerial dry biomass of tomato plants growth, which was improved by 25% under biochar application at 25 t ha⁻¹ with liquid digestate (170 kg N ha⁻¹), compared to the treatment with liquid digestate alone [156].

In addition to digestate role in improving soil and plant N status, adding biochar to anaerobic digestate has also been demonstrated to improve soil biological activity, antioxidant capacity, and carbon sequestration [141,142], and that both may be considered suitable fertilizing substrates or soil conditioners, especially for acidic soils [3,143,148]. Another study by Marchetti et al. [145] revealed the benefit of coupling biochar from animal manure (10 t ha⁻¹) with digestate (170 kg N ha⁻¹) demonstrating an improved organic matter stock and available P for crops, without modifying N availability.

Table 4.

Selected publications studied the potential effect of combining biochar with digestate on plant nutrition and productivity.

Biochar rate	Fertilizer rate	Plant choice	Impact on plant nutrition availability and yield	Ref.
40 t ha ⁻¹	Digestate at 200 kg N ha ⁻¹	Maize	<ul style="list-style-type: none"> Improvement of maize yield by 42 % 	[135]
20 t ha ⁻¹	Digestate doses 100 kg N ha ⁻¹	Taro	<ul style="list-style-type: none"> Improvement of crude leaf and protein content, water holding capacity and N content. Improvement of biomass yield by 1027 g m⁻² compared to 609 g m⁻² obtained with biochar alone. 	[140]
180 t ha ⁻¹	Digestate at 500 mL/pot equivalent to NPK: 120-150-150 kg ha ⁻¹	Chinese melon (Cucumis melo L.)	<ul style="list-style-type: none"> Improvement of the plant height by 36, 22, and 54% compared to biochar, digestate, and unfertilized soil, respectively, under saline water irrigation 	[142]
50-100 t ha ⁻¹	Liquid digestate at 170 kg N ha ⁻¹	Wheat	<ul style="list-style-type: none"> Decrease of nitrate leaching (NO₃⁻) by 82% and 91%, at 50 and 100 t ha⁻¹ respectively, compared to soil treated only with liquid digestate. Improvement of aerial dry biomass production up to 27.5%, at biochar application of 50 t ha⁻¹, compared to soil with only liquid digestate addition. 	[155]
25-50 t ha ⁻¹	Liquid digestate at 170 kg N ha ⁻¹	Tomato	<ul style="list-style-type: none"> Improvement of aerial dry biomass production up to 25%, at biochar application of 25 t ha⁻¹, compared to 	[156]

			soil with only liquid digestate addition.	
10 t ha ⁻¹	Digestate at 170 kg N ha ⁻¹	-	<ul style="list-style-type: none"> Improvement of the available P for crops and the organic matter stock: the organic matter (C_{org}) decreased from 0.5 to 7.4 g C kg⁻¹ using different type of biochars. 	[145]
30 t ha ⁻¹	Digestate	Radish (<i>Raphanus Sativus</i> L.)	<ul style="list-style-type: none"> No effect of co-application of biochar and digestate on dry biomass compared to digestate alone 	[143]
20 t ha ⁻¹	Digestate at 250 kg N ha ⁻¹	Wheat	<ul style="list-style-type: none"> No effect on wheat growth and grain yields, compared to soil with digestate alone. 	[139]

Although no negative impact has been reported yet about coupling digestate and biochar on crop performances, few studies have reported no significant effect under this combination [138,139,143]. For example, Sekar et al. [143] investigated radish cultivation (*Raphanus Sativus* L.) in response to co-application of biochar and digestate in a fifteen month field experiment (**Table 4**). It was found that adding digestate alone significantly improved radish dry biomass (413 g compared to 31 g in soil alone) and that the effect of biochar (30 t ha⁻¹) was not significant as compared to digestate alone [143]. Likewise, Udall et al. [139] reported no effect on wheat growth and grain yields in response to the combination of biochar (20 t ha⁻¹) and digestate (250 kg N ha⁻¹) compared to digestate alone.

4.3 Effects of combining biochar and digestate on soil microbial activity.

Reduced microbial respiration in soil along with improved soil carbon sequestration was measured due to the addition of biochar to digestate [144–147]. Either applied alone or combined with digestate, biochar tended to enrich the C sink of the soil through a reduction in soil respiration due to a reduction in dissolved organic carbon owing to a lower accessibility of microbes to dissolved organic carbon [144]. It seems that the addition of biochar to digestate reduced soluble compounds due to the presence of residual phenol compounds in biochar that exhibit antioxidant capacity in soil microbial communities, and other activities (CO₂ production and dehydrogenase activity) [146]. Similarly, Mukherjee et al. [144]

investigated microbial respiration under a mixture biochar and digestate with the conclusion that addition of digestate to soil led to a significant increase in CO₂ evolution compared to untreated soil. This may be due to a higher proportion of easily degradable C in digestate as opposed to a lower CO₂ release (up to 11-fold difference) observed in soil added with a mixture of digestate and biochar [144]. Interestingly, this effect was also observed when only 1% (w/w) of biochar was mixed with digestate in soil, suggesting that even a low concentration biochar lowered the microbial accessible dissolved organic carbon [144]. Conversely, other findings by Marchetti et al. [145] found no significant effects when biochar (from wood chips and swine manure solids) was added to soils previously amended with slurry digestate. In this study, the digestate-amended soils had an important CO₂ emission rate (1188 mg CO₂ kg⁻¹ dry soil) compared to 874 mg measured in untreated soils. In parallel the addition of biochar did not significantly reduce CO₂ emission, with values of 1059 and 959 mg CO₂ released kg⁻¹ in dry soil for both wood chips and swine manure solid biochar [145]. These findings were also in accordance with the results of the study conducted by Tayibi et al. [156], which demonstrated that adding biochar (25 t ha⁻¹) with liquid digestate (170 Kg N ha⁻¹) did not impact the CO₂ emission rate compared to the application of liquid digestate alone [156].

4.4 Effects of combining biochar and digestate on GHG and soil contaminants reduction emissions.

In parallel with carbon sequestration, recent publications have also investigated the effect of co-addition of biochar/digestate in soil on N₂O emissions [147,148]. To date, few studies have examined the effects of biochar on nitrogen transformations within soil amended with digestate [147]. Nitrous oxide (N₂O) is an important GHG, representing around 8% (in 2004) of the global GHG emissions (as CO₂ equivalent), and the global warming potential of N₂O is 298 times higher than CO₂ over a 100-year period [157,158]. One of the major sector

contributing to N₂O emissions is agriculture especially due to nitrification and denitrification processes in soil [158]. Martin et al. [147] investigated the GHG nitrous oxide (N₂O) fluxes from soil amended with a combination of digestate and biochar incorporated at several concentrations (0%, 1%, and 3% w/w soil). For this purpose, three digestates of different origin coming from 1) cattle dung and potato waste, 2) cattle slurry and maize silage, and 3) maize silage, have been tested [147]. Important nitrous oxide emissions were quantified from soil amended with digestate originating from a maize feedstock. In parallel, biochar amendment reduced N₂O emissions from all digestate tested, and the higher effect was observed in the treatments with a maximum load of biochar [147]. Similarly, Bruun et al. [148] investigated N₂O and carbon dioxide (CO₂) emissions over a period of 55 days by combining biochar at two concentrations (1% and 3% by mass) with anaerobically digested slurry [148]. When biochar (3% w/w soil) was applied with digestate, N₂O emissions was reduced by 47% compared to biochar at concentration of 1%. Interestingly, N₂O emissions reduction was concomitant with microbial activity improvement, creating anoxic conditions and immobilization of soil nitrogen [148]. Besides improving the soil physicochemical properties, biochar addition to soil, either with or without digestate, has been shown to be effective in reducing the leaching of organic and inorganic contaminants [129,130,144]. Interesting synergies have been shown when combining digestate and biochar for simultaneous sorption/sequestration and mineralization of pesticides [144,159]. In laboratory incubation experiments, Mukherjee et al. [159] investigated the mineralization of three pesticides (*i.e* bentazone, boscalid, and pyrimethanil) when biochar from pine woodchips was mixed with digestate in a sandy soil [159]. It was demonstrated that digestate increased pesticide mineralization mainly by improving soil microbial activities due to the available of carbon source. Among all the biochar-digestate mixtures investigated, co-application of digestate and biochar (both at 5%) positively influenced mineralization and simultaneous sorption of the three pesticides [159]. Coupling biochar and digestate has demonstrated

promising results in the literature but such an approach is still in its infancy. Furthermore, the performances appear to be multifactorial and several parameters can influence biochar properties and efficiency for agronomic issues. Biochar properties will be mainly influenced by biomass origin and pyrolysis condition, soil origin (sandy, loamy), climatic conditions, and plant types tested.

4.6. Potential risks of using biochar in agriculture

Beside the environmental and agronomic benefits of using biochar, some studies noticed the potential environmental risks related to biochar applications [160–164]. This knowledge is of importance for biochar utilization in an effective and safe manner, as well as for predicting its environmental behavior [163]. Some risks associated with the addition of biochar such as release of toxic substances, retention of heavy metals, suppression of the effectiveness of applied pesticides due to retention, and ecotoxicological effects on soil microbes have been identified [160–164]. Biochar can be the source of various hazardous compounds such as heavy metals (Cd, Cu, Cr, Ni, Zn), polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzodioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and other toxins such as volatile organic compounds, xylenols, cresols, acrolein and formaldehyde [163,165,166]. In particular, PAHs are very harmful to many plants and other living ecosystems [167,168]. These toxic compounds are produced during the pyrolysis process and their concentrations and compositions are highly dependent on biomass feedstock and pyrolysis operation conditions (i.e. temperature, residence time, ramp time) [163,169]. In addition to organic compounds, biochar may also contain metal contaminants (e.g., Cu, Cd, and Pb), which are mainly derived from feedstocks containing heavy metals, such as sewage sludge [170] and residues from some biogas plants [169]. Stefuniuk et al. [169] have evaluated the chemical and the ecotoxicological of biochars produced at three temperatures of pyrolysis (400, 600 and 800 °C) using biogas residues (taken in three different agricultural biogas plants operating in

mesophilic or thermophilic). The analyses highlighted that with an increased pyrolysis temperature, there was an increase in the contents of PAHs and of certain heavy metals (Cr, Cu, Cd, Pb and Mn).

Due to the specific properties of biochar [171], the sorption of pesticides and herbicides by biochar has been studied with promising results [165]. For instance, Taha et al. [172] studied the adsorption of a mixture of 15 different pesticides (with individual pesticide concentrations of 400µg/l) from water by untreated and treated (using phosphoric acid) biochars and charcoal. The authors reported that treated rice straw biochar reduced the pesticide concentration in water (pH 7) by 99 % in only 2 hours [172]. These results show that biochar can be considered as a double-edged product if the amount of pesticide used to treat plant diseases has not been calculated considering the adsorption efficiency of biochar, making plant disease treatment complicated and poorly managed. Nonetheless, pesticides have been generally used with a higher amount than what plant diseases require for treatment, in this case, the management of bioavailability of herbicides and pesticides using biochar is more likely to be favorable, which helps to improve environmental health and food safety as leading to a more rapid development of weed resistance, reduce crop uptake and leaching of these substances [165,172].

Although biochar has many positive effects for agronomic issues, few works on the literature reported the potential negative effect of biochar on terrestrial ecosystems even though most of the biochar have acceptable content of contaminants (e.g., PAHs and heavy metals), which raised question-related to the risk associated with the application of biochar for soil amendments [173–175]. For instance, Busch et al. [176] reported a reduction in shoot and radical length in maize with the addition of biochar generated at high pyrolysis temperature but not for that produced at low temperature. They concluded that the decline in radical and shoot length was due to higher PAHs content, mainly naphthalene, in the biochar extract at

high temperature pyrolysis [176]. Same effect was also reported by Rogovska et al. [177] that investigated the impact of different biochars extracts on *Zea mays*. Biochars were produced from agricultural residues using several thermochemical processes at various temperatures (slow, fast, and gasification pyrolysis processes). The results showed that all produced biochars from corn hindered shoot length, which was associated with PAHs presence. Naphthalene and Phenanthrene delayed germination by 24 h, but after 5 days, the germination rate of the affected seeds was like the control [177].

Nzanza et al. [178] have reported a microbial inhibition after the use of biochar produced from eucalyptus which conducted to negatively impact the crop yield and leaf nutrient as a reduction of tomato dry weight by 13 % and phosphorus in leave by 26 % [178]. Similarly, Liu et al. [179] have also reported a negative impact of biochar application at rate of 60 t ha⁻¹ (produced from wheat straw), which also lead to a microbial inhibition, conducted to reduce plant beneficial fauna population [179]. Stefaniuk et al. [169] also investigated the potential toxicity of biochars produced from various digestates of agricultural biogas plants on various living organisms (springtails, *Lepidium sativum* plants, *V. fischeri*). In the ecotoxicological tests, the most negative effect on the test organisms was characteristic of biochar produced from non-separated mesophilic digestate. This study shows that the main factors determining the level of toxicity of biochars produced from digestate towards various living organisms (springtails and *Lepidium sativum* plants, *V. fischeri*) are both the feedstock origin and the temperature at which the pyrolysis process is conducted [169]. Finally, the suggested biochar quality guidelines, including the International Biochar Initiative [123], and the European Biochar Certificate [124], focus mainly on the total concentrations of PAHs, PCBs and heavy metals in biochar. Even if, the IBI also required to perform a germination inhibition tests using three plants test species, these analyses cannot fully reflect the potential ecotoxicity of contaminants on overall terrestrial and aquatic ecosystems. Thus, further research should be

conducted in detail on the bioavailability of contaminants in biochar and their risks on the various ecosystems to ensure the reliability of biochar application.

5. Economic, environmental, and societal consideration

To assess the benefit of coupling anaerobic digestion with pyrolysis, it is important to take into consideration the three pillars of the sustainability evaluation: economic, environmental, and societal [180]. To characterize a product or a process in terms of sustainability, a suite of different indicators that cover environmental, economic, and social issues, needs to be adopted. Indicators are quantitative or qualitative factors that provide means to measure a degree of achievement, to reflect changes, or to assess performance or compliance [180]. Multicriteria decision analysis based on the different indicators can be further applied to assess the sustainability of the overall approach [181,182]. First, it is important to consider economical balances when considering the development of innovative processes such as the dual approach coupling anaerobic digestion and pyrolysis process [46,183]. Anaerobic digestion is a well-known technology with wide application in the treatment of high-strength organic wastes. The economic feasibility of this type of installation is usually attained thanks to the availability of fiscal incentives [184]. For this purpose, coupling anaerobic digestion and pyrolysis process can represent an interesting option to improve the economic benefits [184,185]. The techno-economic analysis involves evaluation of a process/technology through process simulation approach often using software such as Aspen Plus and Intelligen Superpro Designer [186]. The economic benefits is generally assess by the use of several indicators such as: capital expenditures (CAPEX), operational expenditure (OPEX), net present value (NPV), payback period (PBP), internal rate of return (IRR), return on investment (ROI) [186,187]. The NPV indicates whether the project is profitable, considering the time value of the cash flows, i.e., revenue streams, capital investments and operational costs. The IRR is the discount rate that produces a zero NPV. The payback period refers to the number of years it

takes to generate enough revenues to pay the investment back, without considering the time value of money [187]. Up to date, very few studies have investigated the economical balances of coupling anaerobic digestion with thermo-chemical processes like pyrolysis [188–190]. Mills et al. [190] have investigated the economical assessment of various energy technologies for the conversion of sewage sludge. In their study several scenarios were compared and among them 1) conventional anaerobic digestion with CHP, 2) Thermal Hydrolysis Process (THP) anaerobic digestion with CHP, 3) THP anaerobic digestion followed by drying, pyrolysis of the digested sludge and use of the both the biogas and the pyrolysis gas in a CHP [190]. Interestingly, the scenario 3 coupling anaerobic digestion with pyrolysis process exhibited higher IRR of 7.64% in comparison of 4.05 % and 5.98 % respectively for scenario 1 and 2. When incentives were considered the IRR of the scenario 3 increase until 17.46 % [190]. Gonzalez et al. [189] have also considered the economical balances of coupling anaerobic digestion of agricultural wastes with subsequent solid digestate pyrolysis [189]. If the energetic interest of coupling the two processes was highlighted, it was clearly demonstrated that the economic aspects were clearly discouraging in absence of important subsidies [189]. If the syngas and bio-oil are generally used as energy carriers, biochar can be used for various applications such as energy, as additive in AD process and agricultural issues. To our knowledge, specific studies on the economic assessment of biochar used as additive in AD process (as enhancer, stabilizer, or biogas treatment) has not been reported. As concerns agricultural sector, biochar can have price varying from 100 to 550 \$ t⁻¹ [135,191]. It is also important to consider that such project can also benefit of government incentives such as Feed-in-Tariffs, Renewable Obligation Certificates, carbon credits that can significantly improve the project viability [187,191,192]. Nonetheless, a lack of data is available among the scientific communities and further research are needed on the coupling of anaerobic digestion and pyrolysis at higher TRL to provide relevant data for assessing economic analysis.

The environmental pillar is generally assessed by means of LCA (Life Cycle Assessment) analysis. The LCA is an established method, both technically and scientifically and is standardized by the International Organization for Standardization ISO 14040 [186,193,194]. Some recent studies have evaluated the environmental benefits or not of coupling anaerobic digestion and pyrolysis on sludge [195], municipal solid wastes [196], food wastes [194] or other industrial wastes [197]. Li and Feng [195] have compared the integrated approach with anaerobic digestion and pyrolysis alone from the points of view of life cycle assessment. It was demonstrated that the integrated approach had better environmental performance and energy efficiency than single pyrolysis because prepositive anaerobic digestion enhanced the conversion of sludge organic matter to energy [195]. Similarly, Wang et al. [196] have investigated the LCA of the integration of anaerobic digestion and pyrolysis for treatment of municipal solid waste. Coupling anaerobic digestion and pyrolysis is more environmentally friendly (-11.53 of total environmental impact /kg OFMSW) than single anaerobic digestion or pyrolysis [196]. Opatokum et al. [194] have also investigated the LCA of energy production from food waste through anaerobic digestion, pyrolysis and integrated energy system [194]. The integrated treatment system indicated similar environmental benefits to anaerobic digestion with the highest benefits in climate change and water depletion in addition to the increased energy generation potential and the production of valuable products (biochar and bio-oil) [194]. In parallel, Opatokum et al. [194] demonstrated that the three treatment options of food waste management are environmentally more favorable than the conventional landfilling of the wastes [194]. Finally, in the development of new technologies, it is important to consider the social acceptance among the various stakeholders (farmers, industrials, policy bodies...). To date and to our knowledge, the social benefits of coupling anaerobic digestion with pyrolysis have not been investigated, although each technology has demonstrated its ability to improve the social sustainability, although impact is highly dependent on site location the location [182,198–200]. Furthermore, anaerobic digestion is a

well-developed industry, while pyrolysis is under development [200]. Key sustainability issues for anaerobic digestion/pyrolysis can involve providing energy services to rural and remote areas, agricultural and agro-industrial development, improve agricultural practices, reduce fertilizers and fossil energy dependency, new job creation, restructuring of energetic and agricultural policy, carbon sequestration and environmental benefits. However, it will be important to also take into account the potential disadvantages that can be generated by the development of this innovative wastes and/or biomasses valorization routes (location near inhabited area, increase of transport, odour problems, risk of real estate depreciation...). For these purposes, it is of prime importance during the project development to consult the local residents' interests but also, more generally, coordination between all of the stakeholders. It will be also important to define or harmonize some standards for the use of digestate and biochar as agriculture purposes including some specific tests to evaluate their potential toxicity on aquatic and terrestrial organisms [201].

6. Perspective: an integrated circular cascading approach

Based on available data, the complementarity between anaerobic digestion and pyrolysis process can be extended in future works beyond the classical framework of anaerobic digestion, as shown in **Fig. 4**.

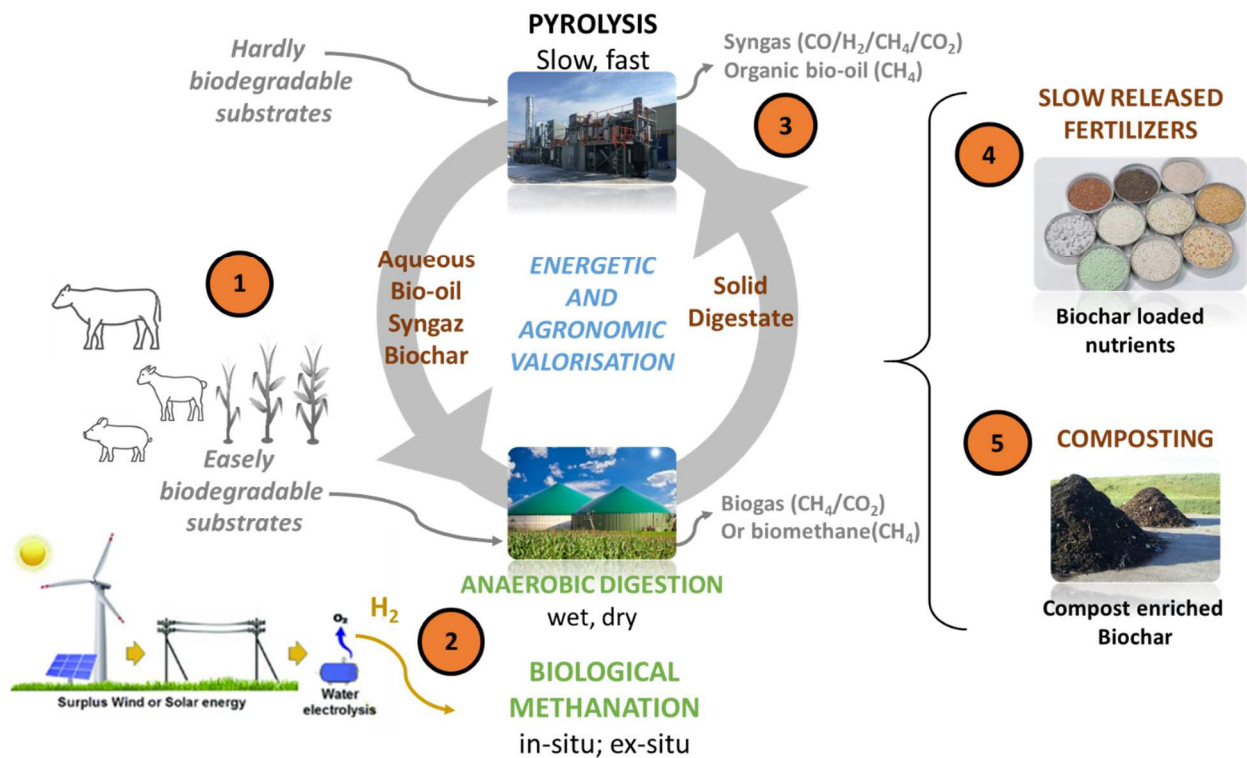


Fig. 4. Mean scheme of perspectives of integrating circular cascading approach by coupling anaerobic digestion and pyrolysis process.

- 1) Biochar can be added before the anaerobic digestion process directly as animal feed additives or in liquid manure storage to reduce gaseous emissions but also in the silage process for the juice recovery. Biochar as animal feed additive has attracted interest recently and has shown positive effects [13,202]. For instance, adding infeed additive biochar resulting in improving animal health, feed efficiency as well as reduce nutrient losses and GHG emissions [202]. Biochar can also be added to uncovered manure storage to reduce odorous emissions of ammonium, ammonia, and other odorous substances, while simultaneously absorbing nutrients in the liquid manure [203,204]. Biochar addition in crops silage dedicated to the anaerobic digestion process can also be interesting because biochar can promote lactic acid fermentation in silage, prevent faulty fermentation, reduce fungus formation and the risk of clostridia infections due to less acetic, and especially butyric, production during silage [203]. Furthermore,

thanks to biochar, there is little or no formation of fermentation juices, reducing environmental burdens due to juice lixiviation to groundwater [203].

- 2) The use of biochar in anaerobic digestion and biological methanation as demonstrated its interest recently to improve the performances [17,72,205]. It seems that biochar addition can stimulate the direct interspecies electron transfer and promote anaerobic digestion and/or in situ biological methanation performance [205]. In parallel, the biological methanation can be done by using the syngas produce during the pyrolysis process [20,72]. Syngas biological methanation is still in its infancy and the implementation of a higher technology readiness level scale is a prerequisite for validation of laboratory-scale results along with the influence of biochar addition that should also be extended. Similarly, research should be reinforced on the APL valorization through the anaerobic digestion process (**Fig. 2**). Indeed, the characteristics of APL, high quantity of COD, numerous complex organic compounds (furans and polyphenols), and ammonia content give them the opportunity to be used as co substrate in anaerobic digestion units [32,206]. Until now, most of the research investigations on the APL valorization through anaerobic digestion has been performed at the batch scale; meanwhile, less is known about continuous pilot-scale trials that deserve to be considered given the influence of APL origin but also its impact on anaerobic digestion stability/performance on microbial communities' diversity. Few information are also available on the APL potential toxicity on anaerobic digestion performances [32] and it seems that biochar addition can attenuate the potential toxicity of some organic compounds on the anaerobic digestion process [29].
- 3) Pyrolysis of solid digestate has attracted the attention of the scientific communities these last decades [50,155]. Several bridges must be studied in the future to better understand the potential of solid digestate in pyrolysis process. First of all, most of the

study has been done until yet with slow pyrolysis process and the impact of fast pyrolysis should be addressed in the future as the products distribution (i.e. biochar, bio-oil, syngas) will not be the same [207]. The typologies (wet vs dry anaerobic digestion process) and solid digestate origin (agricultural, biowastes, sludge...) should be also investigated in the future as the physico-chemical properties of biochar and bio-oil is largely influenced by its pyrolysis conditions and feedstock [208]. Finally, it will be also pertinent to test the pyrolysis of solid digestate in mixture with other co-substrates.

- 4) Another interesting opportunity is to use biochar for the production of slow-release fertilizers and/or organo-mineral fertilizers by absorbing the nutrient content of the liquid digestate creating new markets, and reducing the required land for the liquid digestate [136]. Such an approach can be performed by directly introducing the biochar in digestate before solid/liquid separation using a filter press, screw press, or centrifugal system. This would allow concentrating the available nutrients from the liquid phase in the solid phase and thus producing an “enriched nutrients solid digestate”. Another option is to mix the liquid digestate with biochar for the recovery of “biochar-enriched nutrients”. These two strategies deal with the fact that biochar is able to adsorb available nutrients present in the liquid phase, such as ammonium, potassium, and phosphorous [136,209]. Currently, there are only a few studies on nutrient recovery from the liquid fraction of digestate [112,209]. Kizito et al. [112] have studied the influence of two kinds of biochars (from wood and rice husk) to recover ammonium nitrogen from agricultural digestate through adsorption process. The maximum $\text{NH}_4^+(\text{+})\text{-N}$ adsorption from digestate at an initial concentration of 1400 mg N L^{-1} was 44.64 mg g^{-1} and 39.8 mg g^{-1} for wood and rice husk biochar, respectively. Kizito et al. [209] investigated the performances of phosphate recovery from the liquid fraction of digestate by using four biochars (from wood, corncobs, rice

husks, sawdust pyrolysis). It was demonstrated that biochar can be effectively used to recover P-PO_4^{3-} from digestate and at an initial load of $150 \text{ mg P-PO}_4^{3-} \text{ L}^{-1}$, adsorptions varying from 5.41 to $7.67 \text{ mg P-PO}_4^{3-} \text{ g}^{-1}$ biochar were recorded [209]. Kizito et al. [136] evaluated the addition of digestate-enriched biochar as soil amendment during maize growth. The addition of digestate-enriched biochars resulted in soil organic matter improvement (232%–514%), macronutrients increase (110%–230%). Furthermore, significant increase in biomass yield compared to soil control without biochar enrichment (165% versus 91%, respectively) was reported [136]. Finally, for mineral fertilizer, biochar has been used as a precursor for coating material or in the manufacture of mineral fertilizers with a slow-release properties [210,211]. For instance, Chen et al. [210] investigated an innovative slow-release nitrogen fertilizer based on the coating of urea granules with biochar-based waterborne copolymers. They are tested some biochar produced from different biomasses (maize straw, rice straw, and forest litter) on the fertilizer properties including water resistance, degradability in soil, and leaching behavior of nutrient. Interestingly, the urea particles coated with rice biochar-based copolymer exhibited a better release behavior with 65.28% of nutrient leaching compared to more than 90% from the pure urea [210]. Similarly, biochar (eucalyptus wood, 400°C) granulation with additives (22% bentonite and 5% pregelatinized maize flour) and urea using a granulator dish was investigated by Peregrina et al. [211]. To evaluate the performance of these fertilizers on maize yield, a one-year field experiment with tropical soil was performed. The best results were obtained using biochar-based fertilizers composed of 51% biochar and 10% urea, resulting an increase in nitrogen use efficiency (NUE) and crop yield of 12% and 21%, respectively, compared to pure urea fertilizer [211].

- 5) In addition to producing organo/mineral fertilizers or biochar enriched nutrients, biochar can be incorporated to improve the performances, stability, and agronomic

quality of solid digestate compost if a composting process is applied [149,212,213]. Biochar incorporation inside composting process can improve this latter by several ways: 1) modifying the main physico-chemical properties of compost, 2) enhancing organic matter decomposition through better microbial activities, 3) reducing GHG and ammonia emissions; 4) upgrading compost nutrient quality and maturity through a better stability of the organic matter and 5) improve plant and crops production when compost is used in agronomic issues [214,215].

7. Concluding remarks

While anaerobic digestion and pyrolysis as single processes have been widely studied, their combination in the field of waste management is still in its infancy. Co-application of anaerobic digestion and pyrolysis has proven advantages and benefits in energy recovery, as well as improvement in stability and performance of anaerobic digestion itself along with positive consequences on soil fertility and crop eco-production that could be gained from the combination. This review provided clear evidence showcasing the “energy recovery and efficiency” advantage that could be gained from the combined use of both anaerobic digestion and pyrolysis processes. Notably, through several cascade integrations such as 1) solid digestate valorization during pyrolysis, 2) reuse of bio-oil APL in the anaerobic digestion process, 3) use of the produced syngas in biological methanation, 4) use of biochar in AD process and 5) combination of liquid digestate and biochar for agronomic issues. Biochar has been shown to be a central element in anaerobic digestion and pyrolysis coupling, notably by improving the anaerobic digestion stability/performances and applicability of the “biochar and digestate” combination for agronomic purposes. Nevertheless, the use of biochar in anaerobic digestion or for agronomic issues in combination with digestate is still in its infancy and several avenues are worth exploring. If the energetic benefits have been demonstrated, few data are available on technico-economic analysis of coupling anaerobic digestion and

pyrolysis. To state on the clear benefit of such dual approach it is important to validate the process at higher Technology Readiness Level to have relevant mass and energy flow to be modeled. Future implementation of the dual anaerobic digestion– pyrolysis process does not depend solely on technical constraints. A multidisciplinary approach integrating technical, social, environmental, and economic sciences is worth implementing.

Credit Author Statement

Saida Tayibi: Conceptualization, Investigation, Writing - Original Draft. **Florian Monlau:** Conceptualization, Supervision, Writing, Review & Editing. **Adanae Bargaz:** Writing, Review & Editing. **Raquel Jimenez-prieto:** Writing, Review & Editing. **Abdellatif Barakat:** Supervision, Funding acquisition, Project administration, Conceptualization, Writing, Review & Editing

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Declaration of interest

None.

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Graphical Abstract

