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REVIEW

Biochar from animal manure: A critical assessment on technical feasibility, economic viability, and ecological impact

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Abstract

Animal manure has been used to manage soil fertility since the dawn of agriculture. It provides plant nutrients and improves soil fertility. In the last decades, animal husbandry has been significantly expanded globally. Its economics were optimized via the (international) trade of feed, resulting in a surplus of animal manure in areas with intensive livestock farming. Potentially toxic elements (PTEs), pathogenic microorganisms, antibiotic residues, biocides, and other micropollutants in manure threaten animal, human, and environmental health. Hence, manure application in crop fields is increasingly restricted, especially in hotspot regions with intensive livestock activities. Furthermore, ammonia volatilization and greenhouse gas (GHG) emissions during manure storage, field application, and decomposition contribute to air pollution and climate change. Conventional manure management scenarios such as composting and anaerobic digestion partially improve the system but cannot guarantee to eliminate sanitary and contamination risks and only marginally reducing its climate burden. Hence, this review discusses the potential of pyrolysis, the thermochemical conversion under oxygen-limited conditions as an alternative treatment for animal manure providing energy and biochar. Manure pyrolysis reduces the bioavailability of PTEs, eliminates pathogenic microorganisms and organic micropollutants, and reduces GHG emissions. Pyrolysis also results in the loss of nitrogen, which can be minimized by pretreatment, that is, after removing soluble nitrogen fraction of manure, for example, by digestion and stripping of ammonia–nitrogen or liquid–solid separation. However, conclusions on the effect of manure pyrolysis on crop yield and fertilization efficiencies are hampered by a lack of nutrient mass balances based on livestock unit equivalent comparisons of manure and manure biochar applications. Hence, it is essential to design and conduct experiments in more practically relevant scenarios and depict the observations based on the amount of manure used to produce a certain amount of biochar.

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KEYWORDS

gasification, pyrogenic carbon capture, and storage, liquid manure, negative emissions, organic fertilizer, pyrolysis

1 | INTRODUCTION

Animal manure consists of feces, urine, bedding materials, dropped or indigestible feed, rain and cleaning water and, sometimes, other farm waste in a solid, slurry, or liquid form. The increase in demand for animal products due to human population growth and diet changes led to a substantial increase of animal husbandry in the 20th century (He et al., 2016; Sommer et al., 2013). For example, from 2000 to 2019, livestock units (LSU) per agricultural land area increased from 0.33 to 0.39 LSU ha^{-1} globally (FAOSTAT, 2023). However, in intensive animal farming systems, for example, in many industrialized countries, far higher LSU ha^{-1} are reached. For instance, in 2020, the average livestock density in the EU was 0.7 LSU ha^{-1} of agricultural area and ranged from 0.2 LSU ha^{-1} in Bulgaria, Latvia, and Lithuania to 3.4 LSU ha^{-1} in the Netherlands (Eurostat, 2023). In these cases, farms often import additional animal feed but apply the manure only on fields surrounding the farm. Thus, the generated manure is applied to a smaller area than was necessary to grow the forage, which results in a nutrient surplus in soils.

Manure can be classified into several types based on its total solids, nutrient content, and handling consistency. Liquid animal manure contains less than 1% of solids usually treated in anoxic lagoon systems or in earthen pits and handled with irrigation equipment (Lorimor et al., 2004). Manure slurry contains 4%–10% solids, is treated in deep pits or lagoons, and is applied to the soil using impact head distributors, swivel spreaders, drip hoses, slurry cultivators or trailing shoe distributors. Semisolid manure contains 10%–20%, and solid manure contains more than 20% solid matter (Lorimor et al., 2004). Solid manure can be stacked and piled, which results in fermentation processes. Eventually, it is composted or spread directly on the land. Usually, liquid manure is low in nutrients and contains more soluble nutrients than solid manure. Contrastingly, solid manure contains more insoluble nutrients and carbon than liquid manure (Lorimor et al., 2004). Manure contains all macro- and micronutrients necessary for plant growth. Nitrogen originates mainly from urine as urea, which is quickly transformed into ammonium and ammonia, due to the urease enzyme present in the environment and especially in the feces (He et al., 2016). Hence, chemical properties of manure vary, depending on the animal type, manure management practices, and treatment methods (Figure 1; Tables S1 and S2).

The use of animal manure as an organic soil amendment to improve soil fertility has a long history dating back to ancient civilizations (Araus et al., 2014; Jones, 2013). Due to the high nutrient content, using manure as standalone fertilization or in combination with mineral fertilizers is a common practice among farmers to improve crop yield and land productivity. Due to the variation in their physicochemical composition, animal manures have different impacts on different soil properties. In their meta-analysis, Maillard and Angers (2014) reported an increased level of soil organic carbon (SOC) with the application of manure, and the magnitude of SOC dynamics varied with climate, manure application rate, manure management system, soil properties such as soil texture, and initial SOC content as well as manure type and characteristics. Higher SOC contents result in higher soil porosity and improved aggregation. As a result, infiltration and water-holding capacity may increase whereas bulk density decreases and soil structure improves as shown in various studies (Du et al., 2020; Ferreras et al., 2006; He & Zhang, 2014). On the molecular level, manure decomposition increases the surface carboxyl and phenolic groups in organic matter. Hence, an increase of negative sites in organic matter increases the cation exchange capacity in soil when manure is incorporated (Shi et al., 2019). In the review by Du et al. (2020), an increase in soil enzymatic activity and the abundance of bacteria, fungi, and actinomycetes was noted after repeated manure applications over several years.

In summary, animal manure applications in soil have vast advantages, although it is also associated with several concerns. In intensive livestock farming, high nutrient-containing feed is fed to animals though only 30% of these nutrients are assimilated by the animals (Thangarajan et al., 2013). As a result, a considerable portion of nutrients in animal feed ends up in feed leftovers, partially digested feed, and animal excreta (Melse & de Buisonjé, 2020). Hence, frequent application of manure nutrients may exceed the soil's receptive capacity and lead to runoff and leaching of excess nutrients into nearby surface water bodies and groundwater (e.g., nitrate-N derived from rapid nitrification of manure N) and to accumulation in soil over time (i.e., P buildup in soil; Huygens et al., 2020). In addition, elements such as K, Ca, Mg, Cl, and Na tend to get lost via leaching and runoff due to their high solubility. Higher concentrations of N, P, and K create eutrophication especially in shallow surface water bodies.

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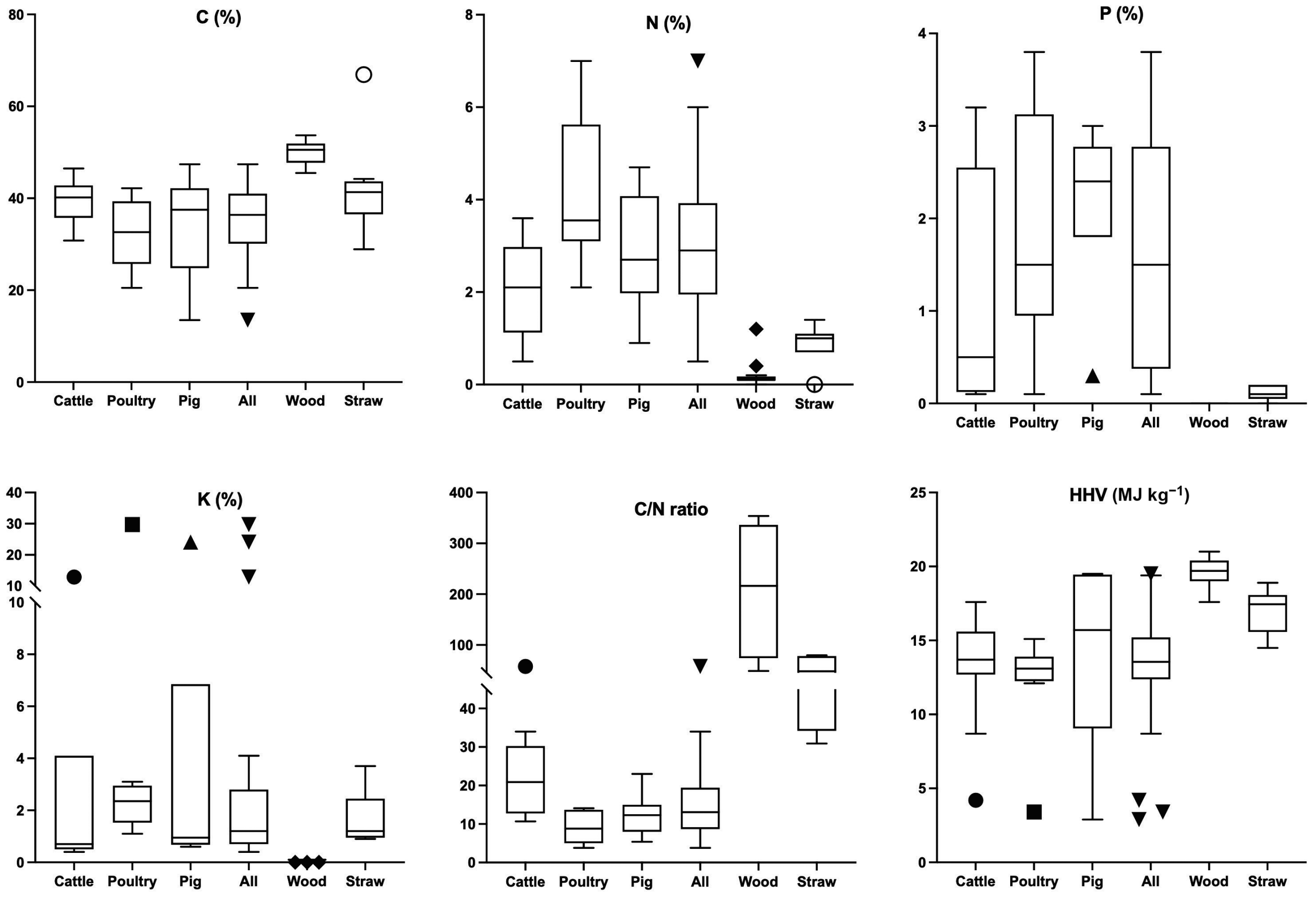

FIGURE 1 Properties of animal manure based on the data presented in Table S2 (C: total carbon, N: total nitrogen, P: total phosphorous, K: total potassium, C/N: C/N molar ratio, HHV: high heating value). Elemental contents are presented in percent dry matter. "All" summarizes the data of cattle, poultry, and pig manure. Data for wood and straw are presented as references with wood and straw being the economically most relevant feedstocks for biochar production, for example, in Europe. Data presented as a boxplot with Tukey whiskery: the box represents the data between the 25th and 75th percentiles, the whiskery are limited to 1.5 times the interquartile distance, further data are presented as individual data points.

For those reasons, the EU Farm to Fork strategy plans to reduce nutrient losses by at least 50% by the year 2030 (EC, 2020). To protect the aquatic and terrestrial ecosystems, the EU needs to reduce its ammonia emissions and N runoff to surface water by at least 38% and 50%, respectively (de Vries et al., 2021). The topic of manure nutrient losses is discussed in more details in the Supplementary Information (Text S1).

Manure storage and treatment (solid–liquid separation, composting, anaerobic digestion) and soil application result in emissions of ammonia, volatile organic compounds, and the potent greenhouse gases (GHGs) N_2O and CH_4 (Shakoor et al., 2021), especially in the summer when higher temperatures promote the release of gases from the liquid phase to the atmosphere (Cárdenas et al., 2021). The presence of ammonium results in nitrification, during which N_2O is emitted as by-product. Elevated concentrations of dissolved ammonium at high pH result in the emission of ammonia due to the chemical equilibrium of both N species. Total N losses during manure slurry storage may be as high as 85% (Kupper et al., 2020). Currently, total GHG-emissions from the global livestock sector equal 7.1 Gt CO_2eq per year, which represents 14.5% of all anthropogenic GHG emissions (Hou et al., 2017; Montes et al., 2013). GHG emissions also occur during manure composting or anaerobic digestion and are discussed in more detail along with processes relevant to GHG emission during and after soil application in the Supplementary Information (Text S2).

Accumulation of potentially toxic elements (PTEs) is another concern associated with animal manure application to soil. Most of these elements originate from feed additives (Meng et al., 2017; Shen et al., 2020). The most abundant PTEs present in animal manure are Cu and Zn (He et al., 2016). However, their bioavailability varies with the manure type, animal type, and feed composition (Sommer et al., 2013). Potential toxic elements from manure soil application could hamper soil microbial

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activity, reduce crop productivity, enter the food chains, and negatively impact animal and human health (Rehman et al., 2018; Zhang et al., 2020).

Pathogens in manure pose a severe risk of spreading zoonotic diseases, for example, Ananna et al. (2021) observed a significantly higher total viable count of fecal microorganisms in manure-amended soils than non-amended soils. Fecal microorganisms in manure can leach into shallow groundwater and pose a threat to human and animal health, for example, by polluting drinking water with fecal bacteria, which may result in diarrhea (Ananna et al., 2021; Sommer et al., 2013). Furthermore, pathogenic microorganisms in animal manure (i.e., *Salmonella*, *Escherichia coli*, *Listeria*, and *Cryptosporidium*) may contaminate fruits and vegetables in manure-applied fields when good agricultural practice is disregarded, and for example, manure is applied late in the growing season. This can cause zoonotic disease outbreaks among animals and humans such as gastrointestinal illnesses, skin irritations, and respiratory illnesses. There are conventional methods to reduce pathogens in manure, which are summarized in the Supplementary Information (Text S3).

Moreover, antibiotics in animal manure pose a significant threat to the environment. Around 30%–90% of antibiotics administered to animals are excreted via the manure pathway (Zahedi et al., 2022). Zhou et al. (2020) identified tetracycline and other antibiotics in manure-derived fertilizers. Plant uptake of such antibiotic compounds could lead to their accumulation in the food chain (Dandeniya et al., 2022; Kumar et al., 2005; Zhou et al., 2020), which may cause fatal diseases due to induced antibiotic resistance in animals and humans (Wang et al., 2019). Concurrently, frequent application of antibiotics in animal husbandry results in the presence of antibiotic-resistant bacteria and antibiotic resistance genes already in manures, as for example, Dandeniya et al. (2022) observed for broiler litter, whose continuous application tends to increase oxytetracycline-resistant epiphytic and endophytic bacteria in carrot. Moreover, the release and accumulation of antibiotic residues through manure application could lead to the development of antibiotic-resistant genes in soil organisms (Hidalgo et al., 2019; Qian et al., 2018). Due to the growing death toll caused by excessive antimicrobial use in the EU, the EU Farm to Fork strategy aims to reduce EU sales of antimicrobials for farmed animals and aquaculture by 50% by 2030 (EC, 2020). The spreading of pathogens and/or microorganisms already carrying antimicrobial resistance genes (ARGs) is another major concern of animal manure application (Checcucci et al., 2020; Congilosi & Aga, 2021; Martín-Marroquín & Hidalgo, 2015).

More than 90% of steroids in the environment come from the livestock sector, and they pose a significant threat to animal and human health triggering endocrine disruptions even at low ppt concentrations (Gudda et al., 2022; Rechsteiner et al., 2020, 2021). Endocrine disruptions and low reproduction development in aquatic species are other detrimental impacts of steroids (Rechsteiner et al., 2020, 2021; Vethaak et al., 2005). Estrone, 17α-estradiol, 17β-estradiol, and estriol are relatively persistent estrogens present in animal manure. Only few microorganisms can completely degrade estrogen in soil by taking them as their carbon source. Exposure to higher levels of estrogen in soils could adversely impact soil microbial activity (Wei et al., 2021).

Converting animal manure into biochar, the solid product of thermochemical conversion under oxygen-limited conditions (pyrolysis), seems a promising approach to address the above-mentioned environmental concerns of manure management. High-temperature conditions (>400°C) could destroy pathogens and organic contaminants (e.g., antibiotics, steroids, etc.) and reduce the availability of PTEs. While it is expected that the environmental harm of manure nutrient losses is reduced, the overall nutrient balance from animal feces to plant growth should be similar compared to traditional manure management. Moreover, labile manure carbon would partly be converted into a long-term carbon sink. Within this context, this critical review of existing literature aims to summarize the knowledge on manure biochar properties and compare impacts of conventional animal manure management and application to soil with the effect of manure pyrolysis, and manure biochar application to soil.

2 | BIOCHAR PRODUCTION FROM ANIMAL MANURE

2.1 | Pyrolysis

Pyrolysis is a thermal conversion of carbonaceous feedstock in the absence of molecular oxygen at temperatures usually well above 400°C resulting in solid, liquid, and gaseous products (Hagemann et al., 2018). The solid product of biomass pyrolysis is called biochar when the H/C$_{org}$ molar ratio and O/C molar ratio are less than 0.7 and 0.4, respectively, which is used in a non-oxidative manner in agriculture or industry to maintain a long-term carbon sink (EBC, 2020). Liquid and gaseous products, that is, bio-oil and noncondensable gases, are usually incinerated to generate energy for drying and heating the biomass and/or other uses like district heating. Production and non-oxidative application of biochar is a negative emission technology called pyrogenic carbon capture and storage (PyCCS), which gained vast attention due to biochar's ability to improve soils and reduce negative side effects of

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agriculture (Schmidt et al., 2019, 2021). The biochar industry is growing exponentially and is currently the most relevant negative emission technology as it is fully commercial (beyond technology readiness level 9) and sequestered about 100,000 t of CO_2 in Europe in 2022 (EBI, 2022). It is based on private (EBC, 2012) and national (e.g., Austria: ÖNORM S2211) standards.

Most historical documents like Allen (1847) or reports on traditional biochar application (Frausin et al., 2014; Liang et al., 2006; Solomon et al., 2016; Woolf, 2008) described the use of wood-based char, not due to its superior properties, but simply as this material was commercially available (charcoal) or the by-product of cooking on open fires. However, with state-of-the-art pyrolysis technology, biochar production for agricultural use became feasible from a multitude of biomass, including primary biomass (forest wood, etc.), nonprocessed by-products (crop residues, etc.) or processed, and marginal biomass (i.e., demolition wood, digested food waste, etc., Buss et al., 2016). Using a variety of biogenic residues from agriculture, industrial activities, and household waste streams as pyrolysis feedstock is growing of interest (Huang et al., 2017; Rathnayake et al., 2021; Rodríguez Alberto et al., 2021). There is an increasing evidence that this more ash-rich biomass results in biochar of at least equal, if not superior, characteristics for use as a soil amendment (Jeffery et al., 2017; Schmidt et al., 2021).

Both phase distribution and characteristics of the products of pyrolysis are controlled by process conditions (heating rate, highest treatment temperature [HTT], residence time, gas flow in the reactor) and feedstock characteristics (chemical composition, moisture, particle size, etc., Hagemann et al., 2018; Rathnayake et al., 2020). Biochar is usually obtained from slow pyrolysis (residence time of minutes to hours) within the temperature range of 350–900°C. Biochar can also be produced by gasification, that is, a thermal treatment with higher, yet sub-stoichiometric oxygen supply optimized for generation of a gas that can be used in an engine to co-produce electricity and heat (EBC, 2012; Hagemann et al., 2018).

Wood feedstocks mainly consist of cellulose, hemicellulose, lignin, and trace amounts of inorganics (Rowell et al., 2005). Manure feedstocks mainly contain cellulose, hemicellulose, lignin, proteins, lipids and fats, and a substantial amount of nutrients. For example, birch wood is composed of 40% of cellulose, 26% of hemicellulose, and 16% of lignin (Mohan et al., 2006), and cattle manure contain 14%–35% of cellulose, 11%–32% of hemicellulose, and 13%–15% of lignin, 8%–30% of proteins, and 15%–32% of lipids and fats (Saady et al., 2021). During the pyrolysis, those compounds progressively break down with increasing pyrolysis temperature. First, hemicellulose and cellulose start to disintegrate in the temperature ranges of 220–315°C and 315–400°C, respectively (Yang et al., 2007). Lignin breakdown occurs in a broad range of temperatures (100–900°C) (Zhao et al., 2017). Higher HTT accelerate the elimination of weaker aliphatic bonds, hydrogen, and oxygen in manure, and create more aromatic organic carbon moieties in the resulting biochar (Ronsse et al., 2013).

2.2 | Feedstock preparation

Pyrolysis of wet manure is an extremely high energy-intensive process (Ro et al., 2010). Hence, there are different strategies to cope with the high moisture content and to make use of the nutrients primarily present in the liquid phase (Figure 2):

1. Solid–liquid separation: The resulting solid phases after separation usually have sufficient dry matter content for pyrolysis at commercial scale directly or after a drying process with lower energy requirement (Meng et al., 2018). For example, Ro et al. (2010) reported that dewatering of wet swine manure to 75% of moisture content before pyrolysis could reduce the energy requirement by 19-folds. The nutrient-rich liquid phase can be combined with the manure biochar to manufacture a biochar-based fertilizer ("charging" of biochar with dissolved nutrients) (Sarkhot et al., 2012; Schmidt et al., 2017). Excess liquid can be applied as a fertilizer separately, used in irrigation or subject to struvite precipitation or similar approaches (Federolf et al., 2016; Katanda et al., 2016). However, direct agronomic use of this liquid portion can impair the benefits associated with pyrolysis such as degradation of manure-derived contaminants (i.e., antibiotics and micropollutants).
2. Drying: In most of the manure–pyrolysis studies available so far, air-drying, solar drying, and oven-drying were used to reduce the moisture content in manure feedstock prior to pyrolysis without prior phase separation (Cantrell et al., 2012; Cao & Harris, 2010; Enders et al., 2012; Gomez-Munoz et al., 2016; Meng et al., 2022). In practice, the excess heat of burning bio-oil and noncondensable gases can be used for this purpose. However, depending on manure water content and heating value, additional energy input might still be needed to sustain drying and pyrolysis (Ro et al., 2010). Drying results in concentrating the nutrients in the solid phase; thus, the biochar may have a higher nutrient content than the biochar produced from solid–liquid separation.
3. Co-pyrolysis/addition of dry plant biomass: Dry matter content of the pyrolysis feedstock can be increased by

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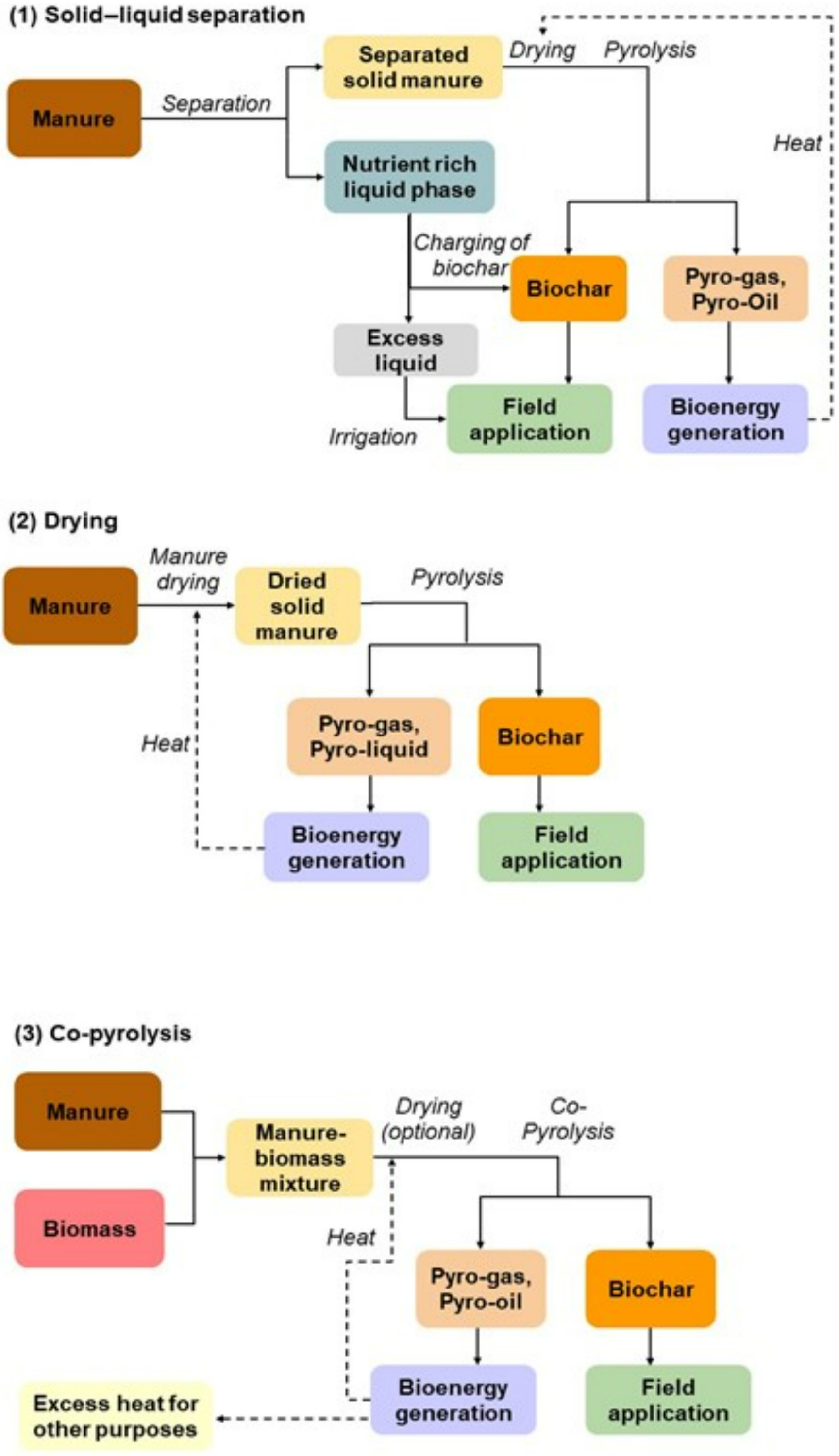

FIGURE 2 Three different approaches to integrate manure in a pyrolysis system (Cantrell et al., 2012; Li et al., 2021; Meng et al., 2022; Ro et al., 2010): (1) Solid–liquid separation—the separated solid phase of manure is pyrolyzed directly while the concentrated nutrient-rich liquid phase can be used to manufacture a biochar-based fertilizer and/or the liquid can be used separately for irrigation purposes. (2) Drying: The excess heat generated during the pyrolysis is used to dry manure prior to pyrolysis without prior phase separation. (3) Co-pyrolysis of manure feedstock with other, more energy-dense biomass. In all approaches, the non-condensable pyrolysis gases and liquids could be used for energy generation.

adding dryer and more energy dense biomass, for example, co-pyrolysis of manure with wood chips or rice straw. This approach may reduce or avoid the need for separate drying, increase overall biochar yield, reduce the content of both nutrients and PTEs, and modify biochar properties, depending on the biomass added (Li et al., 2021; Meng et al., 2018; Qiu et al., 2023). Ro et al. (2010) also reported that biochar could be produced without any external energy by mixing dewatered swine solids with rye grass. Furthermore, this approach facilitates the controlled release of nutrients in manure biochar (Novak et al., 2018), and minimizes the risks associated with PTEs in manure biochar (Li et al., 2020; Qiu et al., 2023).

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In either strategy, the noncondensable pyrolysis gases and oils are burned to generate energy. Alternately, bio-oils could be separated for added value uses (Leijenhorst, 2020). In the present review, we focus on the use of the solid pyrolysis product (biochar).

2.3 | Chemical transformations of manure during pyrolysis and manure biochar properties

2.3.1 | Carbon

The chemical–physical properties of biochar, the solid product of biomass pyrolysis, are largely determined by its carbon compounds. Although carbon is also volatile in pyrolysis, it is released in the form of compounds of low molecular weight, with comparatively high hydrogen and oxygen content. Thus, there is a relative enrichment of carbon with respect to these elements in the biochar (lower molar ratios of H/C and O/C, Hagemann et al., 2018). In the case of biomass with a rather low ash content, like wood, this usually leads to an increase in the carbon content; in the case of manure, the carbon content of the feedstock and biochar is often similar (Figure 3).

During pyrolysis, oxidation is omitted by the absence of molecular oxygen, but still partial oxidation might happen, for example, due to the inherent content of oxygen in biomass and lack of airtightness or dead air entering the reactor within the pore spaces between biomass particles introduced to the reactor (Hagemann et al., 2018). Mass loss during manure pyrolysis can be considerably lower than for plant biomass, which is attributed to the higher content of nonvolatile ash (Figure 3). Overall, manure pyrolysis results in a biochar yield in the range of 40%–60% (d.m.), strongly depending on both the type of manure and pyrolysis conditions, while this values usually lower for wood (20%–30% depending on temperature) (Almutairi et al., 2023; Enders et al., 2012). Interestingly, according to data presented by Cantrell et al. (2012) the recovery rate for total carbon, that is, the ratio of biochar–carbon and biomass–carbon needed to produce the biochar, is similar for wood and manure when pyrolyzed under comparable conditions (60%–40%).

Literature shows considerable variations for both carbon contents and biochar mass yields (Figure 3), which might at least partly be explained by differences in animal husbandry resulting in different content of bedding material in the manure (Figure 1); content of straw or saw dust will affect both biochar properties and mass yield (Figure 3; Tables S3–S5). In addition, manure could have been sampled and dried almost immediately after feces excretion, or after certain storage time, which might be the result of different sampling approaches, stable cleaning intervals and/or manure storage systems, and ultimately caused variations in the carbon content of manure biochar. Storage of manure results in microbial activity and thus changes in feedstock carbon speciation. During pyrolysis, novel carbon compounds are formed by reactions both in the solid phase (primary char) and in the gas phase followed by condensation to form secondary char (Anca-Couce et al., 2017). Their nature depends on both feedstock properties and pyrolysis conditions. Largely depending on the pyrolysis temperature, carbon speciation is dominated by an amorphous arrangement of aromatic polycondensates after pyrolysis at about 300–400°C, which is gradually replaced by graphene stacks and graphitic crystallites with increasing temperature (Keiluweit et al., 2010). Biochars from manure-based feedstock present a lower degree of aromaticity and form less condensed aromatic structures than biochar produced from wood when produced at the same temperature, as evidenced by ^{13}C NMR (McBeath et al., 2014), which may have implications for biochar stability and carbon storage as discussed below. This can partly be explained by differences in feedstock carbon speciation, as, most remarkably, the lignin content of manure is considerably lower (2%–6% for poultry and swine, 12%–13% for cattle; Chen et al., 2003) than in wood (up to 30%). Additionally, higher content of (earth) alkali elements may impact carbon speciation in the biochar, as the deliberate addition of (earth) alkali-rich wood ash reduced the thermal stability of wood biochar (Grafmüller et al., 2022).

2.3.2 | Nitrogen

Manure biochars contain 1%–4.5% nitrogen (Figure 3), which originates from both mineral and organic N compounds in the feedstock. Amino acids can account for up to 8%, 15%–19%, and 7%–17% of the dry matter in cattle, swine, and poultry manure, respectively (Chen et al., 2003). During pyrolysis, they are decomposed quickly and are partially released as ammonia into the gas phase (Weiss et al., 2018).Using state-of-the-art pyrolysis equipment, ammonia and other gaseous N-compounds are degraded via oxidation and reaction with carbon species to innocuous N_2 in the pyrolysis gas combustion chamber. Depending on the pyrolysis gas combustion and exhaust gas filtration technology considerable, low or practically no nitrous oxides emission could be observed. Enders et al. (2012) found that 30%–70% of the manure N remains in the biochar with lower fractions at higher pyrolysis temperatures. Even though the release of N_2 to the atmosphere is not harmful to the environment, the loss of 30%–70% of manure nitrogen during pyrolysis as N_2 is not

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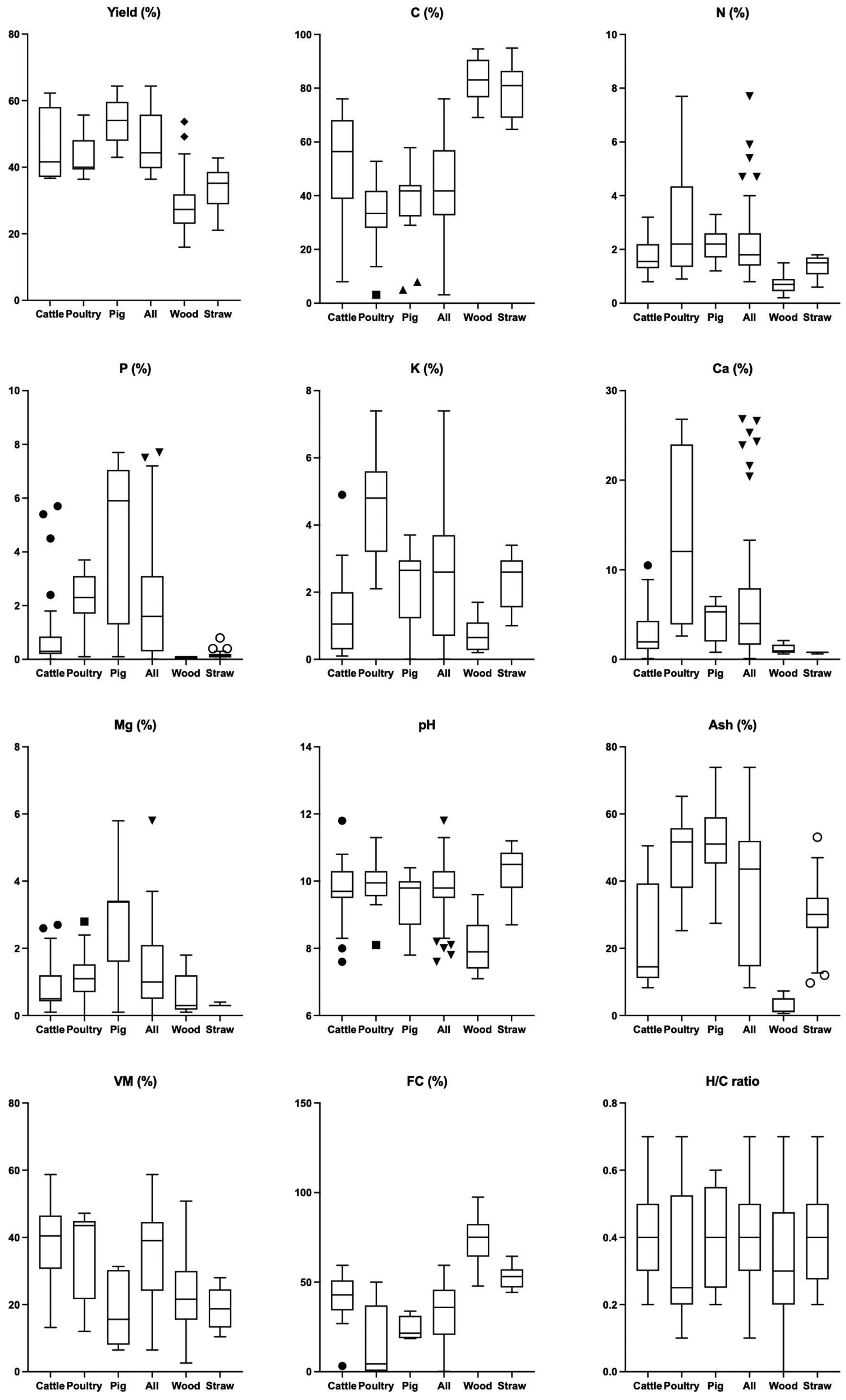

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FIGURE 3 Properties of manure biochar based on the data presented in Table S5 (Yield: biochar yield, C: total carbon, N: total nitrogen, P: total phosphorous, K: total potassium, Ca: total calcium, Mg: total magnesium, VM: volatile matter, FC: fixed carbon, H/C ratio: H/C molar ratio). Elemental contents are presented in percent of dry weight. "All" summarizes the data of cattle, poultry, and pig manure. Data for wood and straw are presented as a reference with wood and straw being the economically most relevant feedstocks for biochar production, for example, in Europe. Data presented as a boxplot with Tukey whiskery: The box represents the data between the 25th and 75th percentiles, the whiskery are limited to 1.5 times the interquartile distance, further data are presented as individual data points.

sustainable from the resource recovery point of view (Hou et al., 2017; Novak et al., 2018).

In the solid phase, inorganic N species of the manure (i.e., ammonium, nitrite, and nitrate) are transformed into organic N species, that is, heterocyclic compounds like pyridinic-N, pyrrolic-N, and graphitic-N or quaternary-N compounds, showing low N availability (Enders et al., 2012; Leng, Xu, et al., 2020; Leng, Yang, et al., 2020; Rombolà et al., 2015). Thus, biochars exhibit lower N availability than fresh manure, manure digestate, and composted manure. Significant reduction of both total N content as well as extractable ammonium-N and nitrate-N content with increasing HTT was reported in Xiao et al. (2018) and Yue et al. (2017) (Table 1). Still, pyrogenic nitrogen was recovered using isotopic labeling in microbial biomass after only a few weeks and in plant biomass 1 year after application (Xu et al., 2022). Piash et al. (2021) observed 49% and 8.9% N release in temperate clay soil amended with chicken manure biochar produced at 300 and 500°C, respectively, after incubating a 2% w/w mixture of biochar and soil for 120 days. Also, Wang et al. (2012) reported lower N recovery when the biochar production temperature increased from 350 to 550°C in cattle manure pyrolysis. The retention of nitrogen during pyrolysis can be increased by the addition of CaO to manure, as has been proposed for the pyrolysis of sewage sludge (Liu et al., 2015).

2.3.3 | Phosphorous

During pyrolysis, P is not volatile, and thus, the total P content increases due to the overall mass loss resulting in a relative enrichment of P by pyrolysis with increasing pyrolysis temperature (Table 2) (Zuo et al., 2020). At the same time, the solubility of P is reduced (Table 2), as some less soluble compounds are formed. Cantrell et al. (2012) found a reduction of water-soluble P content of 68%–99% by pyrolysis with increasing HTT (i.e., from 350 to 700°C) indicating changes in P speciation during pyrolysis. However, the fraction of oxalate-soluble phosphorus was not changed by pyrolysis at up to 500°C and increased after pyrolysis at least 650°C (Uchimiya & Hiradate, 2014).

Speciation of phosphorous can be studied, for example, by K-edge P X-ray adsorption near-edge spectroscopy (P XANES), where X-rays are directly applied on the ground manure biochar. For ^{31}P nuclear magnetic resonance spectroscopy (^{31}P NMR), biochars need to be extracted, and thus, only the NaOH-EDTA-soluble P is characterized (Cade-Menun & Preston, 1996). In manures, P speciation is dominated by orthophosphate. It also contains phytate, and traces of total P content are bound in DNA and lipids (<1% of total P), as assessed by Liang et al. (2018) in composted manure using ^{31}P NMR. They showed that pyrolysis at 400°C or higher eliminates phytate and the obtained biochar contained only mineral phosphate. Within the HTT-range of 400–600°C, 0.4%–4.3% of total P were pyrophosphate ($P_2O_7^{4-}$), which was absent when biochar was produced at 700°C and P was solely present as orthophosphate in the NaOH-EDTA-extract. Those results were confirmed by Sun et al. (2018) for swine feces. Consistent with this, Uchimiya and Hiradate (2014) reported that after the broiler litter pyrolysis at 500°C or higher, the NaOH-EDTA-soluble P is solely present in form of inorganic P species. At 500°C, 3% of total P where present as pyrophosphate, while orthophosphate comprised 97% of total P and was the single P speciation in biochars produced at 650 and 800°C in the NaOH-EDTA-soluble fraction. XANES allows a more in-depth characterization of the orthophosphate. Bruun et al. (2017) showed that pyrolysis of manure digestate increased the contribution of apatite [e.g., hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$)] to total P with increasing temperature (range tested: 350–1050°C) and reduced the content of Fe-bound P and $CaHPO_4$.

At HTT >650°C, P forms gradually become less bioavailable. According to several studies (Bruun et al., 2017; Cantrell et al., 2012; Uchimiya & Hiradate, 2014), this can be due to several mechanisms occurring during the pyrolysis, such as non-extractable P is being occluded in the aromatic structures in biochar, precipitation of amorphous calcium phosphates, formation of nano-size P containing crystals, which can be occluded in the char matrix, and larger sizes of aromatic organic P species of biochar produced at higher HTTs. The formation of water-insoluble hydroxyapatite at HTTs >700°C decreases P availability. For these reasons, Christel et al. (2014) could not quantify any water-extractable P in biochar produced >700°C. Moreover, hydrophobicity of biochar could increase the necessary time until water-soluble P species get dissolved. In the same study, Christel et al. (2014) observed higher P release from raw manure at the beginning of the

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TABLE 1 Total N content and available N forms in different manure feedstock and biochars produced under different pyrolysis conditions.

			NH_4^+-N ($g\,kg^{-1}$)			
	Production temperature (°C)	**Total N ($g\,kg^{-1}$)**	**Deionized water extractable NH_4^+-N**	**2 M KCl extractable NH_4^+-N**	**2 M KCl extractable NO_3^--N ($g\,kg^{-1}$)**	**References**
Chicken manure	n.a. (feedstock)	26	4	7	n.d.	Xiao et al. (2018)
	250	28	0	0	n.d.	
	350	25	0	0	n.d.	
	550	18	n.d.	n.d.	n.d.	
Dairy manure	500	21	n.d.	0	0	Krounbi et al. (2021)
Chicken manure	n.a. (feedstock)	12	n.d.	0	1	Muñoz et al. (2019)
	500	6	n.d.	0	0	
Pig manure	n.a. (feedstock)	18	n.d.	2	0	Muñoz et al. (2019)
	500	18	n.d.	0	0	
Cow manure	n.a. (feedstock)	12	n.d.	0	0	Muñoz et al. (2019)
	500	14	n.d.	0	0	
Poultry litter	550	22	n.d.	0	0	Rahman et al. (2014)
Cow manure	n.a. (feedstock)	16	1	n.d.	0	Yue et al. (2017)
	300	17	0	n.d.	0	
	400	16	0	n.d.	0	
	500	15	0	n.d.	0	
	700	11	0	n.d.	0	

Note: Data presented in this table are presented as reported in the original publications.

Abbreviations: n.a., not applicable; n.d., not determined.

experiment and a decrease over time. Low- to medium-temperature manure biochar (300–600°C) exhibited a higher P-release at the beginning but reached a constant level after 3–6 weeks. Medium- to high-temperature biochar (600–1000°C) presented no P availability at the beginning. Hence, in slow-release fertilizer production, the use of low-temperature biochar (<600°C) was recommended (Christel et al., 2014; Liang et al., 2018; Zuo et al., 2020). When applied to soil, biochar is colonized with P solubilizing microorganisms, which tend to secrete organic acids, which are able to solubilize ortho-P from organo-mineral surfaces (Gul & Whalen, 2016). Buss, Wurzer, et al. (2022) reported that 5% potassium acetate doping prior to swine manure pyrolysis <700°C could enhance the biochar P availability by transforming low soluble P minerals (Al, Ca, Fe, and Mg phosphates) into highly soluble K-phosphates.

2.3.4 | K, Mg, and Ca

The K, Mg, and Ca contents in manure biochar (Figure 3) depend on both their content and speciation in feedstock material as well as pyrolysis conditions (Tables S3 and S4) as with increasing temperature, some of these elements may be volatilized during pyrolysis (Feng et al., 2021). Limitations in mass transfer facilitate higher K, Ca, and Mg retention in the solid phase during pyrolysis (Leijenhorst, 2016). Thus, retention could decrease and reach to a constant level with increasing gas flow rate in the reactor (Lu et al., 2020; Mohanty et al., 2013; Tanner et al., 2016). Their availability (i.e., Mehlich-3 extractable soil Ca, Mg, and K) increases with increasing pyrolysis temperature opposite to major elements (C, N, P), which might be incorporated into the biochar backbone. For instance, the availabilities of Ca, Mg, and K were significantly higher in chicken manure biochar produced at 700°C than at 300°C (Hass et al., 2012). Also, Piash et al. (2021) observed a significant increase of Ca, Mg, and K release in soil incubation when the chicken and dairy manure biochar production temperature increased from 300 to 500°C.

2.3.5 | Formation of pyrogenic contaminants

During pyrolysis, polycyclic aromatic hydrocarbons (PAHs) are formed. However, when adequate process

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TABLE 2 Phosphorous solubility in different manure biochar samples.

Manure type	Production temperature (°C)	Total phosphorous P_{tot} (g kg^{-1})	Soluble P fractions (g kg^{-1})								References
			NaOH-EDTA		Oxalate		Mehlich		Water		
			(g kg^{-1})	% P_{tot}	(g kg^{-1})	% P_{tot}	(g kg^{-1})	% P_{tot}	(g kg^{-1})	% P_{tot}	
Broiler litter	n.a. (feedstock)	22	17	75	19	88	1	4	2	10	Uchimiya and Hiradate (2014)
	350	31	15	49	27	87	1	3	0	0	
	500	39	19	48	35	89	0	1	0	1	
	650	41	17	41	38	92	1	1	0	0	
	800	41	14	33	39	95	0	1	0	0	
Swine manure	n.a. (feedstock)	10	8	78	9	88	1	8	2	14	Liang et al. (2018)
	300	13	8	65	11	89	0	3	1	6	
	400	15	8	56	13	90	0	3	1	4	
	500	18	9	52	16	93	0	2	1	5	
	600	20	10	47	19	94	0	2	0	2	
	700	24	10	40	23	98	0	1	0	1	
Feedlot manure	n.a. (feedstock)	n.d.	7	n.d.	1	n.d.	n.d.	n.d.	3	n.d.	Uchimiya et al. (2015)
	350	n.d.	12	n.d.	1	n.d.	n.d.	n.d.	1	n.d.	
	700	n.d.	5	n.d.	0	n.d.	n.d.	n.d.	0	n.d.	
Swine manure	n.a. (feedstock)	n.d.	20	n.d.	3	n.d.	n.d.	n.d.	11	n.d.	Uchimiya et al. (2015)
	350	n.d.	24	n.d.	5	n.d.	n.d.	n.d.	0	n.d.	
	700	n.a.	23	n.d.	9	n.d.	n.d.	n.d.	0	n.d.	
Swine manure	n.a. (feedstock)	37	20	52	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	Sun et al. (2018)
	300	55	22	40	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	
	450	71	24	34	n.d.	n.d.	n.d	n.d.	n.d.	n.d.	
	600	79	24	31	n.d.	n.d.	n.d	n.d.	n.d	n.d.	
Dairy manure	n.a. (feedstock)	6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2	n.d.	Cantrell et al. (2012)
	350	10	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	
	700	17	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	
Paved feedlot manure	n.a. (feedstock)	7	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3	n.d.	Cantrell et al. (2012)
	350	11	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1	n.d.	
	700	18	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	

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TABLE 2 (Continued)

Manure type	Production temperature (°C)	Total phosphorous P_{tot} ($g\,kg^{-1}$)	Soluble P fractions ($g\,kg^{-1}$)								References
			NaOH-EDTA		Oxalate		Mehlich		Water		
			($g\,kg^{-1}$)	% P_{tot}	($g\,kg^{-1}$)	% P_{tot}	($g\,kg^{-1}$)	% P_{tot}	($g\,kg^{-1}$)	% P_{tot}	
Poultry litter	n.a. (feedstock)	14	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3	n.d.	Cantrell et al. (2012)
	350	21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	
	700	31	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1	n.d.	
Swine solids	n.a. (feedstock)	25	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	11	n.d.	Cantrell et al. (2012)
	350	39	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	
	700	59	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	
Turkey litter	n.a. (feedstock)	16	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	9	n.d.	Cantrell et al. (2012)
	350	26	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1	n.d.	
	700	37	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2	n.d.	

Note: Data presented in this table are presented as reported in the original publications.

Abbreviations: n.a., not applicable; n.d., not determined.

conditions are maintained, they do not condense onto the biochar but are burned as part of the pyrolysis gas (Bucheli et al., 2015; Buss, Hilber, et al., 2022). In practice, product certification, for example, the European Biochar Certificate (EBC, 2012), controls the PAH content of commercial biochar using adapted analytical protocols (Hilber et al., 2012). Since manures contain significant amounts of nitrogen, heterocyclic PAHs, that is, such with at least one nitrogen atom ("heteroatom") in the molecule, are also likely to be formed. We are only aware of one study that investigated these substances in pyrogenic material and quantified 0.3 $mg\,kg^{-1}$ N-PAH in pyrolyzed digestate (4% nitrogen content in the feedstock), with a low PAH content of 5 $mg\,kg^{-1}$ (Weidemann et al., 2018). Since the heterocyclic PAHs are also formed in the gas phase, the content of heterocyclic PAHs on the biochar is also mainly determined by the avoidance of condensation on the solid, that is, by process parameters rather than by the selection of the feedstock. Relevant heterocyclic PAHs have boiling points in the range of 174–355°C, which is in the range of the 16 EPA PAHs already controlled in biochar quality assurance. Thus, an additional check by measurement in the context of quality control does not seem necessary here. Manures may contain relevant levels of chlorine, which can act as precursors for chlorinated organic pollutants. These substances are also monitored as part of the EBC (EBC, 2012). In the available data on the quality control of biochar in Europe of the past 10 years, to date there have rarely been cases where PCDD/Fs and PCBs (polychlorinated dibenzo-*p*-dioxins and dibenzofurans, polychlorinated biphenyls) were at all quantifiable (below limit of quantification) and there has never been an exceedance of the respective limit values in practice (confidential database of Ithaka Institute, 936 analysis). The conditions under which PCDD/F and/or PCB contamination of pyrogenic materials, that is, both formation and sorption to biochar, can occur are the subject of ongoing research. The quality assurance methods already established for biochar (EBC, 2012) also provide sufficient assurance for manure-based biochar.

2.4 | Impact of pyrolysis on bioavailability of manure-derived PTE

Biochar's PTE content is increased compared to its feedstock due to mass loss during pyrolysis and the high boiling points of PTEs (Table 3). Elements including Hg, Pb, Cd can be volatilized at high temperatures (Li et al., 2021), whereas 75%–90% of thermally stable PTEs such as Ni, Cr, and Mn were concentrated in the solid phase (Meng et al., 2017) As a result, a decrease in total elemental Pb/Ni ratio was observed in several studies

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TABLE 3 Comparison between total PTEs concentration in different manure biochar samples and permitted PTE threshold levels.

Manure type	Biochar production temperature (°C)	Pb	Cd	Cu	Ni	Hg	Zn	Cr	As	Reference
		mg kg^{-1}								
Dairy manure	n.a. (feedstock)	1	0	55	6	n.d.	220	n.d.	0	Cantrell et al. (2012)
	350	1	0	99	16	n.d.	361	6	1	
	700	1	<bd	163	25	n.d.	423	10	1	
Feedlot manure	n.a. (feedstock)	2	0	37	3	n.d.	223	n.d.	1	Cantrell et al. (2012)
	350	1	0	92	4	n.d.	359	4	1	
	700	0	0	136	7	n.d.	448	7	2	
Poultry manure	n.a. (feedstock)	1	0	146	4	n.d.	483	n.d.	20	Cantrell et al. (2012)
	350	1	0	213	8	n.d.	712	5	25	
	700	1	0	310	11	n.d.	1010	6	30	
Swine solids	n.a. (feedstock)	1	0	918	10	n.d.	1961	n.d.	1	Cantrell et al. (2012)
	350	3	1	1538	16	n.d.	3181	25	1	
	700	<bd	0	2446	26	n.d.	4981	37	2	
Turkey manure	n.a. (feedstock)	1	1	349	17	n.d.	424	n.d.	102	Cantrell et al. (2012)
	350	2	1	535	29	n.d.	690	8	138	
	700	<bd	1	762	40	n.d.	909	10	166	
Swine manure	n.a. (feedstock)	9	0	220	10	n.d.	764	n.d.	3	Meng et al. (2022)
	300	9	1	287	13	n.d.	927	n.d.	3	
	400	16	1	362	19	n.d.	1078	n.d.	3	
	500	14	0	371	22	n.d.	1279	n.d.	3	
	600	13	0	331	20	n.d.	1070	n.d.	3	
Swine manure	300	21	1	347	53	n.d.	1748	n.d.	3	Meng et al. (2022)
	400	23	1	386	31	n.d.	1619	n.d.	3	
	500	22	1	489	23	n.d.	1629	n.d.	4	
	600	21	1	615	24	n.d.	1611	n.d.	5	
Swine manure	n.a. (feedstock)	13	0	15	6	n.d.	69	17	n.d.	Liang et al. (2018)
	300	17	0	25	9	n.d.	108	22	n.d.	
	400	21	0	33	13	n.d.	115	25	n.d.	
	500	33	0	35	18	n.d.	126	29	n.d.	
	600	39	0	42	23	n.d.	131	30	n.d.	
	700	48	0	56	29	n.d.	141	37	n.d.	

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TABLE 3 (Continued)

Manure type	Biochar production temperature (°C)	Pb	Cd	Cu	Ni	Hg	Zn	Cr	As	Reference
		$mg\,kg^{-1}$								
Chicken manure	n.a. (feedstock)	4	1	69	12	n.d.	412	39	n.d.	Bai et al. (2020)
	300	5	1	95	19	n.d.	767	60	n.d.	
	400	6	2	113	20	n.d.	806	63	n.d.	
	500	6	2	130	22	n.d.	830	72	n.d.	
	600	6	1	143	27	n.d.	848	87	n.d.	
	700	5	1	161	31	n.d.	942	92	n.d.	
Chicken manure	n.a. (feedstock)	n.d.	1	25	4	n.d.	61	8	1	Wang, Zou, et al. (2021)
	350	n.d.	1	41	7	n.d.	104	15	2	
	500	n.d.	1	45	7	n.d.	131	13	2	
	650	n.d.	1	51	8	n.d.	131	16	2	
	800	n.d.	1	55	9	n.d.	160	18	2	
IBI		121–300	1.4–39	143–6000	47–420	1–17	416–7400	93–1200	13–100	IBI (2015)
EBC-Feed		10	0.8	70	25	0.1	200	70	2	EBC (2012)
EBC-AgroOrganic		45	0.7	70	25	0.4	200	70	13	EBC (2012)
EBC-Agro		120	1.5	100	50	1	400	90	13	EBC (2012)
EBC-Urban		120	1.5	100	50	1	400	90	13	EBC (2012)
EBC-Consumer materials		120	1.5	100	50	1	400	90	13	EBC (2012)
EBC-Basic materials		No limit								EBC (2012)

Note: Data presented in this table are presented as reported in the original publications.

Abbreviations: <bd, below detection limit; EBC, European Biochar Certificate; IBI, International Biochar Initiative; n.a., not applicable; n.d., not determined; PTE, potentially toxic element.

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(Bai et al., 2020; Cantrell et al., 2012; Liang et al., 2018). Apart from the production temperature, the PTE composition in feedstock and the type of the pyrolysis unit (e.g., lab vs. pilot scale) also had an impact on the volatilization and retention probabilities of PTEs. For example, Cantrell et al. (2012) reported an increased Pb/Ni elemental ratio with HTT for swine solids and turkey litter, where in the same study, they reported a decreasing trend for dairy manure and feedlot manure. Furthermore, Meng et al. (2022) exhibited an increasing trend of Pb/Ni elemental ratio when the swine manure pyrolyzed in pilot scale pyrolyzer and decreasing trend of Pb/Ni ratio when the swine manure pyrolyzed in lab-scale pyrolysis unit.

In general, PTEs in biochar are less bioavailable and bioavailability is reduced with increasing HTT as evidenced by extractions, soil incubations, and quantification of PTE in plants grown in the presence of manure biochar (Bai et al., 2020; Meng et al., 2022; Uchimiya et al., 2012; Zuo et al., 2020). Hence, the bioavailability of PTEs was shown to be significantly lower in manure biochar-amended soils than manure or manure compost-amended soils (Meng et al., 2022; Sarfaraz et al., 2020; Sun et al., 2018). Availability of PTEs is reduced as they are transformed from exchangeable, carbonate, and iron-bound fractions into more stable fractions such as condensed carbonaceous matter bound fractions (Table S6). For instance, Zuo et al. (2020) reported a reduction of bioavailability of Cu and Zn in chicken and dairy manure and digestate-derived biochar compared to raw manure and digestate, which became more pronounced with increasing pyrolysis temperature. Furthermore, Bai et al. (2020) and Shen et al. (2020) examined the effect of different pyrolysis temperatures (i.e., from 300 to 700°C) on chicken manure-derived biochar and swine manure-derived biochar, respectively, and reported the transformation of exchangeable fraction, acid fraction, and reducible fractions of Cr, Mn, Cu, and Zn in chicken and pig manure into oxidizable and residual fractions when the HTT >500°C. Leachability and bioavailability were reduced with increased HTT. Furthermore, co-pyrolysis of manure with other noncontaminated biomass was suggested to produce biochar with lower PTE levels (Li et al., 2020; Qiu et al., 2023).

2.5 | Elimination of pathogens, antibiotic resistance genes, antibiotics, steroids, and other micropollutants by pyrolysis

Deoxyribonucleic acid (DNA) as a compound is destroyed during pyrolysis starting at 160–200°C and is fully degraded at around 400°C (Alongi et al., 2015), whereas DNA plasmids with actual genetic information were fully destroyed already at 190°C (Karni et al., 2013). Gene destruction by pyrolysis was shown for ARGs in penicillin fermentation residue containing 56 β-lactam resistance genes (Wang, Zhang, et al., 2021). For municipal sewage sludge, pyrolysis at 500°C for 5 min reduced all genes to below detection limits, including 16S rRNA genes, various ARGs, and integrase gene of class 1 integrons (Kimbell et al., 2018). Soils that received biochar obtained from composted pig manure showed no difference in their content of ARGs and mobile genetic elements compared to non-amended control soil, but significantly lower contents of ARGs and mobile genetic elements than a soil amended with non-pyrolyzed, composted pig manure (Zhou et al., 2019). Also, Kimbell et al. (2018) reported a significant reduction of ARGs below the detection limit when pyrolyzed at >500°C during batch pyrolysis of municipal solid waste. Thus, manure pyrolysis considerably reduces biological hazards in soil compared to composting manure.

Estrogenic compounds present in manure have boiling points ranging from 295 to 432°C. Hence, pyrolysis at 400°C for 1 h reduced estrogens in manure by 95% and an almost complete elimination was achieved by 500°C for 1 h (Hoffman et al., 2016). Furthermore, the elimination of pharmaceutical residues (i.e., Ciprofloxacin, Levofloxacin, Clarithromycin, Carbamazepin, 17-α-Ethynylestradiol, Diclofenac, Cefuroxim, Sulfamethoxazol, 17-β-Estradiol, Metoprolol, and Bezafibrate) in sewage sludge pyrolysis was revealed at 500°C in a commercial pyrolysis unit (Stenzel et al., 2019), and it is conceivable that this will also take place when other types of feedstock is pyrolyzed. In essence, manure biochar produced at a minimum 500°C is a safe product with regard to pathogens and organic (trace) contaminants when the residence time is sufficiently long to guarantee complete pyrolysis (Table S6). Since manure biochar performances and avoidance of manure-related environmental threats are highly dependent on biochar properties, adhering to biochar-related regional and international guidelines (Text S4) makes biochar production and application in the environment safer and secure with minimum impact on the human, animal, plant, soil, and environmental health.

3 | IMPACT OF MANURE PYROLYSIS ON GASEOUS EMISSIONS OF MANURE MANAGEMENT SYSTEMS

Storage, conversion, and spreading of manure can result in gaseous emissions including $N_2O$and ammonia due to the high concentration of bioavailable C and N

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and elevated pH. Manure-derived biochar neither provides relevant amounts of available C nor N (Table S7). As detailed above, manure biochar is highly recalcitrant compared to manure feedstock and will not cause N_2O or ammonia emissions during storage.

Methane emissions in manure management occur predominantly in the barn and during storage as the result of microbial activity (Hou et al., 2017). According to Hou et al. (2017), total CH_4 emissions average 3.2% of total manure carbon in the 27 EU countries. If the manure is continuously separated (and/or dried) and the resulting solids are pyrolyzed, there is little time for microbial activity to create biogeochemical conditions favorable for methanogenesis (especially in a highly reduced environment, availability of carboxylic acids or H_2/CO_2 as a substrate for methanogenesis, cf. Lyu et al., 2018). Since the separated liquid contains rather low concentrations of available carbon and is also either filtered or processed into biochar-based fertilizers, very little CH_4 emissions are expected from the separated fraction either. Consequently, CH_4 emissions might be eliminated by separation and pyrolysis (Scenario 1, Figure 2).

Nitrous oxide emissions mainly occur after soil application of manure and only to a small extent during slurry storage as a by-product of microbial nitrification and obligate intermediate of microbial denitrification (Hagemann et al., 2016). According to Hou et al. (2017), nitrous oxide emissions from manure management average 1.5% of total nitrogen in manure across the 27 EU countries. As detailed above, manure biochar does not contain highly available nitrogen, but covalently bound nitrogen, which will hardly contribute to nitrous oxide formation. However, in the case of separation prior to pyrolysis (Scenario 1, Figure 2), most of the nitrogen will be present in the separated liquid fraction. Applying the liquid fraction to the field reduces N_2O emissions by 45% compared to regular manure application (Hou et al., 2015). If the liquid phase is combined with the biochar to form a biochar-based fertilizer (Scenario 1, Figure 2), the nitrogen is retained in the pore structure to improve nutrient retention. According to Hagemann et al. (2017) and the meta-analysis of Borchard et al. (2019), combining nitrogen fertilization with biochar reduces fertilizer-induced formation of N_2O by 38%. However, due to a lack of experimental results specifically on the interaction of biochar with the liquid fraction of manure, it is difficult to quantify the actual extent of nitrous oxide emission reduction, particularly in the long term.

Ammonia emission occurs in the barn as well as during manure storage and application and makes up to 36% of total excreted nitrogen in the EU-27 (Hou et al., 2017). Animals excrete nitrogen predominantly in the form of urea, which is quickly converted to ammonium nitrogen by ureases present in the feces, especially when the urine and feces are mixed. The high pH then provides an equilibrium that favors the chemical conversion of ammonium to ammonia. Thus, manure pyrolysis does not mitigate ammonia emissions at the barn level (13% of excreted N in the EU-27, Hou et al., 2017). Also, energy-intensive thermal drying process prior to pyrolysis (Scenario 2, Figure 2) will volatilize 21%–36% of N in manure as ammonia; however, this technical process allows ammonia scrubbing with efficiencies of up to 71%–99% (Hou et al., 2017). During pyrolysis, ammonia is predominantly oxidized to N_2 in the afterburner (Sørmo et al., 2020). Even though the N_2 emission is environmentally not harmful, in nutrient and resource recovery perspectives, loss of N from the system without making that N plant available or storing it in the soil over time is not desirable. Hence, the separation of soluble nitrogen fraction of manure (by digestion, solid–liquid separation, or stripping of ammonium nitrogen) before pyrolysis is necessary to minimize the loss of nitrogen during pyrolysis.

Commercial pyrolysis units show low levels of non-CO_2 GHG emissions. In summary, a pyrolysis-based manure management approach exhibits substantially lower GHG emissions per ton of manure dry matter than all other conventional manure management systems (Table 4). The overall effect will strongly depend on the actual implementation (drying vs. co-pyrolysis vs. separation, efficiency of drying or separation, handling of separated liquids, duration of manure storage prior to pyrolysis) (Figure 2). Moreover, these data must be interpreted with care: Esteves et al. (2019) reviewed numerous life cycle assessments (LCA) of biogas production from manure. This also includes manure storage and they found considerable variation in the definition of the scope of LCA, the definition of the functional unit, and various other aspects that make the direct comparison between studies difficult.

4 | IMPACT OF MANURE BIOCHAR ON SOIL PROPERTIES AND PLANT GROWTH

The agronomic impacts of biochar application to soil have been subject to numerous studies and meta-analyses. In their systematic review of 26 rigorously selected meta-analyses published in 2016–2020 on biochar effects on soil properties and agronomic performance parameters, Schmidt et al. (2021) found evidence for overall positive, that is, desirable effects of biochar on the selected agronomic parameters. These, among other effects, included increases in crop yield and plant productivity, water use efficiency, root biomass, soil microbial activity, available phosphorous in soil, and SOC as well as decreases in

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TABLE 4 Literature data on GHG emissions for different manure management systems.

Scenario	Total GHG emissions per ton of manure dry matter ($t\,CO_2eq\,t^{-1}$)	Reference
Poultry litter pyrolysis	0.07	Industrial reference (anonymized), Ithaka database
Cattle manure pyrolysis	0.17	Struhs et al. (2020)
Dairy manure management system with belowground concrete storage system	0.17	Aguirre-Villegas and Larson (2017)
Beef cattle manure composting	0.37	Pattey et al. (2005)
Beef cattle manure storage as slurry	0.39	Pattey et al. (2005)
Beef cattle manure stockpiled	0.43	Pattey et al. (2005)
Dairy cattle manure storage as slurry	0.61	Pattey et al. (2005)
Dairy cattle manure composting	0.61	Pattey et al. (2005)
Dairy cattle manure stockpiled	0.74	Pattey et al. (2005)
Biogas digester with biogas capture for energy purposes, swine manure	0.98	Cherubini et al. (2015)
Flare biodigester with biogas flaring, swine manure	2.15	Cherubini et al. (2015)
Composting, swine manure from piglet production	2.22	Cherubini et al. (2015)
Open slurry tanks without natural crust cover, swine manure	2.79	Cherubini et al. (2015)

Note: Original extracted data and necessary calculations are available from https://zenodo.org/record/8032208.

Abbreviation: GHG, greenhouse gas.

nitrous oxide emissions, nitrate leaching, and trace metal concentrations in plants. Most of the listed effects can be explained by the typical porous structure and aromatic carbon backbone of biochar, which enables the retention of water, nutrients, and contaminants as well as interactions with microbes, for example, by acting as electron shuttle and/or alternate intermediate electron donor and acceptor (Chen et al., 2014; Kappler et al., 2014; Schmidt et al., 2021). The dataset considered in Schmidt et al. (2021) comprised biochars from a wide range of feedstock materials, including some manure biochars, and the general results regarding the 26 agronomic parameters can in principle also be applied to the effect of manure biochar.

Comparing manure and manure biochar application at the same application rate (15 Mg ha^{-1}), Sukartono (2011) reported that converting cow manure into manure biochar enhanced organic carbon content in the soil. Joardar et al. (2020) reported enhanced plant growth in poultry litter biochar compared to non-pyrolyzed poultry litter-amended treatments (both amendments at 3% [w/w] application dose). Zolfi-Bavariani et al. (2016) showed a significant increase in organic carbon content, electrical conductivity, and cation exchange capacity in poultry manure biochar-amended treatments compared to poultry manure-added treatments (2% w/w). Kiran et al. (2017) reported reduced bioavailability of Cd in Cd-contaminated soil when amended with cow manure biochar compared to cow manure (all amendments at 3% and 6% [w/w] application dose). Gascó et al. (2016) reported significantly lower C mineralization in pig manure biochar-added treatments than the pig manure containing treatments (all amendments at 8% [w/w] application dose) as the biochar does not contain labile carbon that could be readily mineralized.

Aside from the above comparisons between manure and biochar application in soil, several studies have compared soil enriched with manure biochar to non-amended soils and observed increased nutrient availability (Speratti et al., 2018; Uzoma et al., 2011; Yin et al., 2022). Also, higher SOC content (Abbasi & Anwar, 2015; Subedi et al., 2016; Zhang et al., 2018), improved soil physical properties such as saturated hydraulic conductivity (Gavili et al., 2018), moisture retention (Rahman et al., 2014), water use efficiency (Uzoma et al., 2011), and enhanced soil enzymatic activities (Subedi et al., 2016) were observed in manure biochar-amended soils compared to non-amended soils. As a result of improved soil conditions, many manure biochar-amended soils showed enhanced plant growth compared to non-amended soils. Krounbi et al. (2021) reported increases in tomato, marigold, and radish plant biomass when the soil was mixed with N-enriched dairy manure biochar compared to fertilized control. Chan et al. (2008) reported an increase in radish dry matter yield after poultry litter biochar amendment, Slavich et al. (2013) reported improved productivity in a subtropical pasture due to enhanced N use efficiency after applying

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feedlot manure biochar, and Uzoma et al. (2011) observed a significant increase in maize grain yield in cow manure biochar-amended soils due to enhanced plant water and nutrient use efficiency. In addition to yield increases, Chan et al. (2008) and Uzoma et al. (2011) reported significant increases in Ca, Mg, and K content in plant biomass. Overall, the impact of manure biochar on soil properties and plant growth is heavily depended on manure biochar physicochemical properties, biochar application dose, and soil environmental conditions (Table S8). Apart from above-mentioned positive impacts of manure biochar application over manure application, several studies observed undesired impacts after the application of manure biochar to soil. For instance, Gascó et al. (2016) observed the highest soil enzymatic activity in manure-added treatments, and a negative impact of manure biochar on this parameter. This may be due to the fact that biochar is less labile compared to the manure, that is, biochar does not provide substrate for the enzymatic activity. Also, within the large review dataset in Schmidt et al. (2021), no systematic yield gain from biochar application has been demonstrated for temperate climates with an average annual temperature of less than 10°C. Moreover, Are et al. (2017) reported better performance of composted and non-composted poultry manure than biochar from poultry manure for improving soil physical properties (i.e., saturated hydraulic conductivity and water stable aggregates) under the same application dose (5 t ha^{-1}). When comparing manure biochar, wood biochar, and manure-wood blended biochar, Novak et al. (2018) reported undesirably higher release of P and K in manure biochar-added treatments than the wood-manure blended biochar and wood biochar-added treatments under the similar application dose (20 g kg^{-1}). Hence, they recommend producing biochar by co-pyrolysis of wood and manure to ensure controlled release of nutrients and to not exceeding the soil and plant nutrient requirements.

In summary, most studies mentioned above (as well as in Table S9) compared manure biochar amendment either to a manure amendment at the same application rate or to a non-amended or just mineral fertilizer-added soil. Based on these data, it is currently difficult to make a recommendation pro or con regarding manure pyrolysis from a plant nutrient perspective. It is essential to design and conduct experiments in more practically relevant scenarios and depict the observations based on the amount of manure used to produce a certain amount of biochar. In other words, different manure management practices should also be compared based on the same amount of initial manure. With that, for example, 10 t manure per hectare would be compared, for example, to 4 t manure biochar obtained from pyrolysis of 10 t of manure and/or to 6 t compost obtained from composting 10 t of manure. This or similar approaches would help to get the most accurate picture when comparing manure versus manure biochar applications to support the decision-making of, for example, a farmer who needs to optimize the on-farm nutrient recycling with a given amount of manure available. Such information will facilitate an accurate estimation of net ecosystem and economic benefits (often referred to as NEEB) between manure and manure biochar-based farming systems as, for example, performed to quantitatively compare mineral fertilization with biochar-based organic fertilization (Bi et al., 2022; Li et al., 2015). In addition, further mechanistic information can be obtained, for example, from experiments with normalized carbon or nutrient contents of the organic amendments. Also, when comparing the effects of manure biochar with the application of non-pyrolyzed manure, the manure quality and its preparation (e.g., composting, lactic fermentation, oxygenation, etc.) must be considered. In addition to its physicochemical impacts on soil, the application of manure biochar instead of manure may reduce the risk of soil compaction. In mechanized agriculture, manure, especially manure slurry, is often applied with heavy equipment. Single-axle manure tankers can weigh up to 18 t. Their use may cause considerable soil compaction, especially when soil moisture conditions are not ideal (Vero et al., 2014). Pyrolysis reduces the manure's mass by a factor of 5–10 depending on water content and provides the energy to dry it and concentrate the nutrients. Hence, biochar could be applied with lighter machines, lowering the risk of soil compaction.

5 | MANURE BIOCHAR AS A CARBON SINK

Pyrogenic carbon capture and storage, which includes the production and pyrolysis of biomass and the application of biochar to soils and materials, has been identified as one of the most promising negative emission technologies (Smith et al., 2019; Werner et al., 2018, 2022). To establish a carbon sink, CO_2 must be removed from the atmosphere, transformed into a stable form, and stored for a verifiable long time. Short-lived sinks are also beneficial for the climate but neither short-lived nor long-lived sinks can fully offset the global warming effect of fossil carbon emissions (Leifeld & Keel, 2022).

Carbon in manure originates from animal feed and usually straw-based bedding materials and was thus removed from the atmosphere via photosynthesis during the plant growth of feed and bedding products. Pyrolysis transforms the captured biomass carbon into pyrogenic carbonaceous matter, which are persistent forms of

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carbon when stored, for example, in soil. However, in most current biochar production systems, 40%–70% of the biomass carbon returns to the atmosphere as CO_2 because pyro-gas and oil are combusted to produce heat, or combined heat and power (Enders et al., 2012; Schmidt et al., 2019). Thus, only 30%–60% of the carbon originally fixed by photosynthesis remains stored in the biochar and define the C-sink potential, that is, the carbon content of the manure-based biochar. Moreover, both carbon expenditures, that is, GHG-emissions associated with production of biochar as well as the carbon fraction that decomposes after soil application within a relevant period of time, usually defined as the first 100 years (EBC, 2020) must be accounted for to assess the net negative emissions of PyCCS systems.

Carbon expenditures for the production of manure biochar can be accounted following the EBC Guidelines for the Certification of Biochar Based Carbon Sinks (EBC, 2020). To calculate the net negative emission, the total carbon expenditure of manure biochar production must be derived by summing up all these emissions and can be subtracted from the C-sink potential as CO_2e. Alternatively, the emissions must be compensated externally. Regarding carbon expenditures from pyrolysis and post-pyrolytic activities, manure-derived biochar can be assessed analogous to the production of biochar from vegetal biomass.

To claim carbon sink/net negative emission certificates for the offsetting of CO_2 emissions, only the carbon fraction persistent for more than 100 years shall be accounted for (EBC, 2020). Currently, the common C-sink certification consensus is to assume a persistent carbon fraction of 74% for biochars with an H to C_{org} ratio below 0.4, which accounts for an annual decay of 0.3% for 100 years (Budai et al., 2013; Camps-Arbestain et al., 2015; EBC, 2020). Applying pyrolysis temperatures of >500°C for a sufficiently long residence time, as recommended here to fully eliminate micropollutants and biological hazards, results in H to C_{org} ratios <0.4. However, Singh et al. (2012) reported that mean residence time (MRT) of wood, poultry litter, and cow manure biochar produced at 550°C were 1271, 396, and 313 years, which equals half-life of 881, 274, and 217 years, respectively. Thus, it takes 547, 118, and 93 years, respectively, to degrade 26% of the biochar-C so that the C-sink after this time still accounts for 74% of the initial biochar-C. Based on this study, the current method applied in the EBC C-sink guidelines is very conservative for wood-based biochars (calculated carbon sink underestimates actual carbon sink) and quite accurate for both poultry litter biochar and cow manure biochar. H to C molar ratio of the wood, poultry litter, and cow manure biochar in Singh et al. (2012) were 0.38, 0.55, and 0.72, respectively, that is, higher than 0.4 for the manure biochars. Steam was injected to the pyrolysis of cow manure and poultry litter, which is quite unusual. Wood biochar was produced with and without steam addition; here, this practice reduced the half-life from 1120 to 881 years. Thus, half-life and MRT of the cow manure and poultry litter biochar may be even higher in the absence of steam application. Lower recalcitrance of manure-based biochars in soil coincides with higher content of volatile carbon, which also correlates with higher ash content (determined at 950°C, Enders et al., 2012). A study actively varying ash content of wood-based biochar found a reduction of thermal stability with increasing ash content, which could not be explained by higher reactivity because of the incorporated (earth) alkali metals in the ash. Instead, ash-rich biomass may form relatively less stable carbon compounds (Grafmüller et al., 2022). Nevertheless, based on the calculations above, the current method used to calculate the persistent carbon fraction of biochar applied to soil may also be applicable to manure-based biochars. Biochars made from manure and produced at >500°C present C-contents of 47%–69% which correspond to C-sink potentials of 1.07–1.67 t CO_2e per ton of manure biochar considering only the persistent fraction of at least 74% (persistent over more than 100 years).

6 | CONCLUSIONS

The review revealed that biochar production from animal manure has the potential to overcome most of the environmental concerns associated with manure soil application. Applying a pyrolysis temperature of at least 550°C and a residence time that allows complete pyrolysis, which depends on feedstock particle size and pyrolysis technology used, converting manure into biochar eliminates any hazards related to pathogens, ARGs, and organic pollutants. However, considerably higher pyrolysis temperatures (> 600–650°C) should be avoided as they lead to unnecessary losses of nitrogen and further reduce the availability of phosphorus. Most PTEs and minerals are converted into less available forms during manure pyrolysis. Hence, manure biochars exhibit lower bioavailable PTEs and a more gradual release of nutrients than conventionally treated manure. However, from the point of view of a livestock farmer with a fixed output of livestock manure, there are insufficient data to draw reliable conclusions about the impact of manure pyrolysis on fertilization effects, crop growth, and yield. Hence, it is essential to design and conduct experiments in more practically relevant scenarios: Future research should compare biochar produced from a certain amount of manure with the direct application of the same quantity as manure. In contrast,

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there is clear evidence that pyrolysis of manure would significantly reduce GHG emissions and create long-term carbon sinks that can be certified under current certification schemes. The level of emission reduction, however, strongly depends on the design of manure preparation for pyrolysis, that is, when and how is it separated and/or dried and/or mixed with other feedstock. Since manure biochar performance and avoidance of manure-related environmental concerns are highly dependent on proper implementation of the pyrolysis process, rigid quality control is also essential for the political and societal acceptance of manure biochar.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The dataset used in this study is publicly available in the SI.

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