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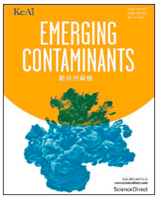

Veterinary antibiotics in contaminated animal manure: Fate, removal challenges, and future perspectives on conventional and emerging technologies

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ABSTRACT

The pollution caused by veterinary antibiotics (VAs) has become a global concern due to their role in promoting antimicrobial resistance in the environment. Animal manure, often referred to as animal slurry, contains substantial amounts of VAs originating from animal urine and feces. Since animal manure is commonly used as a nutrient-rich fertilizer, understanding the degradation of VAs and ensuring their efficient removal are essential for effective manure management. This article critically reviews the emerging technologies effective in VA removal, such as adsorption, membrane separation, advanced oxidation processes, carbonization, and bioelectrochemical systems. While these technologies have been extensively studied for their ability to remove pharmaceuticals from common water and wastewater, their applicability to real manure treatment remains insufficiently explored. This article outlines the challenges associated with each technology, particularly concerning the complex composition of animal manure. It highlights the potential of these technologies as supplementary or post-treatment options to improve VA removal, particularly during periods of high VA usage for therapeutic purposes. Finally, the article offers several recommendations, including advancements in animal manure collection, the importance of VA removal to mitigate antimicrobial resistance, and the potential for integrating multiple technologies to optimize VA removal.

1. Introduction

Modern livestock farming relies heavily on the effective use of veterinary antibiotics (VAs) in animal feeding. These substances can promote animal growth and prevent infectious diseases, thereby ensuring efficient livestock production. By 2030, global VA consumption is predicted to exceed 107,000 tons, with China, Brazil, and the United States being the top three consumers [1]. Currently, over 10 classes of VAs are used in animal farms for various purposes. In China, the total VA consumption surpassed 32,000 tons in 2021, with tetracyclines, penicillins, and macrolides being the most widely used [2]. According to the European Medicines Agency, in 2022, VA consumption across 31 EU countries averaged 73.9 mg/PCU (milligrams per population correction unit), with penicillins, tetracyclines, and sulfonamides accounting for 32.7 %, 23.5 %, and 9.4 % of total consumption, respectively [3]. Particularly, the consumption of VAs may be significantly higher in regions with limited farmer education due to misuse and overuse.

Most VAs administered to animals are poorly absorbed and excreted unchanged into urine and feces, leading to high concentrations of VAs remaining in animal manure. For many decades, VAs have been frequently detected worldwide, with their concentrations ranging from ng/L to μg/L in rivers, lakes, groundwater, and seawater, and from ng/kg to mg/kg in soil and sediments [4,5]. Due to their stability and biorefractory nature, most VAs can persist in the environment for long periods. In environments rich in VAs, microbes are likely to develop antibiotic resistance genes

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Abbreviations

AD	Anaerobic digestion	MIC	Minimum inhibitory concentration
AOP	Advanced oxidation process	MD	Membrane desalination
ARB	Antibiotic-resistant bacteria	MEC	Microbial electrolysis cell
ARG	Antibiotic resistance gene	MF	Microfiltration
BDD	Boron-doped diamond	MFC	Microbial fuel cell
BES	Bioelectrochemical system	NF	Nanofiltration
COD	Chemical oxygen demand	PCU	Population correction unit
DBP	Disinfection by-product	RO	Reverse osmosis
DOM	Dissolved organic matter	SS	Suspended solids
ED	Electrodialysis	TS	Total solids
EDR	Electrodialysis reversal	UF	Ultrafiltration
FO	Forward osmosis	VA	Veterinary antibiotic
HRT	Hydraulic retention time	VFA	Volatile fatty acid
HTC	Hydrothermal carbonization	VS	Volatile solids

(ARGs). As nucleic acid fragments, these substances can be transferred among microbial populations through transformation, transduction, and conjugation [6–8]. This process, known as horizontal gene transfer, can facilitate the spread of antimicrobial resistance throughout ecosystems, posing significant risks to human health. Recently, antibiotic-resistant bacteria (ARB) and ARGs have garnered increased attention due to global antibiotic pollution. In 2014, the World Health Organization identified ARGs as a critical indicator for monitoring water and soil to help address issues related to antibiotic misuse and overuse [9]. Since then, preventing antibiotic pollution and the proliferation of ARGs has become a global long-term task.

Animal manure is a valuable fertilizer; however, as a significant reservoir of VAs, it requires proper treatment to reduce VA levels and minimize its environmental impact. While anaerobic digestion (AD) and composting are commonly used in practice, they are not fully effective in removing all types of VAs. As a result, many emerging technologies have been explored to enhance further the removal of VAs, including adsorption, membrane separation, advanced oxidation process (AOP), carbonization, and bioelectrochemical system (BES). However, most of these technologies have been demonstrated only at the laboratory scale, and their practical applications face challenges from the complex composition of animal manure. The effectiveness of these methods in eliminating VAs remains uncertain, highlighting the need for further research and evaluation. Several key issues related to these technologies can be summarized as follows:

(1) *Effectiveness.* For real animal manure, the effects of salts, organic matter, and suspended particles on the effectiveness of these technologies remain unclear.
(2) *Applicability.* Effective VA removal may require combining conventional and emerging technologies, yet limited research has explored such integrated approaches.
(3) *Cost considerations.* Applying emerging technologies for VA removal necessitates further exploration of strategies to reduce overall costs.

Recently, many researchers have reviewed the degradation of VAs in conventional technologies [10,11], while others have evaluated the effectiveness of emerging technologies in removing VAs from municipal wastewater [12–14]. However, these reviews do not inform readers about the applicability of emerging technologies in treating animal manure. To fill this gap, this article critically reviews the challenges of applying emerging technologies to animal manure treatment, particularly concerning the VA usage patterns in animal farms and the complex composition of animal manure. It also discusses the optimal conditions for applying emerging technologies, their potential to mitigate antimicrobial resistance, and suggestions for reducing associated costs. These insights will contribute to advancing manure management and reducing VA release into the environment.

2. Generation and composition of animal manure

Anaerobic digestion and composting are generally suitable for processing liquid and solid manure, respectively, with their application closely linked to the manure collection methods. Various manure components can also influence the effectiveness of emerging technologies. Understanding the generation and composition of animal manure is crucial for the subsequent discussion on VA removal.

In animal farms, animal manure is typically collected in liquid form due to the intensive use of flushing water. This liquid mixture is transported outside the animal housing for storage, where sedimentation and hydrolysis occur, as shown in Fig. 1. After a specific storage period, solid-liquid separation is employed to separate the manure into liquid and solid fractions for different treatments. Many separation methods, including gravity-based separation and filtration, have been tested for fresh and aged manure, while decanter centrifuge is the most commonly used [15,16]. However, only a maximum of 4100 g centrifuge force has been reported as feasible, indicating that separating small particles remains insufficient [15]. The addition of flocculants can improve separation efficiency, while using coagulants, such as Fe/Al-salt, is discouraged due to their high dosages (up to 15 g/L) [17]. Carbonates in animal manure can consume coagulants and produce massive fine bubbles, negatively impacting separation efficiency. Although many farms now collect solid manure directly from the floor to reduce the use of flushing water, liquid manure is still produced in some cases. For instance, AD requires a well-mixed combination of water and solid manure, which can generate liquid digestate.

Animal manure consists of water, salts, nutrients, sediments, suspended particles, and dissolved organic matter (DOM) derived from urine, feces, and spilled food. As shown in Table S1, the water content typically ranges between 91 % and 98 %, and further increases to 96–99 % after solid-liquid separation [18]. The volatile solids (VS) to total solids (TS) ratio is typically between 60 % and 80 %. The DOM in animal manure includes carbohydrates, volatile fatty acids (VFAs), lipids, proteins, humic acids, cellulose, lignin, and feeding residues, contributing to a chemical oxygen demand (COD) ranging from 25.3 to 130.8 g/L [19]. However, humic acids, cellulose, and lignin are highly resistant to degradation, resulting in a residual COD of up to 2.9 g/L even after complete AD [20]. Our previous study analyzed the composition of digested pig manure, revealing that the DOM in the following molecular ranges accounts for the indicated percentages of COD: 0–1 kDa (1.2 %), 1–5 kDa (5.0 %),

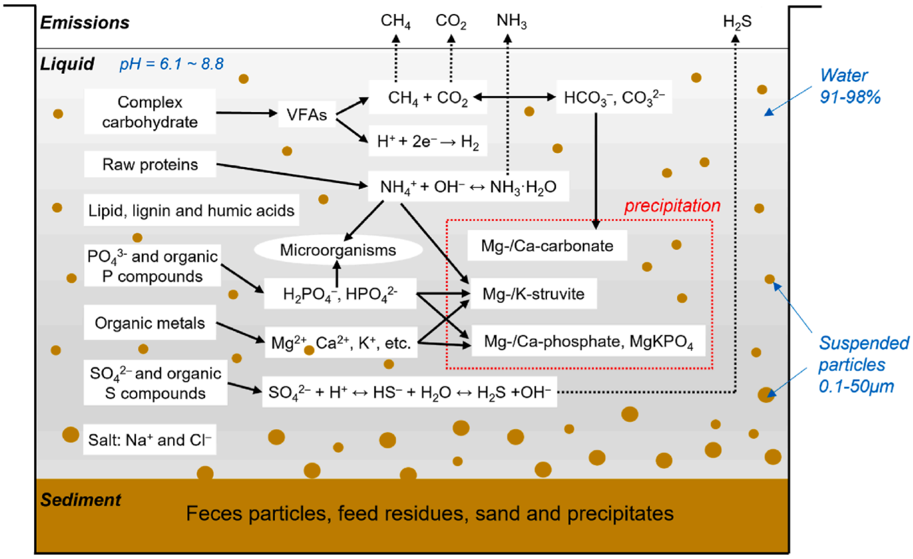

Fig. 1. Composition and evolution of animal manure in the storage tank.

5–10 kDa (2.9 %), 10–50 kDa (8.8 %), 50 kDa to 100 nm (5.7 %), and >100 nm (75.2 %) [21]. The inorganic matter includes N, P, metals (e.g., Na^+, K^+, Mg^{2+}, and Ca^{2+}), and other ions (e.g., Cl^-, SO_4^{2-}, HCO_3^-, and CO_3^{2-}) derived from roughage and salt intake. The concentration of total N in raw animal manure typically ranges from 1.2 to 6.7 g/L, while most of them are transformed into NH_4^+/NH_3 during storage [22]. Phosphorus ranges from 0.4 to 2.7 g/L, while 80–90 % can precipitate with metals, forming amorphous Mg-/K-struvite and Ca-phosphate in the sediments. The Mg^{2+} and Ca^{2+} in the liquid fraction were reported as 0.1 g/L each, but when the pH dropped to 5.0, their concentrations increased to 0.5 and 2.0 g/L, respectively [23]. Na^+, K^+, and Cl^- concentrations are typically up to 0.8, 2.0, and 1.5 g/L, respectively, making them the dominant salt ions in animal manure [24]. Carbonate originates from carbonate intake from feedstock and the dissolution of CO_2. The alkalinity in $CaCO_3$ has been reported to range in 1.5–3.1 g/L but increases to 2.7–4.5 g/L after AD [25]. The pH of animal manure is typically table at 6.1–8.8, with a high buffer capacity due to three components: VFAs (pH 4–6), H_2CO_3/HCO_3^- (pH 5–8), and NH_4^+/NH_3 and HCO_3^-/CO_3^{2-} (pH 8–12).

3. Concentration and distribution of VAs

The frequently detected VAs in animal manure include sulfonamides, tetracyclines, penicillins, fluoroquinolones, macrolides, trimethoprim, and lincosamides, and their basic information is shown in Table S2. Previous studies have often reported high levels of VA contamination in animal manure. In pig manure, up to 9 mg/L (in volume) of sulfamethazine and 4.2 mg/L of chlortetracycline have been detected [26]. Other studies also reported oxytetracycline contamination as high as 136 mg/kg (in dry weight, dw) [27], and fluoroquinolones are often found ranging from several to thousands of mg/kg dw [28]. In addition, the concentration and diversity of VAs in animal manure are likely influenced by the animal's growth stages, which include the starting, feeding, finishing, and breeding phases. When used as growth promoters, VAs are typically used continuously from the starting to finishing phases. In pig farming, 88 % of VAs are applied during the feeding and finishing phases, corresponding to 30–70 and 70–180 days after birth, respectively [29,30]. Poultry and cattle farming follow similar growth and VA usage patterns [31]. It is noteworthy that many countries have banned the use of VAs as growth promoters, making the use of VAs highly related to treating diseases. However, the dosages of VAs used for therapeutic purposes are higher than for growth promotion. Its influence on VA contamination and manure management will be discussed later in this article.

Veterinary antibiotics can be present in solid and liquid fractions, but their precise distributions vary due to VA dosage, feeding methods, and water content. According to existing literature, most studies report VAs in the liquid fraction using μg/L, but in the solid fraction using μg/kg dw, making it difficult to compare their concentrations. This study developed a solid-liquid distribution model with TS varying from 3 % to 7 %, as shown in Fig. 2. Incorporating data from the literature into this model reveals that the distribution of VAs is highly dependent on VA species and total concentrations [32–37]. For instance, tetracyclines at μg/L concentrations are readily sorbed onto the solid fraction, with only 0.5–14.7 % remaining in the liquid fraction. However, if their concentrations increase to mg/L levels, the proportion in the liquid fraction rises to 23.5–88.9 %. This trend is reasonable as the adsorption capacity of VAs depends highly on the TS content, which remains stable throughout the storage period. Sulfonamides and lincomycin are less likely to be sorbed by the solids, with 28.3–70.9 % and 25.9–73.9 % remaining in the liquid fraction, respectively, even at μg/L concentrations. Notably, the adsorption of VAs occurs not only on sediments but also on suspended particles, suggesting that the concentrations of VAs in the liquid fraction may be far higher than the reported values in the literature.

4. The fate of VAs in conventional treatments

4.1. Anaerobic digestion

Anaerobic digestion produces biogas comprising 60–70 % CH_4 and 30–40 % CO_2. The digested manure, known as digestate, is less odorous and pathogenic but richer in nutrients than raw manure. AD is categorized into wet and dry types based on the TS content of the feedstock. Wet AD is generally preferred due to its fast mass transfer, quick biogas production, and low operational costs, so large-scale farms typically use wet AD for manure disposal. In this process, liquid manure can be directly used as the feedstock, while solid manure requires additional water supplementation to

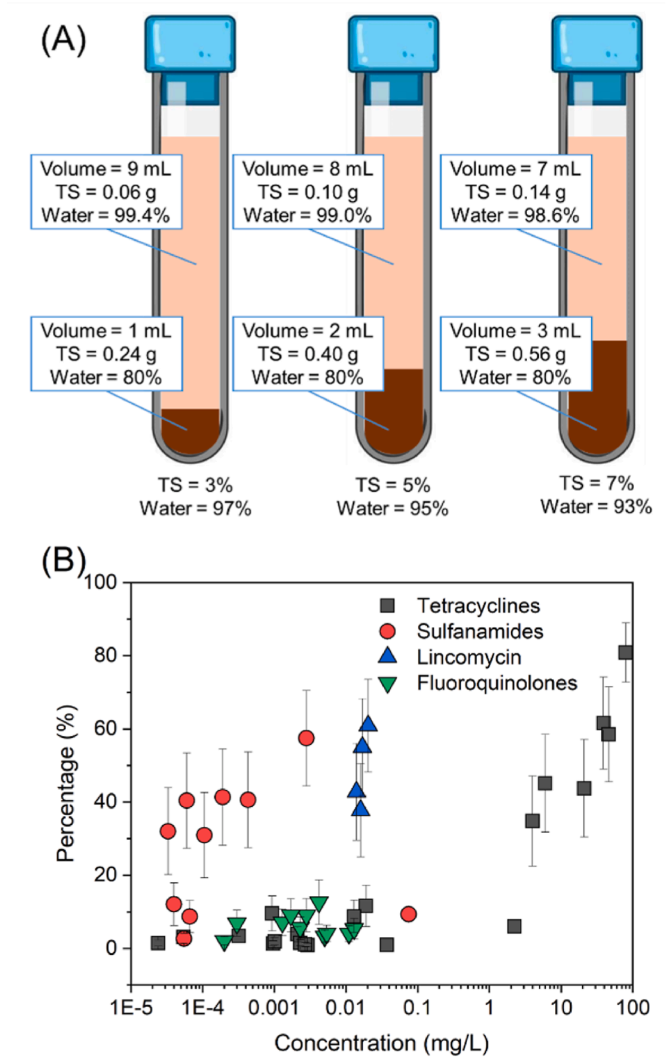

Fig. 2. The solid-liquid separation model (A), and percentage of VAs in the liquid fraction (B) based on the animal manure with TS varying from 3 % to 7 %.

achieve the desired water content [38]. Dry AD offers smaller reactor volume, lower heating requirements, and faster pathogen inactivation [39]. However, it requires complex pre- and post-treatment processes and a more extended digestion period, which can limit its large-scale application.

Anaerobic digestion can partially degrade VAs or convert them into less toxic compounds. Penicillins, for example, are easily degraded and do not pose significant removal challenges. In contrast, tetracyclines, sulfonamides, and macrolides can persist in AD for extended periods. Fig. 3A illustrates the removal efficiencies of VAs during AD in different studies since 2010. The degradation of VAs is highly dependent on the specific VA species and concentrations. Most sulfonamides show limited degradation rates, with some studies reporting no significant decline in their concentration during AD. Tetracyclines and fluoroquinolones typically exhibit degradation rates ranging from 50 % to 90 %. Yin et al. noted that higher concentrations of tetracyclines resulted in poorer removal rates, likely due to the more potent suppression of microbial activity [40,41]. Macrolides, tylosin, and florfenicol exhibit high removal efficiencies but often transform into other metabolites that persist in the reactor [42]. Overall, removing most VAs during wet AD is not efficient. A recent study demonstrated that

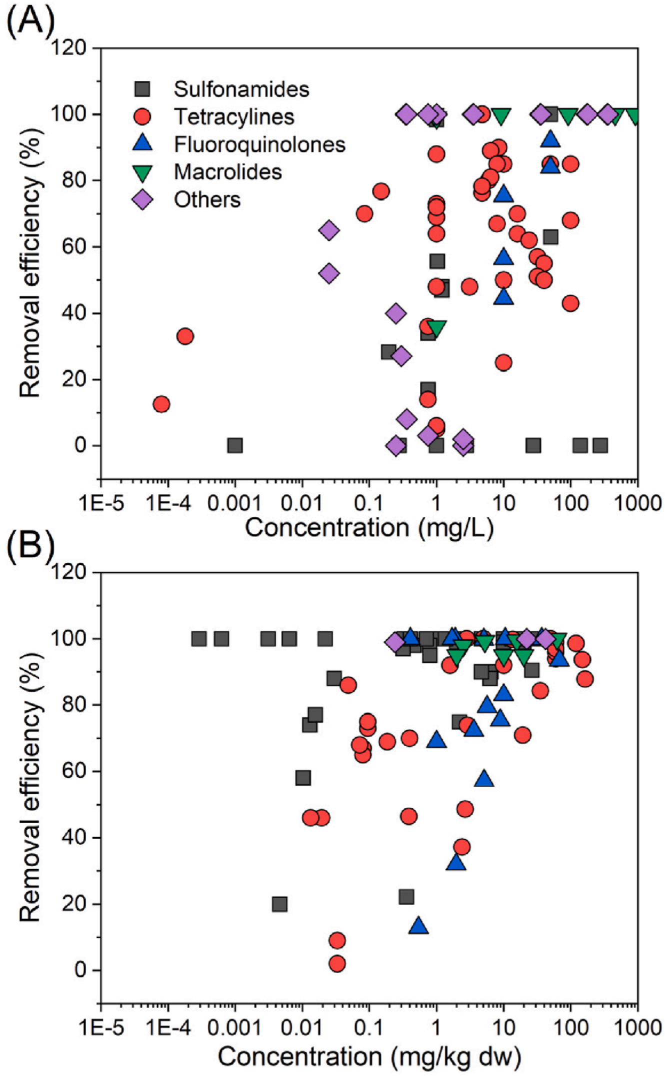

Fig. 3. Removal efficiency versus concentration for different VAs during AD (A) and composting (B) since 2010. All the data was obtained from the literature in Tables S3 and S4.

VA removal in wet AD is much lower compared to an aerobic batch reactor [43], suggesting that additional extension or post-treatment methods are necessary to improve VA removal from AD. Interestingly, dry AD appears to achieve a higher degradation of sulfonamides than wet AD, with removal rates ranging from 40 % to 90 % [44]. However, the number of studies on dry AD is limited, as its application is less frequent in practice.

4.2. Composting

Composting is only applicable for solid manure treatment. It involves three successive phases: mesophilic (25–40 °C), thermophilic (40–65 °C), and maturation phases (10–40 °C). The optimal conditions include a carbon-to-nitrogen (C/N) ratio in the range of 20–25, with moisture content ideally between 40 % and 60 %. One notable aspect of composting is the potential for mixing domestic waste (such as rice straw and husk) with animal manure, which helps to adjust the C/N ratio and maintain optimal moisture [45]. Compared to AD, composting exhibits a higher degradation of organic matter due to the significantly faster growth of aerobic microbes (6–24 generations per day) compared to anaerobic microbes (0.2–0.4 generations per day). However, composting of animal manure causes undesired emissions of CO_2 and NH_3. The loss of NH_3 to the atmosphere also results in a reduced fertilization value of the composting residues.

Fig. 3B summarizes the removal of VAs during composting, as reported in studies since 2010. The removal of certain VAs is generally more efficient than those observed in AD. Notably, sulfonamides were rapidly removed during composting, in contrast to their poor removal in AD. This may be attributed to the higher temperatures (up to 65 °C) and the greater microbial diversity in compost piles. However, tetracyclines and fluoroquinolones showed slower degradation compared to sulfonamides. Several studies have reported that composting at 25 °C led to very slow degradation of VAs, suggesting that temperature plays a critical role in the breakdown of these compounds [46,47]. Unlike AD, the concentration of VAs has less impact on composting efficiency. Even with high concentrations of sulfonamides and tetracyclines used, only mild inhibition of composting occurs, indicating that aerobic microbes are less susceptible to the inhibitory effects of VAs than anaerobic microbes [48]. It has also been reported that the presence of heavy metals, such as Cu^{2+}, can inhibit the degradation of VAs during composting, while the addition of lime or activated carbon can alleviate this inhibition [49]. In practice, AD and composting are sometimes used together to maximize their environmental benefits [50,51]. The solid fraction of digestate can be further composted before being applied to land. It helps to enhance the removal of VAs from animal manure, contributing to more sustainable waste management practices.

5. Emerging technologies and challenges

Overall, AD and composting are ineffective in removing all types of VAs from animal manure. In particular, AD is less effective in removing VAs from the liquid manure. During intensive use of VAs in animal farms, the high concentration of VAs can also cause shock suppression stress to biological processes, leading to poor removal efficiencies. In such cases, additional treatments may be considered to enhance the removal of VAs further.

5.1. Adsorption

To date, two main categories of adsorbents have been used for the removal of VAs: (1) carbon-based materials, including activated carbon, carbon nanotubes, graphene, and biochar, and (2) clay minerals, including montmorillonite, kaolinite, and zeolite. Previous studies have reported adsorption capacities for tetracyclines, sulfonamides, and fluoroquinolones on activated carbon of up to 588, 282, and 613 mg/g, respectively, while carbon nanotubes demonstrated 1530, 510, and 933 mg/g for the same VAs [52]. Despite the high adsorption capacities, cost reduction and recyclability remain significant challenges, especially for high-value nanomaterials. Therefore, using cheap materials to remove the VAs from complex wastewater remains a widely accepted approach in engineering applications. Biochar, derived from carbonization of wood, straw, yard trimmings, and animal feces, is a cost-effective adsorbent. Recently, there has been growing interest in using biochar and modified biochar to remove VAs [53–57]. Biochar typically has a surface area ranging from 20 to 200 m^2/g, with a maximum adsorption capacity for various VAs reported to be 12 mg/g [58,59]. Despite its low adsorption capacity, biochar production is over 10 to 10,000 times cheaper than activated carbon and nanomaterials [60]. However, it is more fragile than activated carbon, making it more suitable for soil remediation. Its porous structure enables efficient soil retention of nutrients, heavy metals, and VAs. Clay minerals also play significant roles in VA adsorption, with ion exchange, π-π interactions, and hydrogen bonding being the key mechanisms. Montmorillonite has an ion-exchange capacity of 0.95 mmol/g and a BET surface area of 35 m^2/g, allowing it to adsorb up to 298 mg/g of ciprofloxacin and 400 mg/g of tetracycline [61,62]. Zeolite, with a higher BET surface area up to 914 m^2/g, generally exhibits lower adsorption capacities, typically below 100 mg/g [63]. The ionization of VAs under different pH conditions can alter their interactions with clay minerals. Maintaining the pH within an optimal range ensures that the ionization of VAs opposes the surface charge of clay minerals and thus makes high adsorption capacities.

Most studies have simulated real conditions using synthetic wastewater spiked with salts and humic acid to investigate the specific components affecting adsorption. As shown in Fig. 4, the adsorption of VAs can be affected by foreign ions and DOM. While Cu^{2+} and Zn^{2+} can bridge the interaction between VAs and adsorbents [64], increasing heavy metals in animal feeds is unfeasible. In most cases, foreign ions can inhibit the adsorption rate due to competitive adsorption, complexation, and electrostatic screening effects. It has been reported that 40 mg/L of NH_4^+ decreased the adsorption of levofloxacin on zeolite by 58 %, primarily due to competitive adsorption, while 400 mg/L of Ca^{2+} and Mg^{2+} reduced the adsorption of levofloxacin by 50 % due to complexation [65]. Na^+ and Cl^- typically only create an electrostatic screening effect, which can generate a double electric layer on adsorbent surfaces and repel ionized VAs. Their poor binding with activated carbon and VAs makes competitive adsorption and complexation reactions less likely. A previous study has shown that 0–0.5 mol/L NaCl slightly reduced sulfadiazine adsorption on graphene due to the electrostatic screening effect but with no influence on activated carbon [66]. Humic acid has a strong affinity for VAs through hydrogen bonding, which can potentially bridge the adsorption process [67,68]. One study found that 100 mg/L of humic acid slightly increased the adsorption of tetracycline on kaolinite under acidic conditions but showed minimal effect under neutral conditions [69]. In another study, humic acid increased the adsorption of fluoroquinolones on biochar [70]. However, many other studies reported its suppression of adsorption [65,71,72]. The inconsistency in these findings may be attributed to using different types of humic acids. It has been reported that competitive adsorption becomes more significant when organic molecules approach the pore size in adsorbents [73]. Molecules larger than the pore size can be entirely excluded, while molecules close in size to the pores are more likely to be trapped. Unlike the humic acid used in these studies, the DOM in animal manure can range from hundreds to millions of Da, which can severely block various pores of different sizes. Notably, colloidal particles can clog adsorbents or compete for the adsorption of VAs. Their difficult removal from the liquid fraction often results in low VA removal efficiency.

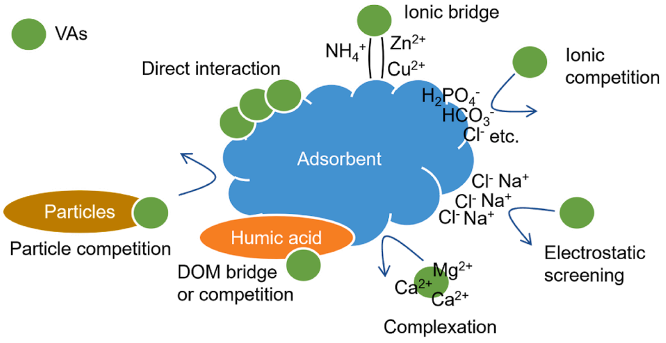

Fig. 4. Different factors influencing the adsorption of VAs in animal manure.

Overall, adsorptive removal of VAs from animal manure presents greater challenges due to three mechanisms: (1) competitive adsorption, complexation, and electrostatic screening caused by multiple ions; (2) pore blocking caused by DOM; and (3) the binding of colloidal particles with VAs. Converting raw manure into digestate does not significantly alleviate these issues, as most challenges remain unchanged. However, digested manure produces less odor than raw manure, making the reuse of adsorbents more acceptable. In addition, applying inexpensive adsorbents along with the spreading of digested manure on land provides a promising approach. However, this method does not lead to the degradation of VAs. Despite its ability to immobilize VAs, the long-term risk regarding antimicrobial resistance requires further study.

5.2. Membrane separation

Pressure-driven membrane technologies, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), are commonly used for separating contaminants from liquids. MF and UF employ porous membranes to remove particles larger than 0.45 μm and 0.1 μm, respectively. VAs can pass through the pores along with water, salts, and DOM in these processes, while larger particles remain on the membranes. As shown in Fig. 5, retention of VAs on MF and UF is difficult, as their pore size is far larger than the molecular size of VAs. However, they still present 18.6–89.6 % removal rates because of the adsorption of membrane base and large organic matter [74,75]. Due to processing defects and membrane heterogeneity, commercial membranes exhibit a lognormal pore size distribution, and their interior structures may contain interlaced configurations [76]. Substances whose size can match the membrane pores can inevitably reduce separation efficiency. For MF and UF, pore blockage is primarily caused by colloidal particles. Under hydraulic pressure, these particles can quickly embed into the membrane pores, forming a fouling cake on the membrane surface. NF and RO membranes can retain VAs, as the molecular weight of VAs generally exceeds 200 Da (except for loose NF membranes). RO can produce pure water, whereas NF allows salts and nutrients to pass. In practice, MF, UF, NF, and RO are often used in combination. For example, an MF/UF-RO system has been applied to treat pharmaceutical wastewater, resulting in an MF/UF permeate containing VAs and an RO permeate free of VAs [77,78]. However, these technologies are rarely employed for treating animal manure. One study used vibrating RO to concentrate the nutrients from pig slurry, but long-term assessment is still lacking [79]. Another study used MF/UF-RO to treat digested pig manure with SS ranging from 0.06 % to 0.22 %, observing a 40 % decrease in flux within 1 h [80,81]. However, this SS content was low compared to most studies, likely because the slurry was derived from flushing wastewater, which had not been mixed with the majority of solid manure. Overall, membrane pore blockage caused by the transmembrane pressure difference is the primary challenge. Although Ca^{2+} and Mg^{2+} can potentially form membrane scaling, the membranes typically become blocked in a short time, making the duration insufficient for significant scaling formation.

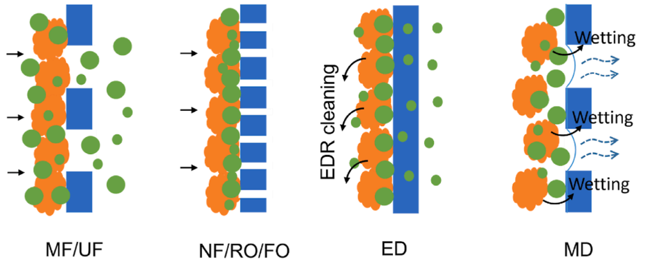

Fig. 5. Schematics of VAs retention in different membrane technologies.

Non-pressure-driven membrane technologies include electrodialysis (ED), membrane distillation (MD), and forward osmosis (FO). ED utilizes an electric field to drive ions passing ion-exchange membranes, enabling the separation and concentration of nutrients from the feed solution. It is suitable for treating water with 0.5–10 g/L of salinity. Colloidal particles and DOM can foul the anion-exchange membrane, while Ca^{2+} and Mg^{2+} may form scaling on the cation-exchange membrane. In practice, electrodialysis reversal (EDR) with electrodes reversed at 15–30 min intervals is often employed to mitigate membrane fouling and scaling. Our previous study demonstrated that EDR can recover nutrients from digested manure, with only a slight decline in membrane resistance and ion-exchange capacity after 770 L/m^2 of treating load [20], and stable ion-exchange capacity was observed even after 5000 L/m^2 of treating load over 8 months [82]. The DOM smaller than 10 kDa is responsible for membrane fouling, indicating that removing colloidal particles is not critical for ED [21]. Depending on their ionization properties, VAs can migrate toward either the anode or cathode in this process. Large VAs, such as tetracyclines, are typically retained on the membrane surface, while small VAs, such as sulfonamides, can pass membranes slowly. However, this does not mean VAs can be efficiently removed in ED, as their migrations depend on their concentrations and electric field strength. When desalination approaches completion, VAs are usually removed less efficiently due to the low current density [83]. In addition, zwitterionic VAs, such as tetracycline, exhibit a high affinity for humic acid, leading to its spatial associations with humic acid. Consequently, around 36 % of tetracycline and 51 % of sulfadiazine remained in the feed solution during EDR, compared to 12 % and 36 % in conventional ED, respectively [84].

Membrane distillation utilizes thermodynamic pressure to recover water and other volatile substances. This process involves maintaining a high temperature of 40–70 °C to drive vapor passing a hydrophobic membrane (with pore sizes ranging from 0.2 to 20 μm) and condensing on a cooling surface [85]. If the pH exceeds 10, NH_3 can be effectively removed along with water evaporation [86]. MD has been shown to reject nearly 100 % of negatively charged VAs while allowing 22 % of positively charged tobramycin to pass, indicating that positively charged substances may cause permeate impurities [87]. Moreover, membrane wetting from semi-hydrophobic compounds, such as phenols, proteins, and humic acids, poses significant challenges. These substances can reduce the hydrophobicity of the MD membrane, which in turn facilitates the adsorption of hydrophilic substances. This creates a medium that connects the membrane base to hydrophilic substances, leading to severe membrane fouling. For example, in a laboratory-scale MD system treating digested manure, the flux remained stable at 17.5 $L/m^2/h$ during the first 24 h but decreased to 5 $L/m^2/h$ after 72 h [88]. Similar results were observed in another study treating raw animal manure, where MD flux decreased by 90 % over 25 h of operation [89]. Although membrane cleaning with citric acid and NaClO recovered 75.5 % of the flux, long-term fouling remains a significant challenge [90]. Additionally, MD requires pre-treatment with MF or UF to minimize the impact of particles [91], making it less suitable for long-term treatment compared to ED.

Similar to RO, FO uses semi-permeable membranes but relies on natural osmotic pressure as the driving force. In this process, water migrates from a feed solution to a draw solution with high salt concentrations. FO membranes are generally thinner, less tortuous, and more porous than RO membranes, as they are designed to minimize water diffusion paths [92]. This design results in poorer mechanical properties, making FO membranes more susceptible to wear and cracking during operation. From an energy consumption perspective, the diluted draw solution must be re-concentrated to maintain high salinity, which requires more energy than RO or MD alone. Another drawback of FO is reverse solute flux, which has been reported at 14.1 and 3.98 $g/m^2/h$ for two commonly used membranes [93]. Although FO membranes tend to experience less fouling than RO due to their spontaneous diffusion process [94], many studies suggest that FO actually faces more severe fouling than RO, leading to inconsistent research findings [95]. In most studies, membrane cleaning does not fully recover flux to 100 % [93,96]. Currently, there is a lack of long-term FO testing in the treatment of animal manure. Similar to RO and MD, FO cannot separate VAs from the feed solution. Since the primary goal of manure treatment is not to produce pure water, applying NF, RO, MD, and FO remains contentious.

5.3. Advanced oxidation processes

Advanced oxidation processes, such as ozonation, Fenton, persulfate oxidation, photolysis, electro-oxidation, and their hybrid variants, can be established to remove VAs from animal manure, as summarized in Table 1 [97–101]. Ozonation can produce reactive O and $O_2^{\cdot-}$. Under certain specific conditions, such as high temperatures, UV radiation, and catalysts provided, $^{\cdot}OH$ can also be formed. However, the mineralization of VAs using this method ranges from 30 % to 80 %, with stable small organic molecules, such as acetic, oxalic, and oxamic acids, being formed as by-products [102,103]. One study used ozonation as a pretreatment step to increase the biodegradability of organic waste, and it significantly enhanced the CH_4 yield [104]. This improvement is attributed to the ability of reactive oxidation species to break down large organic molecules into more biologically accessible forms. Fenton process relies on the reaction between Fe^{2+} and H_2O_2 to generate $^{\cdot}OH$ under acidic conditions (pH 2.5–4.5). However, in practice, neither the Fenton nor the modified Fenton are suitable for treating animal manure due to the difficulties in acidifying animal manure. Persulfate-based oxidation can produce $SO_4^{\cdot-}$ as the primary reactive radical, co-existing with $^{\cdot}OH$ and $O_2^{\cdot-}$, and non-radical species like $^1O_2$. This process can be activated by heat, ultraviolet light, ultrasound, electricity, carbon materials, and transition metal oxides. Compared to the Fenton process, $SO_4^{\cdot-}$ can be generated over a wider pH range, although it can react with OH^- and form $^{\cdot}OH$ under alkaline conditions [105]. In addition, the reaction between DOM and $SO_4^{\cdot-}$ is an order of magnitude slower than that between DOM and $^{\cdot}OH$ [106], indicating that more radicals remain available for decomposing target VAs in this process.

Photolysis alone is relatively ineffective but can be significantly enhanced through catalysis using metallic oxides, such as TiO_2, ZnO, and CuO, forming what is known as photocatalysis. Photocatalysis narrows the energy bandgap of metallic oxides, enabling the generation of h^+ after losing e^-, which then efficiently oxidizes H_2O and OH^- into $^{\cdot}OH$. However, photolysis and photocatalysis face challenges when treating animal manure due to poor light penetration in turbid liquids rich in suspended particles. Humic acid exhibits both photon filtration and photosensitization effects, which can stimulate the generation of $^{\cdot}OH$. The addition of 1 mg/L humic acid can double the $^{\cdot}OH$ yield during photolysis; however, it can clog TiO_2 surfaces, thereby suppressing $^{\cdot}OH$ generation during photocatalysis [107]. Electrode materials critically influence the performance of electro-oxidation. Titanium coated with ruthenium dioxide (Ti/RuO_2), platinum, and boron-doped diamond (BDD) anodes have shown significant removals of various micro-contaminants. Among these, BDD anodes are the most expensive but also the most efficient, achieving up to 90 % mineralization of VAs due to their high oxygen evolution potential (2.7 V) [108]. The generated $^{\cdot}OH$ on the BDD anode can be efficiently released into the solution, unlike Ti/RuO_2 anodes, which exhibit only 35 %

Table 1
Summary of AOPs effective for VAs oxidation.

Conventional AOPs	Required materials	Oxidation mechanisms	Advantages	Disadvantages	Hybrid AOPs
Ozonation	1–4 % of O_3	(1) O and $O_2^{\cdot-}$; (2) O_3 generates $^{\cdot}OH$.	Wide pH adaption	(1) High equipment and operational costs; (2) Less soluble in water; (3) Disposal of end gas.	$Ozonation/H_2O_2$; Photocatalytic ozonation.
Fenton	H_2O_2 and Fe^{2+}	H_2O_2 produces $^{\cdot}OH$ in the presence of Fe^{2+}.	Coagulation caused by Fe^{3+}	(1) $Fe(OH)_3$ wastes generated; (2) pH restrictions in 2.5–4.5.	Electro-Fenton; Photo-Fenton; Photo-electro-Fenton.
Persulfate	Persulfate	(1) Generation of $SO_4^{\cdot-}$ and other radicals.	(1) Wide pH adaption; (2) less reactive with other DOM; (3) lower costs on storage and transportation	Rely on activation methods.	Various physical and chemical methods can be used for activation.
Photolysis	Solar light, UV or γ radiation	(1) Absorption of photos by VAs; (2) Photos loss from photosensitizers.	(1) Wide pH adaption; (2) No chemicals required.	(1) Slow degradation; (2) Poor generation of $^{\cdot}OH$; (3) Low light penetration in turbid water.	$Photolysis/H_2O_2$; Photocatalysis; $Photocatalysis/H_2O_2$;
Electro-oxidation	Non-sacrificial electrodes	(1) Direct oxidation; (2) Production of $^{\cdot}OH$.	(1) Wide pH adaption; (2) No chemicals required	(1) Expensive electrode materials; (2) Limited electrode area. (3) Producing Cl_2.	Electro-chlorination.

mineralization due to the adsorption of $^{\cdot}OH$ onto the anode [109]. In addition, electro-oxidation is highly reactive with DOM, and complete mineralization of humic acid has been reported after 7 h of oxidation using BDD anodes [110]. However, it is noteworthy that Cl_2 can be produced during electro-oxidation, which is undesired for the treatment of animal manure.

Various substances in animal manure can influence the effectiveness of AOPs. NH_4^+/NH_3 can react with multiple radicals to form nitrate, with reaction rates of $^{\cdot}OH$ and $SO_4^{\cdot -}$ with NH_3 ($\sim 10^7$ $M^{-1}s^{-1}$) being much higher than with NH_4^+ ($\sim 10^5$ $M^{-1}s^{-1}$) [111]. Additionally, $^{\cdot}OH$ and $SO_4^{\cdot -}$ can react with Cl^- to generate $ClO^{\cdot}$ and $Cl^{\cdot}$ ($\sim 10^8 - 10^9$ $M^{-1}s^{-1}$), while carbonate as a radical scavenger can produce less reactive $CO_3^{\cdot -}$ ($\sim 10^8$ $M^{-1}s^{-1}$) and $HCO_3^{\cdot}$ ($\sim 10^6$ $M^{-1}s^{-1}$) [112]. Certain DOMs, such as phenols, proteins, and humic acids, can also react with radicals. For example, humic acid has been reported to achieve a removal efficiency of 94.15 % through persulfate oxidation [113]. However, ozonation, while capable of decolorizing manure, typically achieves only low mineralization of humic acids (20–60 %) [102]. These competitive reactions can increase the demand for AOP reagents. Our previous study on electrode oxidation demonstrated that the addition of digestate extended the time required for complete VA removal from 5 min to 60 min [114]. However, humic acids were only reduced by approximately 20 %, suggesting that VAs are more preferentially oxidized than humic acids. Apparently, digested manure is more suitable for AOP treatment than raw manure due to its lower COD content. The dosage of AOP reagents should be optimized to ensure efficient removal of VAs without fully oxidizing all DOM. Specifically, humic acids should be preserved as valuable organic fertilizers rather than fully oxidized. In this regard, $SO_4^{\cdot -}$ offers an advantage because their reactivity with DOM (10^6–10^7 $M^{-1}s^{-1}$) is weaker compared to $^{\cdot}OH$ (10^8–10^9 $M^{-1}s^{-1}$) [112].

5.4. Carbonization

Pyrolysis and hydrothermal carbonization (HTC) are widely studied methods for producing biochar from animal manure. Pyrolysis requires extremely high temperatures, typically ranging from several hundred to over 1000 °C. Only materials with low moisture content can be used as feedstock, indicating that liquid manure is unsuitable for pyrolysis. VAs in animal manure can be thermally mineralized during pyrolysis, similar to other organic matter. One study has shown that tetracyclines and sulfonamides are completely removed during pyrolysis of animal manure at temperatures around 600 °C, while heavy metals are significantly immobilized in the resulting biochar [115]. However, pyrolysis can release NH_3 into the atmosphere, which is undesirable compared to biological treatments. HTC operates at much milder temperatures, typically between 160 °C and 250 °C, with a heating duration of several hours. This process can handle semi-solid or liquid waste, meaning that manure does not need to be dried before being fed into the reactor. As the temperature increases, HTC generates more hydrophobic substances. NH_3 and other volatile compounds can be retained within the reactor, preventing their release into the atmosphere. HTC also effectively reduces suspended particles by altering their hydrophilicity [116], making solid-liquid separation much easier. Like pyrolysis, tetracyclines and sulfonamides can be entirely destroyed during HTC, and 69–82 % of organic carbon is fully mineralized, demonstrating an effective removal of VAs using this technology [117]. While the liquid by-product of HTC may exhibit biotoxicity, its nutrients and DOM can be used as fertilizer to promote crop growth after proper dilution [118]. One advantage of carbonization over other emerging technologies is that the complex composition of animal manure does not lead to process inhibition. Despite their high energy consumption, carbonization treatments can be effectively implemented at sites with excess heat energy. In addition, applying carbonization to raw manure is time-efficient and straightforward, as it avoids prolonged treatments required for AD and composting.

5.5. Bioelectrochemical system

Bioelectrochemical systems (BES) can be configured in single- or two-chamber designs. In a microbial fuel cell (MFC), microbes use DOM as a carbon source to drive electron movement in an external circuit, thereby converting bioenergy into electrical energy. In a microbial electrolysis cell (MEC), an external voltage is applied to stimulate electrode reactions, such as H_2 production at the cathode. Raw animal manure, rather than its digested form, can be fed to BES because it contains abundant DOM and nutrients. Numerous studies have shown that BES can achieve 70–99 % removal of VAs, primarily due to the activity of exoelectrogenic microbes on the bioanode and ambient microbes in the reactor [119]. For example, the degradation of ceftriaxone, sulfonamides, and tetracyclines in MFCs has been reported to range from 80 % to 99 %, significantly higher than the 20 %–63 % degradation observed in control AD systems [120,121]. Yang et al. also reported a higher removal of sulfamethoxazole in MFCs than in open-circuit MFCs [122]. Interestingly, the suppression of suspended methanogens reduced sulfadiazine removal, emphasizing the crucial role of ambient microbes in VA degradation [123]. However, VA removal is typically less efficient during the start-up of BES. For instance, during a 10-month acclimation period, the removal of sulfamethoxazole was initially 20 % but eventually reached 80 %. [124]. In addition, an oligotrophic environment can force microbes to derive nutrients and carbon from VAs. Yan et al. reported a significant increase in VA removal when acetate was replaced with oxytetracycline as the carbon source in MFCs [120]. Our previous study also showed that the degradation of roxarsone was significantly enhanced in BES when acetate was absent [125]. Acetate, a readily available carbon source, can directly participate in the tricarboxylic acid cycle or methane production, requiring less enzymatic involvement and lower energy than the cleavage of VAs. It is well established that anode potential influences the oxidation rate, while the ambient redox potential also contributes to the VA degradation. For instance, acetate concentrations above 556 mg/L can create a reducing environment with low redox potential, which competes with micro-contaminant degradation on the bioanode [126]. However, further research is needed to understand this competition better in real animal manure, as it contains more DOM than synthetic wastewater. On the biocathode, VAs can be reduced using the cathode as an electron donor. The reduction products typically exhibit similar structures to the parent VAs but with lower toxicity. For instance, biocathodes can dechlorinate many VAs containing -Cl through both microbial and abiotic reduction [127], and the $-NO_2$ on the aromatic ring can be reduced to an $-NH_2$ [128]. However, only these two types of reductions without cleavage of the structure have been reported so far.

Specific components in animal manure may inhibit the performance of BES. For instance, NH_3 can penetrate cell membranes and disrupt ionic balance. A previous study has shown that when the NH_4^+/NH_3 concentrations exceeded 800 mg/L, the current density in an MFC system was significantly reduced [129]. In another study, an MFC system performed well with NH_4^+/NH_3 concentrations reaching 750 mg/L [130]. Over time, however, microbes in BES can gradually adapt to 4000 mg/L of NH_4^+/NH_3 [131]. Hydrogen sulfide (H_2S) can bind to the metal ions in enzymes and disrupt cellular metabolism. It has been reported that BES performance declined when the H_2S concentration in the headspace exceeded 3 % [132]. However, high levels of H_2S (up to 1.95 %) may be volatilized into the atmosphere during manure storage, alleviating this impact [133]. Furthermore, anode-associated microbes can oxidize H_2S to SO_4^{2-}, reducing its toxicity within the system [134]. Similarly, high salinity and substrate concentrations may also affect BES performance. It has been reported that 11,000 mg/L of different ions (Cl^-, SO_4^{2-}, and HCO_3^-) did not inhibit BES performance [135]. In fact, such high salt concentrations enhanced the conductivity of bulk solutions, lowered internal resistance, and improved system efficiency. Inhibition begins at NaCl concentrations reaching 33,000 mg/L [136], significantly higher than the salt content in animal manure. While high substrate concentrations may inhibit BES performance [137], numerous studies have successfully used pig manure, dairy manure, and poultry litter to initiate BES [130,138,139]. Humic acids may enhance BES performance due to their role as redox mediators. One study showed that adding 2 g/L of humic acid to an MFC increased the current density by 84 % [140], suggesting that humic acids might positively facilitate the electron transfer in BES.

The concentrations of various substances in animal manure generally fall within the permissible range for BES operation; however, they may still inhibit the startup of BES. Therefore, initially diluting animal manure is an efficient strategy for BES startup, allowing the functional microbes to acclimate gradually in the system. In the BES, the microbial consortium in the bulk solution contributes most of the DOM degradation, while both the bioanode and biocathode may play roles in removing VAs. The conversion of all bioenergy into weak current in MFCs remains an unsolved challenge, as the utilization of this electricity has not been fully addressed. Therefore, integrating BES with AD systems for specific purposes, such as removing VAs, nutrient recovery, and upgrading biogas, presents promising options.

6. Outlook and perspectives

6.1. Possible disposal pathway for VA removal

The removal of VAs should be assessed throughout the entire manure disposal process. Three scenarios can be applied to dispose of animal manure, as depicted in Fig. 6. The black route represents the disposal of liquid manure using wet AD. The advantages of AD include CH_4 production, reduced CO_2 and NH_3 emissions, and odor elimination. The resulting digestate can be separated into liquid and solid fractions, which can then be used as liquid and solid fertilizers, respectively. While the solid fraction is generally safe for land application, composting is often employed to reduce residual DOM and water content further. Alternatively, the liquid manure can be separated into liquid and solid fractions before AD (the red route). However, composting before AD inevitably releases odor, CO_2, and NH_3 into the atmosphere. A key limitation of using wet AD is the disposal of liquid digestate, despite its many benefits over composting. Large-scale farms often face challenges with the limited carrying capacity of adjacent lands and logistical issues associated with transporting liquid digestate to distant locations. Therefore, managers must consider the destination of liquid digestate before establishing large-scale operations. The blue route involves the disposal of solid manure, which can be manually or mechanically collected from animal housing. The solid manure can be directly composted to enhance its fertilization value before land application. This approach has gained increasing attention due to its minimal wastewater generation, as water is primarily used to flush residual feces and urine on barn floors. In this scenario, the resulting wastewater contains low levels of nutrients and can be directed to wastewater treatment plants. Dry AD could be an alternative to composting but requires advanced pre- and post-treatment systems.

Regardless of the disposal route chosen, two types of fertilizers can be produced from animal manure: (1) liquid digestate from wet AD and (2) solid fertilizer from composting or dry AD. However, both fertilizers may still contain residual VAs, as neither AD nor composting can completely remove all VAs from animal manure. The effectiveness of AD and composting in removing VAs can be reduced during periods of high VA usage in animal farms. In such cases, emerging technologies may serve as an emergency solution to ensure the safe management of contaminated manure. Given the high costs of emerging technologies, it is recommended that animal manure with high VA contamination be managed separately. This approach can avoid the need to treat all the manure produced by animals. From another perspective, only manure with severe VA contamination is suitable for these treatments, as high VA concentrations contribute to high removal kinetics. Since 2006, Europe has banned the use of VAs for growth promotion. In the United States, the Food and Drug Administration has encouraged farmers to phase out VA growth promoters [141]. This indicates that manure with high VA contamination is primarily derived from therapeutic applications. Studies have shown that VA concentrations in feed typically range from 2.5 to 50 mg/kg for growth promotion, while therapeutic dosages are much higher [142]. For instance, sulfamethazine used for pigs typically requires 464–773 mg/kg of feed dosages, and the initial doses can be doubled depending on the severity of infection, according to supplier guidelines (Merck Veterinary Manual). Notably, following the phase-out of VAs for growth promotion, therapeutic dosages have slightly increased in some countries [143], further supporting the possibility of separately managing contaminated manure. Since 2020, China has also banned the use of commercial feed

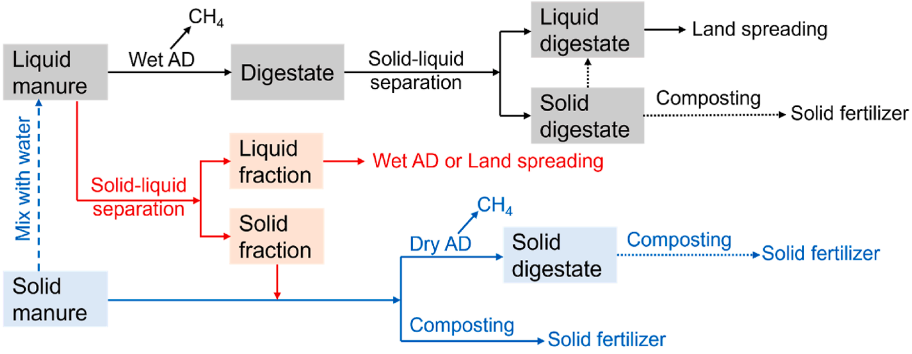

Fig. 6. Technical routes for the disposal of liquid and solid manure.

containing VA growth promoters, as outlined in Announcements No. 194 and 246 [144]. This includes many common VA additives, such as oxytetracycline, nosiheptide, and enramycin. However, medicated feed intended for therapeutic purposes, including chlortetracycline and kitasamycin, as well as prescription medicines, such as various oral and injectable VAs, remain permitted. These treatments are only allowed during disease outbreaks and are subject to strict limitations on treatment duration (typically less than 7 days), facilitating the categorized collection of VA-contaminated manure. Despite these regulations, a complete ban on VA growth promoters remains a significant challenge in most developing countries, as it could lead to substantial economic losses in a short time. Future efforts should focus on progressively implementing legislation to regulate VA use and improve livestock production practices.

Currently, there are no standardized guidelines for managing VA-contaminated manure. However, the following measures can be considered to improve management efficiency: (1) isolating and treating infected individuals in a designated area at the earliest stage; (2) collecting feces into a separate tank during the intensive treatment period; (3) reducing disease by improving farm hygiene, administering probiotics, and utilizing non-antibiotic treatments; and (4) conducting systematic research on the VA degradation to improve farmers' ability to identify high concentrations of VAs in animal manure.

6.2. Concerns about ARB and ARGs

Another issue that warrants further discussion is whether ARB and ARGs can be minimized alongside the removal of VAs. This is very important because it is related to assessing the environmental impact of treated manure. It has been reported that at least 109 distinct ARGs have been detected in animal manure, which may either be hosted within their corresponding ARB or exist independently as DNA-like molecules [145]. While both AD and composting are known to inactivate ARB efficiently, removing ARGs remains more contentious in most studies [146–151]. ARB can be inactivated at high temperatures, such as those found in thermophilic AD and the thermophilic phase of composting. However, the decomposition of DNA-like fragments typically requires temperatures above 70 °C, which is not achievable in biological processes [152]. Many studies report a reduction in ARGs during AD and composting, but others observe no change or even an increase in specific ARGs, such as *sul*1, *sul*2, *tet*A, *tet*X, *tet*W, and *tet*Q [150–152]. These discrepancies have not been fully understood, as the fate of ARGs can be influenced by various factors such as VA concentrations, species, HRT, temperature, and substrate type. Nonetheless, it is evident that the presence of ARB and ARGs is strongly linked to the levels of their parent VAs in the substrate. It is known that VAs above the minimum inhibitory concentration (MIC) can reduce ARB generation and cause a low ARG abundance, while sub-MIC levels of VAs may promote the transfer of ARGs. The differences in VA concentration and degradation rate across these studies may help explain the inconsistencies in ARG removal.

Up to now, no studies have investigated the adsorptive removal of ARB and ARGs from animal manure, despite the likelihood that they may adhere to adsorbents. However, several studies have incorporated biochar and activated carbon into AD and composting, with positive results showing enhanced removal of ARB and ARGs [146,150,153]. ARB and ARGs can also be retained by MF and UF membranes [154–156], while studies on their removal using ED, MD, and FO are limited. Among AOPs, $^{\cdot}OH$ and $SO_4^{\cdot-}$ are particularly effective at removing ARB and ARGs. Studies have shown that ARB can be inactivated entirely and ARGs reduced by 1–4 logs with sufficient $^{\cdot}OH$ supply [157]. Similar results have been obtained with persulfate oxidation [158,159]. However, insufficient oxidant supply may increase bacterial tolerance to VAs and their intermediates [160]. Small organic compounds generated during VA oxidation can serve as a carbon source for bacteria, potentially causing undesired regrowth of pathogens and an increase in ARB and ARGs following oxidation [160,161]. In the BES, long-term cultivation may lead to the development of antibiotic resistance in microbes. However, whether BES systems increase or reduce ARB and ARGs remains unclear, as some studies report an increase while others show a decrease [119,162].

Most emerging technologies have demonstrated significant efficiency in removing ARB and ARGs. However, even if ARGs are significantly reduced in animal manure, there remains a substantial risk of ARGs re-proliferation once they are released into the environment. This risk can be attributed to several reasons: (1) ARGs remain stable in the environment for long periods; (2) horizontal gene transfer facilitates the spread of ARGs; and (3) even if parent VAs are removed, ARGs can still be expressed under other selective pressures, such as heavy metals, non-antibiotic pharmaceuticals, aromatic compounds, estrogen, microplastics, inorganics, and pH changes [163,164]. These compounds can trigger co-selective mechanisms, including co-persistence, cross-persistence, and co-regulation, which further promote the spread of ARGs [165]. One study found that sulfonamides and quinolones in rivers negatively correlated with their corresponding ARGs, while tetracyclines positively correlated with their corresponding ARGs [166]. Similar results have been observed in leachate, sediments, and farmlands irrigated with municipal secondary effluent [167–169], suggesting that the spread of ARGs may extend beyond the selective pressure from parent VAs in these environments. However, numerous studies have demonstrated that applying animal manure to farmlands can significantly increase the abundance of ARGs in soil [164]. This may be due to the high VA concentration in animal manure, which exerts intense selective pressure on ARGs, making the selective pressure from other factors appear insufficient. Therefore, rather than focusing on the difficulties in removing ARGs, reducing the concentration of VAs in animal manure remains a more pragmatic and reliable strategy currently.

6.3. Application challenges for emerging technologies

Emerging technologies can play significant roles in VA removal if the contaminated manure is collected separately. Their key challenges must be thoroughly understood, as summarized in Table S5. Adsorptive removal of VAs is less effective for liquid digestate. Nonetheless, using biochar and minerals for remediation holds promise due to their low cost, enhancing AD and composting performance, and strong immobilization of VAs. Among membrane technologies, pressure-driven separations, MD, and FO raise concerns about the rationale of producing pure water. These technologies cannot separate VAs from humic acids and colloidal particles, meaning their further removal still relies on other methods, such as AOPs. EDR can separate VAs from the feed, with long-term operation demonstrated up to 5000 L/m^2. In this process, certain VAs, which can be easily sorbed by membranes, tend to migrate into the cleaning and product solutions, benefiting subsequent treatment using AOPs. Additionally, the treated manure can be used as a low-salinity fertilizer. While AOPs can efficiently degrade VAs, the complex composition of liquid digestate raises operational costs, restricting their use mainly to situations with high VA contamination. It is important to note that excessive oxidation, which removes humic acids beneficial for plant growth, is undesirable. Therefore, the use of $SO_4^{\cdot-}$ is more widely accepted, as its reaction rate with humic acids is lower than that of ·OH. The carbonization of animal manure is straightforward and is suitable for regions with surplus thermal energy. BES can enhance the performance of wet AD, while only a limited range of VAs have been removable using this approach. Scaling up BES is constrained by challenges such as unstable enrichment of functional microbes and the intensive use of electrodes.

Currently, no laws or standards are available for removing VAs from animal manure, as the correlation between VA contamination and their environmental impact has not been adequately quantified. This knowledge gap stems from the complexity of research, complicating efforts to control VA contamination. A comprehensive understanding of animal manure composition and the degradation of VAs is essential to enhance the research efficiency. Reducing the costs associated with emerging technologies remains challenging in the foreseeable future. Therefore, it is recommended that animal manure with high VA contamination be managed separately, as discussed in Section 6.1, which is currently an economically viable approach for animal farms. Moreover, VA removal can be potentially enhanced through the integration of multiple technologies, including:

(1) Integration of membranes and AOPs. Among membrane technologies, EDR can concentrate VAs into specific solutions, facilitating their destruction through AOPs. This integration can reduce the size of ozonation distributor, the amount of persulfate required, and the necessary electrode area for electro-oxidation. By increasing the voltage and concentration ratio in EDR, VA removal can be enhanced, decreasing the treatment load on AOPs. However, this approach presents a trade-off regarding energy input for EDR and the chemical dosage for AOPs, which warrants further exploration.
(2) Utilization of inherent energy sources. When animal manure containing VAs is managed separately, the biogas energy generated from uncontaminated manure could potentially support the treatment of contaminated manure. In this scenario, pyrolysis and HTC could be considered, as biogas is more efficient in generating thermal energy than electricity. Key trade-offs will focus on the energy required for treating contaminated and uncontaminated manure.
(3) Development of multi-purpose technologies. For example, membrane technologies can recover nutrients and reclaim water, BES can upgrade biogas, and carbonization can produce biochar, thus enhancing their overall economic value. Additionally, many emerging technologies can reduce the concentration of heavy metals and other pollutants in animal manure. When evaluating a technology, its added value should be considered comprehensively.

7. Conclusions

This article reviews the species, concentrations, and distribution of VAs in animal manure and their removal efficiencies using conventional and emerging technologies. The findings indicate that VAs may be present in both the liquid and solid fractions of animal manure with high concentrations, particularly during periods of intensive VA usage in animal farms. AD and composting are insufficient for removing all types of VAs, resulting in unpredictable reductions of VAs in many cases. In such cases, emerging technologies offer potential as supplementary and post-treatment methods to further enhance the removal of VAs. Promising approaches include biochar amendments, EDR, persulfate oxidation, carbonization, and BES. Rapid removal of VAs may contribute to lower ARGs spread risks in the environment. However, the high costs associated with these technologies remain a primary barrier to their practical applications. The authors propose an advanced manure management strategy that separately collects manure during periods of intensive VA usage for additional treatment, which will make the overall cost acceptable. It is further suggested that integrating multiple technologies and utilizing inherent energy can improve the economic viability of emerging technologies.

CRediT authorship contribution statement

Chen Ding: Writing – original draft, Investigation, Formal analysis, Data curation. **Yun Lang:** Investigation. **Liwen Xiao:** Supervision, Resources. **Xinmin Zhan:** Supervision, Resources. **Lin Shi:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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