A critical review of ammonia recovery from anaerobic digestate of organic wastes via stripping

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Abstract

Anaerobic digestion is a well-established technology which valorises organic wastes to produce renewable energy as biogas. The ammonia released during this process can be inhibitory if it exceeds the threshold concentration. Additionally, ammonia is a harmful environmental pollutant and also a renewable source of energy, thus establishing a need for the recovery of ammonia. Gas-liquid stripping is one such widely researched technology to recover ammonia from wastewaters. However, there have not been any reviews focusing solely on the application of stripping to recover ammonia from the anaerobic digestate of organic wastes. This study reviews the current state of research in this field, focusing on the factors affecting the ammonia recovery, stripping column designs and linking of an anaerobic digester to a stripping column. An overview of the work done on the modelling of the ammonia stripping from digestate is also included. This review offers a direction for further research in this field; namely experimental and modelling studies on i) the effect of digestate composition on ammonia recovery and ii) the effect of stripping gas on ammonia removal.

Highlights

- Process conditions and the %CO_2 in the stripping gas influence ammonia recovery
- Stripping columns designs are not a limiting factor determining ammonia removal
- Stripping can alleviate ammonia inhibition in anaerobic digesters
- Activity coefficients are used to estimate ammonia speciation in the digestate

Keywords: stripping; ammonia; anaerobic digestion; digestate; organic waste; modelling; ammonia recovery

Word Count: 9823

List of abbreviations

AD	Anaerobic Digestion
VFA	Volatile Fatty Acids
FA	Free Ammonia
TA	Total Ammonia
FAN	Free Ammonia Nitrogen
TAN	Total Ammonia Nitrogen
G/L	Gas-to-Liquid Ratio
TS	Total Solids
TCOD	Total Chemical Oxygen Demand
COD	Chemical Oxygen Demand
VS	Volatile Solids
$[\boldsymbol{NH_3}]$	FA Concentration (mol/m^3)
$[\boldsymbol{NH_3} + \boldsymbol{NH_4^+}]$	TA Concentration (mol/m^3)
$\boldsymbol{K_d}$	Acid dissociation constant (mol/m^3)
$\boldsymbol{\gamma_{NH_4^+}}$	Activity coefficient of NH_4^+ ion using Pitzer approach
$\boldsymbol{I}$	Ionic strength (mol/m^3)
$\boldsymbol{T}$	Temperature (K)
$\boldsymbol{f^{H^+}}$	Specific activity coefficient of H^+
$\boldsymbol{F}$	Fraction of free ammonia
$\boldsymbol{Q_g}$	Gas flow rate (m^3/s)
$\boldsymbol{V_L}$	Volume of liquid (m^3)
$\boldsymbol{\rho_{NH_3}}$	Density of free ammonia (mol/ m^3)
$\boldsymbol{H_{NH_3}}$	Henry's constant for free ammonia (mol/ m^3)
$\boldsymbol{K_{OL,1}}$	Mass transfer coefficient for free ammonia (m/s)
$\boldsymbol{a}$	Specific volume (m^2/m^3)
$\boldsymbol{V_E}$	Water droplet volume trapped per unit volumetric unit total gas flow
$\boldsymbol{Q_{ST}}$	Volumetric steam flow rate (m^3/s)
$\boldsymbol{N_{OL}}$	Number of overall liquid-phase transfer units
$\boldsymbol{H_{OL}}$	Height of an overall liquid-phase transfer unit
$\boldsymbol{K_{OL,2}}$	Mass transfer coefficient for ammonia on the basis of overall liquid-phase resistance (mol/ m^2.s)
$\boldsymbol{L}$	Liquid loading rate (mol/ m^2.s)
$\boldsymbol{R}$	Universal Gas Constant (J/mol.K)
$\boldsymbol{S}$	Horizontal cross-sectional area (m^2)
$\boldsymbol{l}$	Depth of re-circulation gas inlet under the liquid surface (m)
$\boldsymbol{P_{NH_3}}$	Partial pressure of free ammonia (Pa)
$\boldsymbol{t}$	Stripping time (s)
$\boldsymbol{M_{NH_3}}$	Moles of ammonia stripped (mol)
$\boldsymbol{m_{NH_3}}$	Molar flow rate of ammonia (mol/s)

1. Introduction

The rapid increase in human population since the 20th century has laid pressure upon food and livestock resources, leading to a rapid growth in these industries. A consequence of this growth is the large amount of organic wastes generated annually. For example in 2017, 322 million tonnes of cattle manure were produced in the EU [1],[2]. Organic wastes such as livestock, food, agricultural, etc. must be treated before being discharged into the environment to prevent surface water, ground water and soil pollution [3],[4]. The most common methods of organic waste treatment are (i) physicochemical processes such as extraction and coagulation-flocculation, (ii) thermochemical processes such as combustion and gasification, and (iii) biochemical processes such as fermentation and anaerobic digestion (AD) [5],[6]. Among all these, AD is an attractive technology as it produces biogas, a renewable source of energy for heat or power generation [7].

During the AD process, specific groups of micro-organisms convert organic matter into biogas through four stages in oxygen-free conditions: hydrolysis, acidogenesis, acetogenesis and methanogenesis [1]. In short, during hydrolysis, extracellular enzymes degrade complex organic polymers into soluble monomers. The acidogenic stage entails the conversion of these soluble monomers to volatile fatty acids (VFAs) and hydrogen. In the acetogenesis stage, VFAs are further transformed into acetate, carbon dioxide, and/or hydrogen. Then, during methanogenesis, acetate, carbon dioxide and hydrogen are converted to methane [8]. During hydrolysis, between 33% to 80% of the nitrogenous organic matter such as urea, proteins and amino acids are degraded to ammoniacal nitrogen [9],[10]. This includes dissolved ammonia (NH_3 (gaseous)) in equilibrium with ammonium ions (NH_4^+) [11],[12]. This gaseous ammonia, also called free ammonia (FA), is known to be inhibitory to the AD process as it can permeate through the cell membranes of micro-organisms. This can cause proton imbalance, change in intracellular pH, reduction in efficiency of enzymatic reactions or increase in the energy required for cellular maintenance [11],[13],[14],[15]. Methanogens are the most sensitive micro-organisms to FA at high concentration [16]. At the process level, ammonia inhibition can cause a reduction in steady-state biogas production, and an accumulation of volatile fatty acids (VFAs) in the digester, which can ultimately cause the AD process to collapse [17],[18]. Multiple studies have reported a wide range of inhibitory ammonia concentrations in terms of total ammonia (TA), defined here as the NH_4^+ and NH_3 species together, with concentration units of milligrams of nitrogen in TA per litre (mg TAN/L) [16],[19],[20],[21]. An inhibition threshold of 4000 mg TAN/L was reported for thermophilic AD of cattle manure, 11000 mg TAN/L for thermophilic AD of swine manure, 8000-14000 mg TAN/L for mesophilic AD of chicken manure and 800-1400 mg TAN/L for mesophilic AD of swine slurry mixture [16],[19],[20],[21]. However, the overall negative effect of FA is generally attributed to the differences in operating conditions (temperature and pH), substrate composition, inoculum type, and acclimatization periods [22]. It must be noted that even though numerous studies have determined that FA is the inhibitory species, the concentrations mentioned above are reported in terms of TAN. There is no common consensus among studies about reporting ammonia inhibition in a specific concentration format, and these values are herein reported directly from the studies without alteration throughout this review.

The effluent of the AD process (i.e., digestate) still contains a large fraction of organic matter. Digestate can be applied directly to agricultural fields as a fertilizer, but this practice is no longer allowed by the EU directives due to its negative environmental consequences [23]. FA can volatilise from the digestate and react with compounds in the atmosphere to form ammonium aerosols and particulate matter (such as ammonium nitrate and ammonium sulphate particles), which contribute to atmospheric pollution [24]. FA can also be oxidized in the atmposphere to form HNO_3, causing acid rain. Furthermore, runoff from ammonia-rich waste streams into water bodies can cause eutrophication, causing a decrease in dissolved oxygen levels, which is harmful to aquatic organisms [25].

Various techniques have been studied to recover ammonia from organic waste streams, including gas-liquid stripping, ion exchange, electrodialysis, membrane separation, adsorption, struvite precipitation, and biological processes (nitrification and denitrification) [26],[27],[28],[29],[30],[31]. These studies investigated ammonia recovery from several matrices such as synthetic wastewater, sewage sludge, piggery wastewater and digestate. Ammonia stripping is the process of mass transfer of ammonia from the liquid phase to the gas phase [32]. It is a particularly attractive technology since it is insensitive to solids in the liquid phase and only requires a relatively limited energy input [33]. Baldi et al. [34] described the following steps in the ammonia stripping process: (i) conversion of ammonium ions (NH_4^+) to FA, (ii) diffusion of FA from the bulk liquid phase to the interface between liquid and gaseous phase, (iii) transfer of FA across the interface into the gaseous phase and (iv) diffusion of FA into the bulk of gas phase. The gas used for stripping (also called the stripping agent) containing FA is then finally removed from the system.

The recovered ammonia can be subsequently recovered and valorised to realise its energetic potential. Ammonia can be decomposed to produce hydrogen, which is a clean and renewable fuel. Ammonia is also used (i) directly as a fuel for fuel cells, internal combustion and spark ignition engines, (ii) as a fuel blend in gas fuels and for various other combustion processes, and (iii) as a working fluid in power cycles for devices such as solar cells or refrigeration systems [35],[36] [37],[38],[39]. The application of ammonia for power generation is a broad and widely researched topic, and the interested readers can consult numerous sources of literature on this topic cited above.

This paper will focus on reviewing the application of gas-liquid stripping to remove ammonia specifically from anaerobic digestate of organic wastes. The paper will discuss (i) the factors governing ammonia removal, (ii) the different stripping reactors that have been designed and implemented, (iii) the different configurations of integrating a stripping column in an anaerobic digestion process, and (iv) good practices in the modelling of ammonia stripping.

2. Stripping factors affecting ammonia recovery

The primary factors affecting ammonia recovery during stripping are temperature, pH, and gas flow rate. This section will discuss the effect of these three factors, as well as some secondary factors (such as CO_2 in the stripping gas and the composition of the digestate) on ammonia stripping.

2.1 Temperature

The temperature is an important factor affecting ammonia recovery from the digestate. Ammonium ions and dissolved FA are maintained in equilibrium, as shown in Equation (1) [40].

$$NH_4^+ \leftrightarrow NH_3 + H^+ \tag{1}$$

$$\frac{[NH_3]}{[NH_3+NH_4^+]} = \frac{1}{1+[H^+]/K_d} \tag{2}$$

$$pK_d = 0.09019 + \frac{2729.92}{T} \tag{3}$$

where K_d is the acid dissociation constant related to Equation (1) [[41]]. As the temperature increases, so does the value of K_d (according to Equation (3)). This, in turn, shifts the equilibrium stated in Equation (1) to the right, thereby increasing the amount of FA [42]. Unlike NH_4^+ ions, FA can be physically desorbed from the liquid phase by gas stripping, and therefore an increase in temperature should foster ammonia recovery [32]. An increase in temperature also decreases the solubility of FA in solution [43]. Furthermore, elevated temperatures increase the saturation vapour pressure of the FA in the solution, thereby increasing the driving force for transfer from the liquid phase to the gas phase, as well as increasing the diffusion rates and mass transfer rates of molecules across liquid and gas phases [41],[44],[45]. Experimentally, increasing the liquid phase temperatures (generally in the range of 15-100 °C) was demonstrated to successfully increase ammonia recovery in multiple studies [41],[45],[46]. Zeng et al. [46] reported an increase in ammonia recovery from 86 to 97% as the temperature was increased from 40 to 80°C respectively during steam stripping of the liquid fraction of cattle manure digestate. Zeb et al. [41] reported ammonia recovery of 20%, 40% and 90% at 35, 55 and 70°C, respectively, for air stripping of dairy manure digestate. Provolo et al. [45] demonstrated a higher ammonia removal as the temperature increased during slow-rate ammonia volatilization of animal manure digestate (64% and 87% at 30°C and 40°C, respectively).

2.2 pH

Similar to the effect of temperature, the pH of the liquid phase affects the concentration of FA present in the solution. As the pH of the solution increases, the equilibrium in Equation (1) shifts to the right, due to the consumption of protons by hydroxyl ions. This shift in equilibrium increases the concentration of FA in the solution. The effect of an increase in pH is shown analytically in Equation (2) [44]. This can also be explained by the pKa of the ammonium ion as a weak acid, which is 9.2 [47]. At a pH higher than this pKa, the concentration of FA is greater than the concentration of NH_4^+ ions, which is preferable for ammonia stripping.

Increasing the liquid phase pH (generally in the range of 8 to 12) was demonstrated to increase ammonia recovery in various studies [44],[48],[49],[50], although some researchers did not observe the same trend. Studies reported that increasing pH above a critical value of 10 did not result in a proportionally higher ammonia removal [51], [52], [53]. While these studies did not offer a definite conclusion, a possible explanation can be postulated via the empirical relationship in Equation (2). At a constant temperature of 57.5 °C and at a pH 9, the available FA is 74.5% of TA. At pH 10 and 11, available FA percentages are 96.5% and 99.6% of TAN, respectively. It is, therefore, clear that the FA does not increase significantly with an increase in pH above a critical value of 10, which could also explain why the authors did not observe a proportionally higher ammonia recovery. Another explanation can be drawn using the pKa of ammonium ions (value of 9.2). As the pH of the solution is increased above this pKa, the concentration of FA also increases. However, this increase is not significant above a critical pH value of 10.

2.3 Gas flow rate

An increase in the stripping gas flow rate leads to an increase in ammonia recovery. However, unlike temperature and pH, the gas flow rate does not influence the concentration of FA in the liquid phase. Instead, an increased gas flow rate maintains the concentration gradient of FA between the liquid and gaseous phases, which is the driving force for the mass transfer of ammonia. An increase in gas flow rate also causes a depletion of the boundary layer and thereby a decrease in the mass transfer resistance [54],[55]. Moreover, it causes an increase in the interfacial area, which allows for more diffusion of gaseous ammonia from liquid to gas phase [32], [56]. Conversely, higher flow rates can also lead to decreased liquid temperature, foaming and evaporation of the liquid phase [41].

Some studies have combined the gas flow rate, stripping time, and volume of the substrate into a dimensionless number. This number G/L is defined as the ratio of the total volume of used gas to the volume of the stripped liquid [52],[57],[58]. This is mathematically described in Equation (4),

$$G/L = Q_g . t / V_l \tag{4}$$

Where Q_g is the gas flow rate, t is the stripping time and V_l is the volume of liquid stripped. Working with this dimensionless number enables comparison of different studies in an efficient way. Lei et al. [56] showed that increasing gas flow rates from 3 to 10 L/min over 12 h (G/L ranging from 2160 to 7200) increased the ammonia recovery from 72% to 95% for air stripping of pig manure digestate. Liu et al. [58] demonstrated that increasing G/L from 0 to 3000 yielded an increase in ammonia recovery from 0 to 96% during air stripping of pig manure digestate. Conversely, De la Rubia et al. [59] reported no increase in ammonia recovery when G/L was increased from 585 to 1170 during biogas stripping of food waste digestate. This observation was attributed to low gas flow rates and a low concentration of FA in the digestate, as well as a moderate pH of 8 and a temperature of 35 °C.

2.4 CO_2 in the stripping gas

Digestate typically contains the inorganic carbon species CO_2, HCO_3^- and CO_3^{2-}. These species are in equilibrium with each other as shown in Equations (5) and (6) [41], [60].

$$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+ \tag{5}$$

$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+ \tag{6}$$

If the stripping gas contains CO_2, there is a concentration gradient between the CO_2 in the liquid and gaseous phases. The lower the concentration of CO_2 in the stripping gas, the higher the driving force for mass transfer of CO_2 from the liquid to gas phases and vice versa [61]. Stripping CO_2 from the liquid phase causes a shift in equilibrium towards to the left, decreasing the concentration of protons in the solution, which, in turn, increases the pH (Equations (5) and (6)). This increase in pH improves ammonia recovery, as discussed in Section 2.2 [43],[61],[62]. Stripping CO_2 from wastewater treatment plant effluent to increase the pH of the solution was successfully demonstrated by Cohen et al. [61]. While there have been studies discussing the influence of CO_2 in the stripping gas on ammonia removal from model solutions, not many are related to digestate [43]. Bousek et al. [63] investigated ammonia recovery from a model ammonium bicarbonate solution and organic waste digestate by stripping with a gas mixture containing CO_2 at concentrations ranging between 0 and 40%. For both experiments, the ammonia recovery decreased from 81% to 25% as the concentration of CO_2 in the gas increased from 0% to 40%. A study from Wang et al. [64] on pig manure digestate showed a similar decrease in ammonia removal with inrceasing CO_2 concentration (approximately 95%, 65% and 55% ammonia removal at 10%, 20% and 40% CO_2, respectively).

2.5 Composition of digestate

Digestate is a complex matrix containing organic and inorganic matter as well as soluble and non-soluble components, whose composition varies with the nature of the waste source. Up till now, there have not been systematic studies investigating the effect of the composition, origin or the type of digestate on ammonia recovery. Some studies discussed the effect of individual parameters (such as total solids percentage) with contradictory findings. Georgiou et al. [33] reported similar levels of ammonia recovery when comparing model solutions and digestate, thereby concluding that the composition of the digestate (e.g., dissolved organics, suspended solids, etc.) did not have any effect on the ammonia removal. Conversely, Laureni et al. [65] reported higher ammonia recovery for substrates with lower organic matter (greater than 90% for concentrations below 10 g COD/L as compared to 50% for concentrations below 27 g COD/L). A possible explanation was that the FA available to be stripped was lower at higher organic-containing substrates due to the tendency of ammonium ions to adhere to organic matter.

Theoretically, the total solids (TS) in the digestate can also affect the ammonia removal efficiency. Conventional digestate typically has a TS content of 1 to 5% on mass basis. Lower percentages of total solids allow more FA to be trapped by the stripping gas, due to better gas-liquid association and fluidity [42],[44],[66]. Since ammonium ions can be trapped or adsorbed onto suspended organic matter, stripping of the liquid fraction of the digestate after solid-liquid separation should theoretically yield higher ammonia recovery [45],[48]. However, both Huang et al. [67] and Abouelenien et al. [68] demonstrated a maximum ammonia removal efficiency of 95% and 85.5%, respectively, in spite of the high solids TS content of the AD digestate (i.e., 20% TS) and the absence of solid-liquid separation. Provolo et al. [45] also concluded that for slow-rate ammonia volatilisation, the TS content in the digestate did not have any effect on ammonia removal. It can be concluded that the above studies show no consensus and do not provide enough information about the influence of the digestate composition on ammonia recovery. A comprehensive study experimentally determining ammonia recovery over a wide range of values for different parameters such as TS, organic content and different types of digestate could provide this information.

3. Stripping column design

The design of the stripping column is important as it defines the intensity of contact between the gaseous and liquid phases. More intensified contact between the phases allows a higher mass transfer, at minimum energy costs [34]. This section will describe the different stripping column designs reported in the literature. Figure 1 schematically depicts the different types of stripping columns.

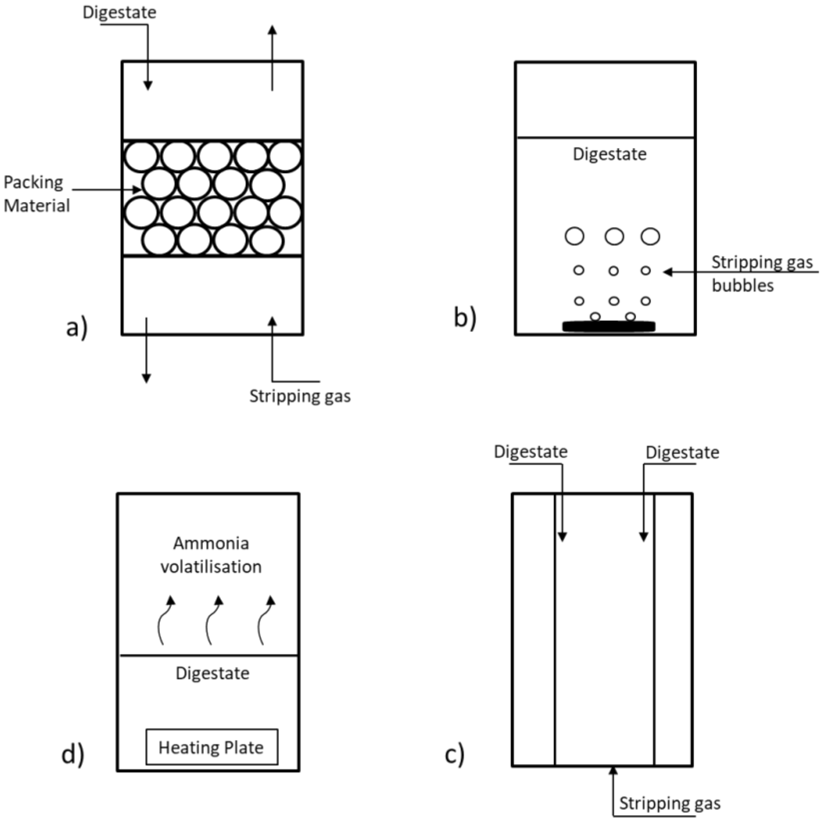

Figure 1: Schematic representation of different stripping column designs. Clockwise from top left: a) Packed bed column, b) Bubble aeration column, c) Wet-walled column and d) Thermal stripping

3.1 Packed Columns

Packed columns have been extensively researched for a variety of applications and have also been widely used for stripping ammonia from digestate. A packed column is a hollow column filled with packing materials (Raschig rings, plastic rings, etc.) to maximise the surface area for mass transfer [69]. A potential disadvantage of using digestate in packed columns is that the suspended particulate matter in the digestate can cause fouling on the packing material which, in turn, decreases the effective area available for mass transfer [70]. Ammonia recovery of up to 80% was demonstrated when dairy manure digestate was stripped in a packed column [53]. The same study also reported a decrease in TS between the influent and effluent across the packed column, indicating the deposition of solid particles on the packing material. The authors recommended an upgrade in the design of columns and trays for future experiments to avoid fouling. Laureni et al. [65] used a packed column to strip pig slurry digestate and demonstrated up to 95% ammonia recovery without discussing any fouling or scaling problems. Some studies have reported stripping from the liquid fraction of the digestate to eliminate any problems associated with solid particles. Guštin and Marinšek-Logar stripped the liquid fraction of pig slurry and other organic digestate in a bench-scale packed column, containing plastic carriers as packing material and reported 92% ammonia recovery [52]. Zeng et al. [46] observed a maximum ammonia recovery of 96% when steam-stripping applied to the liquid fraction of cattle manure digestate. A packed column with plastic rings was also used to strip the liquid fraction of slaughterhouse waste digestate, achieving a 90% ammonia recovery [71]. The above studies show that the design of the stripping column itself is not a limiting factor to ammonia recovery, as high ammonia recoveries of up to 95% were reported. Risk of clogging and fouling of the packing material can be considered as a drawback for commercial application of this technology. As discussed, stripping with the liquid fraction of digestate is a way to mitigate the negative effect of the digestate's solids content. However, this solid-liquid separation requires an additional process step, and therefore its implementation in future studies depends upon its feasibility. Another method to overcome the problems of clogging and fouling is a 'cleaning step' to regenerate the column. One such example is described in a patent by Schrader et al [72], where gas (air or N_2) is bubbled through the column to remove the clogged debris. A prerequisite is that the specific gravity of the packing material in the column must be less than that of the flooding gas. This allows for the formation of a fluidised bed, allowing sufficient cleaning. The authors recommend cleaning once a week for two hours, although that might vary, depending upon the nature of the clogged material.

3.2 Bubble aeration

Bubble aeration is another commonly used stripping technology in which gas bubbles are introduced into the liquid phase by an air stone, gas diffuser or a submerged pipe to transfer the FA from the liquid phase to the gas bubbles. These gas bubbles containing ammonia are then collected at the outlet. While aeration is an easily and widely implemented technology, foaming in digestate and subsequent volume loss, water evaporation and cooling of the digestate are some disadvantages of this technology [49]. Ammonia stripping by aeration has been tested in multiple studies [41],[56], [59], [73], [74]. Some studies have used air diffusers to strip 95% ammonia from the supernatant of settled pig waste digestate and pig waste digestate, respectively [41], [56]. De la Rubia et al. [59] reported a maximum ammonia recovery of 44% using bubble aeration with synthetic biogas. Garcia-Gonzalez et al. [73] reported a maximum ammonia recovery of 99% from raw swine manure by bubbling air through an air stone. A study by Park and Kim [74] also demonstrated 95% ammonia recovery using aeration through an air stone for ammonia stripping from waste activated digestate in which pH was increased by a water electrolyser system.

3.3 Miscellaneous designs

In addition to packed columns and bubble aeration, there have also been novel stripping configurations that have been implemented to strip ammonia from digestate. A water-sparged aerocyclone is one such example. In this system, the ammonia-containing liquid phase is sparged into a reactor through a porous wall creating fine droplets, while the stripping gas flows tangentially through the column. This configuration greatly increases the contact area between the liquid and gaseous phases, leading to increased mass transfer. Ammonia removal greater than 91% was reported when stripping with the liquid fraction of anaerobically digested piggery wastewater at pH 11 [70], [75]. Limoli et al. [66] investigated the application of turbulent mixing to strip out ammonia from a mixture of cow, pig and poultry manure digestate. The turbulent mixing was created by a vortex at the water surface in a vessel, using a magnetic stirrer. The authors reported a maximum ammonia recovery of almost 97%, at a pH of 12. Thermal stripping is a design that does not utilize any stripping gas flow [76]. In this system, the digestate is heated to very high temperatures (90 °C and above), which volatilises the FA. One study thermally stripped the liquid fraction of dairy manure digestate by heating the filtrate of dairy manure digestate in a lab vessel at different temperatures (between 76 °C and boiling temperature). Ammonia removal greater than 95% was observed at 102 °C and a pH in the range of 9 to 11. It was observed that when boiling temperatures were reached, the ammonia recovery and mass transfer coefficients were much higher than in the sub-boiling temperature regimes (the mass transfer coefficient was 10 to 100 times higher) [76]. This can be attributed to turbulence in the liquid phase due to boiling, which decreases mass transfer resistance. A very recent study by Cao et al. [77] used an innovative spraying technology to strip out ammonia from piggery wastewater. Wastewater was dispersed as droplets, which were then allowed to come into contact with spiral hot water pipes. The formation of droplets increased the surface area for mass transfer, and the collision force and temperature difference released FA due to the breaking of the phase equilibrium. The study reported almost 95% ammonia removal at pH of 8.06 and water re-circulation at a temperature of 55 °C. Bonmati and Flotats [48] used an isothermal wet-walled column, where the walls of the column provided the contact area for mass transfer between liquid and gaseous phases. The authors demonstrated up to 98% ammonia recovery at a pH of 11.5 and temperature 80 °C. It can be concluded that the extent of ammonia removal is not limited by the choice of stripping column design, and is instead dictated by process conditions and the choice of stripping gas. The choice of the stripping column design can therefore be influenced by process or experimental constrainsts such as the presence of suspended solids in the digestate, materials and manufacture costs or availability of equipment. For example, aeration can be preferred over a packed column if it is not feasible to carry out solid-liquid separation. Table 1 summarises the advantages and disadvantages of each stripping column design.

Table 1: Advantages and disadvantages of different stripping column designs

Stripping Column Design	Properties	Advantages	Disadvantages	Reference
Packed Column	Liquid and gas phases circulated in co- or counter-current manner	Extensively researched and implemented	Suspended particles in digestate may cause fouling in the column	[46], [52],[53], [65], [71]

Bubble Aeration	Liquid phase is stationary, while stripping gas is bubbled through	Easily implemented, and widely used	-Foaming of digestate -Cooling of digestate	[41], [49], [56], [59],[73],[74]
Water-sparged aerocyclone	Bubbles created by sparging, stripping gas flows tangentially	- Higher mass transfer coefficients -Low cost and easy operation	Upscaling difficult to implement	[70],[75]
Thermal stripping	Stationary liquid phase is heated in the absence of a stripping gas	No external stripping agent required	Possibly high energy requirements due to high temperatures	[76]
Spraying	Liquid phase is sprayed as droplets; External stripping gas is not required	Lower energy costs as water heated in re-circulation pipes instead of wastewater	-	[77]

3.4 Full scale ammonia stripping

While the studies discussed above were all implemented at a laboratory scale, stripping of ammonia from digestate has also been implemented at pilot and industrial-scale plants, with reportedly 9-10 large scale air-ammonia stripping plants in the USA and 15 in Germany [78]. In the literature, packed bed and aeration stripping columns are the designs that have been most commonly implemented at large scale. Baldi et al [34] operated a packed tower and a bubble column at full scale, in continuous and batch-wise modes to strip ammonia from the liquid fraction of livestock manure and corn silage digestate. The liquid-phase capacity of the plants was in the range of 5 to 12 m^3, and the range of gas flow was between 500 and 4500 Nm^3/h. A packed stripping column was used at a large scale to strip ammonia from the urine and anaerobic digester sludge liquid [79]. Katehis et al. [51] also reported using a packed column to strip ammonia from the centrate of digestate.

4. Anaerobic digester- Stripping column configurations

This section will describe how ammonia stripping in an anaerobic digester can mitigate ammonia inhibition. A stripping column can be integrated in the digestion process via four different configurations [44]: (i) pre-AD stripping, (ii) in-situ stripping, (iii) side-stream stripping and (iv) post-AD stripping (Figure 2).

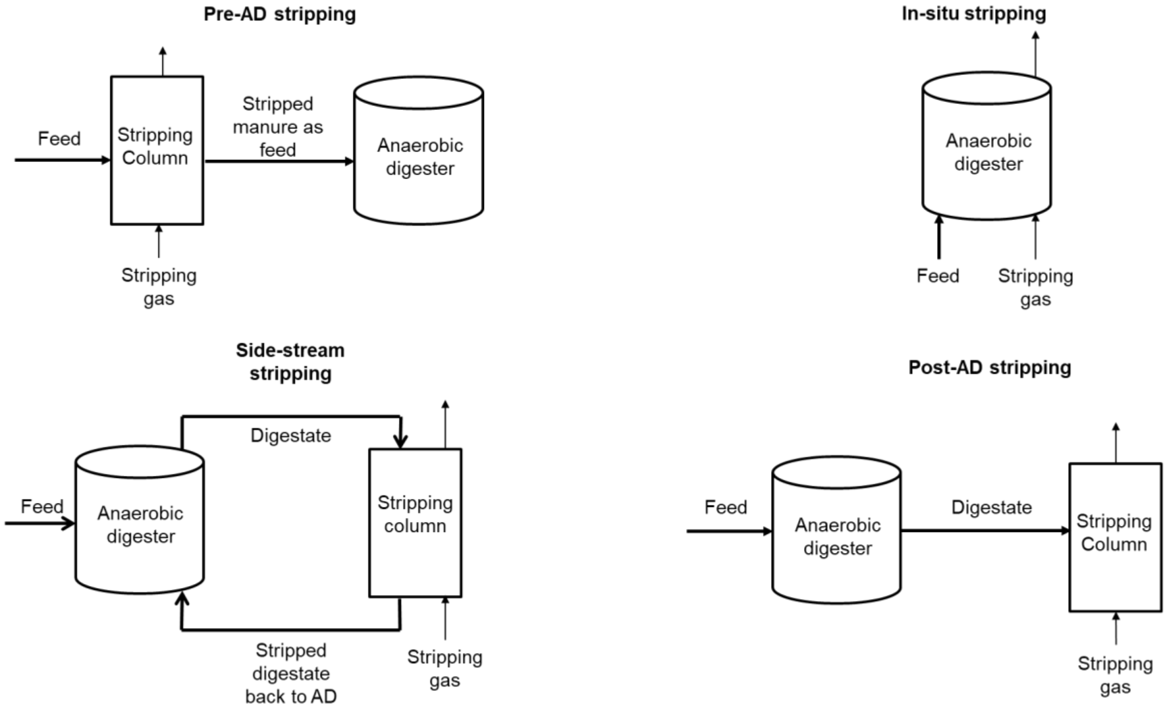

Figure 2: Anaerobic digester- Stripping column configurations [44]

4.1 Pre-AD stripping

In this configuration, ammonia is stripped from raw organic waste substrates before they are fed to the anaerobic digester. Stripping ammonia from raw substrate should decrease the amount of ammonia entering the anaerobic digester, thereby preventing process inhibition. Multiple studies have implemented pre-AD stripping and reported higher methane yields [40],[48],[80],[74],[81]. For example, Zhang et al. [40] demonstrated that during semi-continuous biomethane potential tests, pre-AD stripping of raw piggery wastewater improved the methane production potential by a factor three compared to non-stripped wastewater (170 and 49 mL CH_4/$gCOD_{added}$, respectively, where COD stands for Chemical Oxygen Demand). The same study also reported that pre-AD stripping increased the removal of organic matter in terms of total chemical oxygen demand (TCOD), soluble COD, and VFA (45, 50, and 57% as compared to 20, 13, and 10%, respectively). Another study also reported that stripping of raw piggery wastewater before AD increased the methane yield by a maximum of 5 times (50 compared to 250 mL CH_4/g COD_{added}) as compared to no stripping [80]. Furthermore, pre- AD stripping of ammonia from waste activated sludge yielded a 2.9 times higher specific methane yields (667 compared to 234 mL CH_4/g VS_{added}) and a 1.8 times higher substrate degradation as compared to the control digester without stripping [74]. The higher methane yields and higher organic removal reported in these studies can be attributed to the lower ammonia concentration in the digester with pre-AD stripping (as opposed to without). Stripping of ammonia most probably prevents its accumulation beyond inhibitory concentrations, thereby inhibiting disruption of methanogenesis.

Conversely, some studies did not report enhanced methane yields due to ammonia stripping. Bonmati and Flotats [48], for example, observed a decrease in methane production of 75% after pre-stripping of pig waste slurry (an average of 22 compared to 88.8 L CH_4/ kg VS). Possible explanations for this reduction in methane production were the presence of metal cations (Ca, Zn and Cu) that were introduced during the alkaline treatment before stripping and high values of pH before AD. Li et al. [82] also reported that stripping ammonia from the raw chicken manure before it was fed to a digester did not improve the biomethane production potential. This was explained by the reduction of total COD due to the addition of $Ca(OH)_2$ (as an alkaline agent to increase the pH), thereby decreasing the organic matter in the feed for anaerobic digestion.

In conclusion, pre-AD stripping has been demonstrated to be an effective technology to address the issue of ammonia inhibition. It can be especially useful for treating nitrogen-rich substrates such as pig and chicken manure, thus recovering ammonia before AD and preventing its accumulation. However, the alkaline agents used to increase the pH before stripping can be a limiting factor, as reported by Bonmati and Flotats [48] and Li et al. [82]. The use of NaOH and KOH can cause cation inhibition of the micro-organisms, affecting the methane yield, while the use of $Ca(OH)_2$ can precipitate active micro-organisms out of the digester. The use of alternative alkaline agents (as discussed in Section 2.5) could overcome this problem. Furthermore, it must also be pointed out that the pre-AD configuration does not allow maximum ammonia recovery, as the ammonia produced as a result of the AD degradation process itself cannot be recovered. Therefore, this configuration could be an effective technique to improve AD performance, but not necessarily the best for ammonia recovery. Lastly, most studies were conducted in batch or semi-continuous mode, so there is no information on whether it is equally effective for continuous processes.

4.2 In-situ stripping

In the in-situ configuration, the stripping and AD process take place in the same reactor. This configuration is advantageous as it prevents the accumulation of ammonia in the reactor and hence prevents inhibition of the methanogenic micro-organisms [44], [59]. Moreover, additional equipment or post-treatment steps are not required, which makes it potentially cost-effective. A disadvantage of in-situ stripping is that the physical conditions for the stripping and anaerobic digestion cannot be changed independently from each other. While high temperatures and pH are beneficial for ammonia stripping, they are not suitable for the AD process, due to both FA and temperature inhibition of the micro-organisms [15].

Abouelenien et al. [83] demonstrated the feasibility of in-situ stripping as a proof of concept, with ammonia stripping and chicken manure digestion carried out simultaneously in the same reactor. However, this study did not make a comparison with a control reactor and hence, the actual advantage of using in-situ stripping to improve the AD process was not proven. In another study, in-situ stripping was implemented in an anaerobic digester fed with chicken manure with high TS content (15%) at mesophilic temperature [84]. While ammonia recoveries were not reported directly, TAN concentrations in the stripping digester were 14.7% lower than in the control digester (i.e., 5.8 g N/L compared to 6.8 g N/L) after in-situ stripping. Methane yields were 34% higher compared to the control digester (an average of 250 compared to 187 mL/g-VS_{feed}). The digester with in-situ stripping also showed a 30% reduction in VFA concentration compared to the control (an average of 15.2

compared to 21.6 g/L). The authors concluded that lower TAN concentrations in the stripping digester prevented the inhibition of micro-organisms, which subsequently resulted in higher methane yields as compared to the digester without stripping. This could also be corroborated by the lower VFA concentrations in the stripping digester. Furthermore, the authors also investigated whether ammonia stripping caused any changes in the microbial community. They reported that, while the dominant pathway was syntrophic acetate oxidation coupled with hydrogenotrophic methanogenesis (SAO-HM), in-situ stripping caused a partial shift towards acetoclastic methanogenesis. This was also attributed to the lower TAN concentrations in the stripping digester.

Both Sun et al. [85] and Jiang et al. [86] compared two different reactor configurations of in-situ stripping; biogas circulation through (i) the liquid phase of the reactor and (ii) the headspace of the anaerobic digester. While the study did not report ammonia removal in terms of efficiency, it compared the performances of the digesters. In both digesters, the steady state methane yield increased as the organic loading rate (OLR) was increased. At an OLR of 4 g volatile TS/L/d, the digester with biogas re-circulation through the liquid phase demonstrated a steady state methane production of 2900 mL/L/d. Conversely, the reactor with re-circulation through the headspace was not able to achieve steady-state methane production. This difference was attributed to lower TAN concentrations and correspondingly higher VFA accumulation in the first digester as compared to the second (with an average of 2050 mg/L compared to 2850 mg/L). Gas re-circulation through the liquid phase of the digester showed twice the ammonia removal as compared to re-circulation through the headspace, resulting in lower ammonia concentration in the liquid phase and a more stable anaerobic digestion process.

4.3 Side-stream stripping

In the side-stream stripping configuration, a part of the digestate in the anaerobic digester is stripped in a separate stripping column and then recycled to the digester. An advantage of this configuration is that the stripping column and anaerobic digester can be operated independently in terms of temperature and pH.

Several studies have reported a decrease in ammonia concentration in the digester when side-stream stripping was implemented [14],[87],[88],[89]. Serna-Maza et al. [87] reported that introduction of side-stream stripping led to a decrease in TAN concentration (5 gN/kg to 3 gN/kg). In studies by Pedizzi et al. [14] and Fernandez-Gonzalez et al. [88], a decrease in TAN concentration from 2.5 to 1.1 g N/L in the digester and by a factor of 1/3, respectively, was observed upon re-circulation of stripped digestate. Inspite of the decreasing ammonia concentration in the stripping digesters, no significant change in methane yields was reported. This is in contrast to the observations reported during pre-AD and in-situ stripping. This could possibly be explained by the fact that conditions of high temperature and pH during the stripping process (and, in the case of Pedizzi's study, the presence of air) have been known to negatively affect microbial communities, thereby possibly de-activating methanogens. Further, upon re-circulation, the now low methanogenic activity of the stripped digestate could be positively compensated by the micro-organisms in the digester, thus explaining the lack of change in methane yields.

All the above studies reported a change in the methanogenic communities in the digester upon the application of side-stream stripping. Possible causes for this shift could be the decrease in TAN and VFA concentration which is known to promote the acetoclastic methanogenesis pathway, or the exposure of digestate to air during re-circulation promoting facultative anaerobes [82], [90]. Li et al. [82] operated three side-stream stripping columns with different stripping conditions and reported an increase in relative abundance of more robust hydrogenotrophic methanogens in the digesters in which harsher stripping conditions were applied. Conversely, Serna-Maza [89] reported that side stream stripping caused a change in the dominant pathway from syntrophic acetate oxidation to acetoclastic methanogenesis, even though the same stripping conditions were applied. The difference in shifts could possibly be explained by the higher OLR (9 g VS/L/d compared to 2 g VS/L/d) and average VFA concentrations (12 g/L compared to 250 mg/L) in [82] as compared to [89]. While the large difference in operating conditions does not allow for the most accurate comparison, it does show how side-stream stripping can cause a change in the microbial community composition inside the digester.

4.4 Post AD stripping

In the post-AD stripping configuration, ammonia stripping is implemented on the digester effluent as a post-treatment step. This configuration has some practical advantages: (i) the stripping conditions can be optimized regardless of the process conditions needed for stable AD, (ii) the ammonia-stripped digestate could be used as fertilizer, and (iii) stripping at high temperatures could also work as a post-treatment pasteurisation step [44]. While post AD stripping is a suitable configuration to recover ammonia from the system, it does not decrease ammonia concentrations in the anaerobic digester, and thus it is not a solution to prevent ammonia inhibition. This could be one reason for the low number of studies that have investigated post AD stripping. As an example, Wang et al. [64] stripped ammonia from the liquid fraction of anaerobic digestate of pre-treated wheat straw and the stripped digestate was subsequently used as fertilizer.

4.5 Overview of AD stripping

Table 2 is a compilation of the different publications studying ammonia recovery from digestate via stripping. This table lists the anaerobic digester-stripping column configuration, the inlet ammonia concentration (mg NH_3 /L, unless specified), the maximum ammonia recovery reported by the study (in %), the process conditions (pH temperature and G/L) at which the maximum ammonia recovery was observed, the stripping gas used, the AD organic waste substrate, and any additional remarks from the study.

In addition to the discussions in sections 4.1 to 4.4 , a few interesting trends can be observed in the data presented in Table 2. Most of the studies reported in Table 2 investigate the stripping process in an independent stripping column. This is probably because characterisation of the stripping process is the first step to be studied before integration with the AD process. Further, it is relatively less expensive and time-intensive to study the stripping process without linking it to the AD process. It can also be observed that air is the most commonly used stripping gas in the studies with an independent stripping column and pre-AD studies. This is because air is readily available, inexpensive, and there is no direct contact of air with the AD process, thereby preserving anaerobic conditions. For the same reason, for in-situ and side-stream stripping studies nitrogen and/or biogas are the preferred stripping gases (with the exception of one study) to maintain anaerobic conditions inside the digester.

It is difficult to compare the ammonia recovery reported by different studies in Table 2 directly. This is because different studies carry out the stripping process over a wide range of process conditions values (temperature, pH or G/L). The value of these process conditions determines the FA and the driving force for mass transfer of ammonia, and therefore the wide variance in values makes comparing ammonia recoveries difficult. A possible solution to overcome this problem is to theoretically calculate the FA available at those process conditions for each study. This could be one basis for comparison, provided the G/L ratio is similar. These calculations have not been shown in this study as they are out of its scope.

Table 2: A compiled list of publications studying ammonia stripping from digestate. The configuration 'Stripping column' refers to the publications in which ammonia stripping from digestate has been reported without any preceding or succeeding AD process. The term 'n.d.' stands for not defined. Biogas compostion is defined as 60% CH_4-40% CO_2 unless specified.

Configuration	Inlet ammonia concentration (mg NH_3 /L, unless specified)	Max Ammonia removal (%)	Process Conditions	Stripping Gas	Waste Source	Additional Remarks	References
Stripping Column	7170	44	pH = 8.3 T= 70 ℃ G/L = 840	Synthetic biogas (65% CH_4-35% CO_2)	Food waste		[59]
Stripping Column	821	80	pH = 10 T= 34 ℃ G/L = n.d.	Air	Dairy wastewater		[53]
Stripping Column	4.35 g N/kg	92.5	pH=9.95, T= 55 ℃ G/L = 5730	Biogas	Source Separated Domestic food waste		[91]
Stripping column	6 g NH_4^+-N/L	96	pH= 7.7 T= 65 ℃ G/L =1200	Air	A mixture of pig manure, fodder, sugar	Increase in %CO_2 in the stripping gas decreased ammonia recovery	[63]
Stripping column	2200	92	pH=10 T = 70 ℃ G/L =1800	Air	Pig slurry and other organics		[52]
Stripping Column	1544	95.3	pH=12 T=15 ℃ G/L =36000	Air	A mixture of pig excreta and kitchen garbage		[56]
Stripping Column	2.5-3.5 gN/L	79	pH=9.4–10.1 T=52 ℃ G/L =200	Air	A mixture of livestock manure and corn silage	- Ammonia stripping carried out at full-scale - Investigation of ammonia recovery at the semi-continuous and continuous operation of stripping column	[34]

Stripping column	120	91	pH >11 T = (28–30 ◦C) G/L =n.d.	Air	Piggery wastewater	Implementation of a water-sparged aerocyclone as a novel stripping column design	[75]
Stripping column	5000	97.1 +/- 2.6	pH=12 T= 22 ℃ G/L =n/a (Velocity gradient = 300 min^{-1})	Air	A mixture of raw cow, pig and poultry manure	- Turbulent mixing was implemented as a means of stripping - CaO, NaOH and H_2O_2 were compared as a means to control pH. NaOH was shown to be the most suitable	[66]
Stripping column	1440	95	pH= 8 T = 70 ℃ G/L =n.d.	Air	Dairy manure	Air stripping was concluded to be an effective way of increasing pH without the addition of alkali	[41]
Stripping column	1000 mg N/L	85	pH=12 T= not specified G/L =n.d.	Air	Landfill leachate	Economic analysis showed that at high ammonia recovery, ammonia stripping is more feasible than struvite precipitation and membrane treatment	[92]
Stripping column	615-775	85	pH=7.8 T= 67-70 ℃ G/L = 1133	Air	Sewage sludge	Multiple scrubbing agents were investigated for most efficient ammonia recovery	[57]
Stripping column	1050 mg N/L	73	pH=9.5 T= 60 ℃ G/L = 700-750	Air	Human urine and sewage sludge mixture (Full scale)		[79]
Stripping column	414 mg N/L	53	pH=11 T= 40 ℃ G/L =2429	Air	Dairy manure	Economic analysis of ammonia stripping and recovery at a commercial dairy farm	[50]
Stripping column	910	96	pH=11 T= 80 ℃ G/L = 71-80	Steam	Cattle manure		[46]
Stripping column	1000	98	pH > 11 T= 70 ℃ G/L =850-900	Air	Biosolids		[51]

Stripping column	1.84 g/kg	87	pH = 9 T= 40 ℃ G/L =n/a	Air	Animal manure	Demonstration of slow rate ammonia stripping; the air was pumped across the headspace of the reactor, and not through the liquid phase directly	[45]
Stripping column	1652 mg $N\text{-}NH_3$/L	95	pH = 12 T= 50 ℃ G/L =2000	Air	A mixture of cattle manure (95% cow manure)	- Several types of lime were investigated as an alkali to increase the pH of digestate before stripping. - Hydrated lime showed the fastest reaction rates, along with being the most cost-effective.	[33]
Stripping Column	22 g/kg VS	96	pH = 11 T= 55 ℃ G/L =240	Air	Swine excreta	Demostration of ammonia stripping from solid state AD digestate (~20% TS content)	[67]
Stripping column	3013 mg N/L	80	pH = 8.17 T= 50 ℃ G/L =2250	Air	Swine manure	Ammonia recovery greater than 80% for substrates with organic matter < 10 g COD/L	[65]
Stripping column	250	95	pH = 9.5 T= 40 ℃ G/L =1440	Air	Swine Wastewater	Ammonia recovery by stripping and struvite precipitation	[93]
Stripping column	~635	95	pH = 8.06 T= 55 ℃ G/L =n/a	Air	Piggery Wastewater (raw manure)	Ammonia stripping via an innovative spraying technology	[77]
Stripping Column	1449 mg N/L	96	pH = no info T= 36 ℃ G/L =3000	Air	Pig manure		[58]
Stripping Column	3173	68	pH = 12 T= 20 ℃ G/L = n.d.	Air	Mixture of cow manure and grass		[94]
Stripping column	800 mg N/L	>95	pH=9-11 T= 102 ℃ G/L =n/a	No gas (thermal stripping)	Dairy manure	Ammonia recovery by thermal stripping at liquid boiling point	[76]
Pre-AD stripping	4950	88	pH=11 T= 37 ℃ G/L =2700	Air	Piggery WW		[40]

Pre-AD stripping	7600 mg N/ Kg	97	pH = 11 T= 85 °C G/L =1440	Nitrogen	Waste activated sludge	Ammonia stripping before AD was beneficial for increased methane production	[81]
Pre-AD stripping	3.5 gN/kg	98	pH= 11.5 T= 80 °C G/L =75	Air	Pig slurry		[48]
Pre-AD stripping	4.95 g N/L	71	pH = 10 T= 37 °C G/L =1440	Air	Piggery Wastewater (raw manure)	- Evidence of the type of alkali and dosage used in stripping can affect methanogenesis. - Na$^+$ and K$^+$ ions can have a negative impact on AD due to cation toxicity. - Ca^{2+} ions have a lower negative impact due to lower solubility	[80]
Pre-AD stripping	9230	95	pH = 9.5 T= 35 °C G/L =21600	Air	Waste activated sludge from	Electrolysed water system is used to increase the pH before stripping	[74]
In-situ stripping	1.25 g N/L	64	pH=8.1 T= 55 °C G/L =200	Nitrogen	Dairy manure		[42]
In-situ stripping	369 mg N/L	95	pH = 10 T= 35 °C G/L =7200	Nitrogen	Waste-Activated Sludge	Improved short-chain fatty acids production during alkaline fermentation	[95]
In-situ stripping	6800	14.7	pH = 7.8 T= 37 °C G/L =2328	Biogas	Chicken manure	Anaerobic digester with in-situ stripping showed greater biogas yields and lower VFA than the control	[84]
In-situ stripping	194 mg N/L	n.d.	pH = no info T= 53 °C G/L = n.d	Biogas	Distillery WW by ethanol production from kitchen waste	Higher ammonia recovery when biogas was bubbled through a liquid phase than flushing the headspace of the reactor	[86]
Side-stream stripping	3500	72	pH=9.5 T=80 °C G/L = n.d. (450 bar vacuum)	Steam (Vacuum pump)	Chicken Manure		[96]
Side-stream stripping	5.1 g N/kg	32%/day	pH=10, T= 85 °C	Biogas	SS-DFW		[87]

Side-stream stripping	8.13 g N/L	98	pH=10.25 T= 70 ℃ G/L =830	Biogas	SS-DFW		[44]
Side-stream stripping	2100	45	pH=10 T=70 ℃ G/L =432	Nitrogen	Chicken manure (Digestate)		[82]
Side-stream stripping	N.d.	90	pH = 8.5-9 T= 65 ℃ G/L = n.d.	Air	Slaughterhouse waste		[71]
Side-stream stripping	2500	94	pH = 9.5 T= 35 ℃ G/L =60	Air	Poultry manure	Decreased ammonia concentrations and increased biogas yield, COD removal, and OLR.	[89]
Side-stream stripping	2.4 g N/L	50	pH = n.d. T= 65 ℃ G/L = 180	Air	Pig manure and Ecofit		[14]
Post-AD stripping	1750	97	pH= n.d. T= 67-70 ℃ G/L = n.d.	90% CH_4-10% CO_2	Pre-treated wheat straw		[64]
Pre-and-post AD stripping	10.5 gN/Kg TS	85.5	pH = 10 T= 85 ℃ G/L = n.d.	Nitrogen	Chicken manure		[68]
Pre and Post AD stripping	17.3 g/kg VS	90	pH = 10.2 T= 55 ℃ G/L =360	Air	Swine manure		[97]

5. Modelling

The modelling of the ammonia stripping process from digestate has been specifically studied in a few papers [78], [98]-[99]. These models can be described by the governing equations of mass transfer and the composition of the digestate. Digestate is a complex matrix, containing organic compounds and inorganic ions (e.g., VFAs, CO_3^{2-}, K^+ etc.) along with multiple species in equilibrium, which can affect the speciation of ammonia, Equation (1). Therefore, the composition of the digestate must be appropriately defined to construct sensitive models. This section will describe (i) the mass transfer equations used in various studies, (ii) the methods that describe the composition of the digestate, and (iii) the studies that have described modelling of ammonia stripping from digestate.

5.1 Mass transfer equations

Many studies have formulated analytical equations to describe the mass transfer of ammonia via stripping, some examples of which are shown in Table 3. While these equations have been described for stripping of ammonia from a variety of wastewaters, these can also be applied to digestate, as the physical process of ammonia stripping is the same [98].

Table 3: Final mass transfer equations for models describing ammonia stripping

Description	The final form of the equation	Remarks	Reference
Desorption of ammonia from the liquid phase by aeration	$\ln\frac{[NH_3+NH_4^+]_{t_2}}{[NH_3+NH_4^+]_{t_1}} = \frac{Q_g \cdot \rho_a}{V_l \,.\, H_{NH_3}} \,.\, F.\,(t_2 - t_1)$		[32]
Mass transfer of ammonia into rising gas bubbles	$[NH_3](t) = [NH_3]_{t=0} \cdot \exp\,[-K_{OL,1}a \cdot t]$	Assuming exit gas is far from saturation	[100], [101]
Ammonia desorption into the air stream	$[NH_3](t) = [NH_3]_{t=0} \cdot \exp\,[-V_E(Q_A + Q_{ST}) \cdot t/V_L]$	Takes ammonia loss due to entrainment in water vapour into account	[101]
Ammonia stripping in a packed column	$Z = N_{OL}\ X\ H_{OL}$ $N_{OL} = \int_{[NH_3]_{in}}^{[NH_3]_{out}} \frac{d[NH_3]}{[NH_3]^* - [NH_3]}$	Validated experimentally	[51]

	$H_{OL} = \frac{L}{K_{OL,2}\alpha}$		
Flux of ammonia into gas bubbles	$K_{OL}a =$ $-\frac{Q_g}{R \cdot T \cdot S \cdot l \cdot H_{NH_3}} \cdot \ln\left(\frac{[NH_3] - P_{NH_3} \cdot H_{NH_3}}{[NH_3]}\right)$	Validated experimentally	[102]
Mass Balance of ammonia	$\frac{dM_{NH_{3,liq}}}{dt} = m_{NH_3,in,dig} - m_{NH_3,out,dig}$ $+ K_{OL}(K_d x [NH_4^+](t)$ $- [H^+](t)[NH_3](t))V_L$ $- \int m_{NH_{3,gas}} dh$	Mass balance over an imaginary slice of stripping column is integrated over the entire length	[98]

It is clear from Table 3 that most studies focused on the mass transfer of ammonia from the liquid phase into the stripping gas bubbles, while one study lists the mass transfer equations for stripping in a packed column. The assumptions on which these models are based vary between the studies, and therefore only the final form of the governing equation is listed in Table 3.

5.2 Ammonia speciation in the liquid phase

The speciation of ammonia is influenced by the ionic strength due to the complex chemistry of digestate and the different ionic interactions [103],[104]. Only a few studies have defined parameters to estimate the effect of the ionic interactions on ammonia speciation in digestate. Hafner et al. [104] used the Pitzer ion-interaction approach to introduce activity coefficients, which improved the accuracy of estimating FA concentrations in organic waste digestate. This approach is appropriate for concentrated solutions with ionic strengths greater than 0.1 mol/kg [62], [104]. Equation 7 describes the mathematical relationship to calculate the concentration of FA, taking into account these activity coefficients. The mathematical relationships describing the various constants required to calculate the activity coefficients are given in Equations (8) to (13) [105].

$$\frac{[NH_3]}{[NH_3 + NH_4^+]} = \frac{1}{1 + \frac{[H^+]}{K_d \times \gamma_{NH_4^+}}} \tag{7}$$

$$\log(\gamma_{NH_4^+}) = \frac{A \cdot \sqrt{I}}{1 + 2.5 \cdot B \cdot \sqrt{I}} \tag{8}$$

$$A = \frac{1.82483 \cdot 10^6 \cdot d^{1/2}}{(\epsilon T)^{3/2}} \tag{9}$$

$$B = \frac{50.2916 \cdot d^{1/2}}{(\epsilon T)^{1/2}} \tag{10}$$

$$\epsilon = 2727.586 + 0.6224107 \cdot T - 1075.112\ logT - \frac{52000.87}{T} \tag{11}$$

$$d = \frac{1+0.1342489 \cdot c^{\frac{1}{3}} - 0.003946263 \cdot c}{3.1975 - 0.3151548 \cdot c^{\frac{1}{3}} - 0.001203374 \cdot c + 7.48908 \cdot 10^{-13} \cdot c^{4}} \tag{12}$$

$$c = 647.26 - T \tag{13}$$

Nielsen at al. [102] attempted to correct hydrogen ion concentrations measured by a pH meter for the ionic strength in organic wastes such as pig manure. The term $[H^+]$ in Equation 2 could be replaced by the more accurate term $\{H^+\}$, which takes into account the ionic strength of the medium. Equations (14) to (16) describe the relationship between $[H^+]$ and $\{H^+\}$.

$$pH = -log_{10}\{H^+\} \tag{14}$$

$$[H^+] = \frac{1}{f^{H^+}}\{H^+\} \tag{15}$$

$$log_{10}f^{H^+} = -0.5 \cdot \left(\frac{\sqrt{I}}{1+\sqrt{I}} - b \cdot I\right) \tag{16}$$

The coefficient *b* depends on the nature of the waste and the ionic strength of the solution [106]. To conclude, it is recommended that the methods mentioned in the above studies are included to factor the effect of digestate chemistry on the speciation of ammonia. This would enable the construction of more sensitive models and can be extended to a variety of organic wastes.

5.3 Software-based modelling

Different software tools were developed and used to model the ammonia stripping process, such as Aspen and Modelica [78],[98],[99]. Errico et al. [78] used Aspen to model ammonia stripping from the liquid fraction of digestate, and used air as stripping gas and a flash drum to simulate the stripping out of CO_2, which reduces the buffer strength and increases the pH. The model predicted the increase of ammonia recovery as a function of gas flow rate. However, some simplifications were made. Speciation of ammonia was assumed to follow ideal behaviour and governing mass transfer equations were not defined. Furthermore, the digestate was assumed to only consist of dissolved salts, while solids and soluble molecules were neglected. Redford et al. [98] used Modelica to model ammonia stripping from digestate. They also assumed ideal solute behaviour and did not define the composition of the digestate. To define mass transfer, the authors described an ammonia mass balance across an infinitesimal slice of the stripping column and then integrated it over the entire length of the column. The study predicted an increase in pH of the digestate during the stripping process, which was validated experimentally. In [99], a Computational Fluid Dynamics (CFD) analysis of ammonia stripping from digestate in a packed bed column was carried out, and modelled the temperature and velocity profiles of the liquid and gaseous phases along the dimensions of the column. Here again, the composition of the digestate was not specified, while ammonia speciation and gas-liquid equilibria were defined considering ideal behaviour. Contrarily to the previously mentioned studies, the governing equations for mass transfer, continuity, and momentum were all defined in detail. While the studies above did not define the composition of digestate in detail, a study by Vaneeckhaute et al [107] developed a bio-physiochemical three-phase mathematical model that described ammonia stripping from digestate. The study used the software combination PHREEQC-Tornado/WEST to define the chemical speciation and governing mass transfer equations. Digestate composition was defined in detail in terms of organic and inorganic compounds based on the existing literature. This study was a demonstration of a proof of concept to model ammonia stripping and showed that the length of the stripping column did not have any effect on ammonia removal efficiencies, since the ammonia equilibrium between gas bubble-water was obtained very quickly.

6. Outlook and future research perspectives

Stripping has been proven by numerous studies to be an efficient technology for ammonia recovery from digestate, with extensive research performed on the positive influence of the governing process conditions (temperature, pH, and gas flow rate). Some noteworthy trends can be observed among the majority of the ammonia stripping studies reviewed in this paper, listed in Table 2. Most papers set a minimum of one process condition to its extreme value to ensure maximum ammonia recovery and most of them have used air as stripping gas, possibly due to its cost-effectiveness and ease of use.

Studies investigating side-stream stripping or in-situ configurations tend to avoid using air as stripping gas, as the presence of air can disrupt the AD process.

Linking a stripping column to an anaerobic digester can be seen as a two-fold method of extracting renewable gaseous fuels from the system, in the form of ammonia and methane. While all configurations have been successfully demonstrated, some motivations for the choice between these configurations could be the apparent tradeoff between extent of ammonia recovery and increasing methane yields. Side-stream stripping offers more flexibility in the stripping column and could, therefore, be a preferential choice for maximising ammonia recovery, albeit without a substantial increase in methane yield as discussed in Section 4.3. Conversely, pre-AD and in-situ stripping have been demonstrated to improve methane yields with a lower flexibility for increasing ammonia recovery in the stripping column.

In spite of the extensive research on ammonia stripping from digestate, there are still many avenues for further research. Table 2 shows the different stripping gases that have been used across various studies. There is no systematic study directly comparing these different gases and if a particular gas is better for ammonia removal. Such a comparison could provide information to future researchers to determine which stripping gas would be suited best for their application. The other topic is with regards to the composition of the digestate. Digestate from different organic waste sources (food waste, livestock waste, agricultural wastes etc.) can vary in terms of their TS content, organic content, TAN concentrations etc. These factors, specific to the waste source and composition, can influence the inter-phase area available for mass transfer and the amount of ammonium ions adsorbed onto solid particles, and thereby ammonia recovery. There is a lack of such a comprehensive experimental knowledge, investigating the effect of the type of digestate and digestate-specific factors on ammonia recovery. This type of studies would provide additional information on factors affecting ammonia recovery, and also be useful to determine whether it is easier to strip ammonia from digestate of a specific origin.

Another area that requires further research is the modelling of the ammonia stripping process. Comprehensive modelling of the process of ammonia stripping from digestate can be broken up into the following parts: (i) equations defining the mass transfer mechanisms of ammonia from liquid to gas phase, (ii) factors affecting ammonia recovery, (iii) composition of the digestate and (iv) composition of the stripping gas. While the mass transfer equations have been studied in much detail (see Table 3), there are fewer studies modelling the effect of the composition of the digestate and/or the stripping gas on ammonia removal. With respect to the digestate, the equilibrium between FA and NH_4^+ is an important stripping parameter. To factor the influence of the digestate composition on this equilibrium, some studies have incorporated activity coefficients. These activity coefficients depend on the interactions between various ions and present chemical species. However, not many studies have modelled the effect of physical digestate characteristics of the digestate on this equilibrium. Incorporating parameters such as TS, viscosity and organic compounds, and investigating their potential effects would greatly improve the accuracy and sensitivity of the models that utilise this equilibrium. With respect to the stripping gas, as discussed in Section 2.4, the fraction of CO_2 in the gas also affects the ammonia speciation. Whereas the influence of CO_2 on ammonia removal has been experimentally established, there has not been any modelling work performed on how the composition of the stripping gas or how the type of stripping gas itself (e.g., air, N_2, biogas, etc.) affect ammonia removal. Taking the stripping gas into account while developing the model or mass transfer equations would be another step in bridging the knowledge gap currently existing in the literature. The ideal practical model would be a black box with inputs like the process conditions, % CO_2 in stripping gas, type and composition of the digestate, reactor design, and the resulting output would be an accurate estimation of the ammonia removal from the liquid phase.

While the studies discussed in this review were all implemented at a laboratory scale, stripping of ammonia from digestate has also been implemented at pilot and industrial-scale plants. While full-scale ammonia stripping is an established process, there are still some potential disadvantages that can be addressed. Increasing temperature and gas flow rate to improve ammonia recovery can result in large heating and pumping energy costs. Similarly, increasing the pH of the liquid phase using alkaline agents will greatly increase the costs for chemicals, and also increase operational complexity. The use of $Ca(OH)_2$ to increase the pH has been known to precipitate carbonates, which can cause unwanted fouling or scaling problems. There are some potential solutions to compensate for the above-mentioned tradeoffs. Locating the plant near an energy source whenever possible is a way to minimise energy transport costs. In the cases where biogas is used as a stripping agent, the same gas stream can be used to produce energy in the grid. The usage of alkaline agents to increase the pH can be avoided by stripping out CO_2 as a form of pre-treatment, which gives the additional benefit of supplementary ammonia removal. In case the usage of alkaline agents is unavoidable, the addition of MgO is an alternative solution. While the addition of MgO increases the pH of the solution, it can be used to recover phosphorus as well as nitrogen via precipitation of struvite ($MgNH_4PO_4.6H_2O$). The precipitated struvite could then be valorised to offset chemical costs.

7. Conclusions

Ammonia recovery from the AD process does not only prevent inhibition but also prevents air and water pollution. While temperature, pH, and gas flow rate are the established process conditions that positively affect ammonia removal, the composition of the stripping gas can also have an important impact. Multiple studies have also linked a stripping column to an anaerobic digester in various configurations to control the ammonia concentration in the digester. While each configuration has its advantages and disadvantages, studies show that all configurations can successfully reduce the ammonia content in the digester and increase methane production. Similarly, multiple stripping column designs have been implemented in the literature to successfully strip ammonia from digestate. Finally, the ammonia stripping process can be modelled by defining mass transfer equations and estimating the true extent of ammonia speciation in the digestate by incorporating activity coefficients.

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