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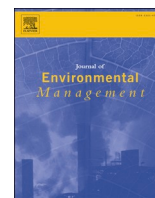
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Recovery and applications of ammoniacal nitrogen from nitrogen-loaded residual streams: A review

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ABSTRACT

Total ammoniacal nitrogen (TAN) is considered to be a pollutant, but is also a versatile resource. This review presents an overview of the TAN recovery potentials from nitrogen (N)-loaded residual streams by discussing the sources, recovery technologies and potential applications. The first section of the review addresses the fate of TAN after its production. The second section describes the identification and categorisation of N-loaded (≥ 0.5 g L⁻¹ of reduced N) residual streams based on total suspended solids (TSS), chemical oxygen demand (COD), total Kjeldahl nitrogen (TKN), TAN, and TAN/TKN ratio. Category 1 represents streams with a low TAN/TKN ratio (< 0.5) that need conversion of organic-N to TAN prior to TAN recovery, for example by anaerobic digestion (AD). Category 2 represents streams with a high TAN/TKN ratio (≥ 0.5) and high TSS (> 1 g L⁻¹) that require a decrease of the TSS prior to TAN recovery, whereas category 3 represents streams with a high TAN/TKN ratio (≥ 0.5) and low TSS (≤ 1 g L⁻¹) that are suitable for direct TAN recovery. The third section focuses on the key processes and limitations of AD, which is identified as a suitable technology to increase the TAN/TKN ratio by converting organic-N to TAN. In the fourth section, TAN recovery technologies are evaluated in terms of the feed composition tolerance, the required inputs (energy, chemicals, etc.) and obtained outputs of TAN (chemical form, concentration, etc.). Finally, in the fifth section, the use of recovered TAN for three major potential applications (fertilizer, fuel, and resource for chemical and biochemical processes) is discussed. This review presents an overview of possible TAN recovery strategies based on the available technologies, but the choice of the recovery strategy shall ultimately depend on the product characteristics required by the application. The major challenges identified in this review are the lack of information on enhancing the conversion of organic-N into TAN by AD, the difficulties in comparing the performance and required input of the recovery technologies, and the deficiency of information on the required concentration and quality of the final TAN products for reuse.

1. Introduction

1.1. Production and fate of ammonia

Ammonia (NH₃) is the world's second most produced chemical, of which major use is as nitrogen (N) fertilizer. About 80% of the NH₃ produced by industry is used in agriculture while the rest (20%) is used as raw material for the fabrication of chemical compounds and explosives (Fig. 1) (Erisman et al., 2008; Galloway et al., 2004). Nearly 1 – 2% of the total world energy production is used in the Haber-Bosch (HB) process to produce N-fertilizers, which represents a 3 – 5% of the global annual natural gas consumption and generates 4 – 8 ton CO₂e (equivalent carbon dioxide) per ton N-fertilizer per year. The

well-established HB process synthesises NH₃ from nitrogen (N₂) and hydrogen (H₂) at high temperatures and under high pressure. The generation of H₂ accounts for the largest part of the energy consumption of the process. When the H₂ is generated via methane (CH₄) reforming, the energy consumption of the HB is 28 MJ·kg-N⁻¹; if the H₂ is generated via water electrolysis, the energy consumption of the HB is 107 MJ·kg-N⁻¹ (Cherkasov et al., 2015). Even though the HB process has been largely improved and is very efficient and economically attractive, alternative methods for NH₃ production such as electrochemical-routes are being actively studied and developed (Garagounis et al., 2019; Giddey et al., 2013).

The successful synthesis of artificial NH₃ boosted the food production in the 20th century and brought great prosperity to the human

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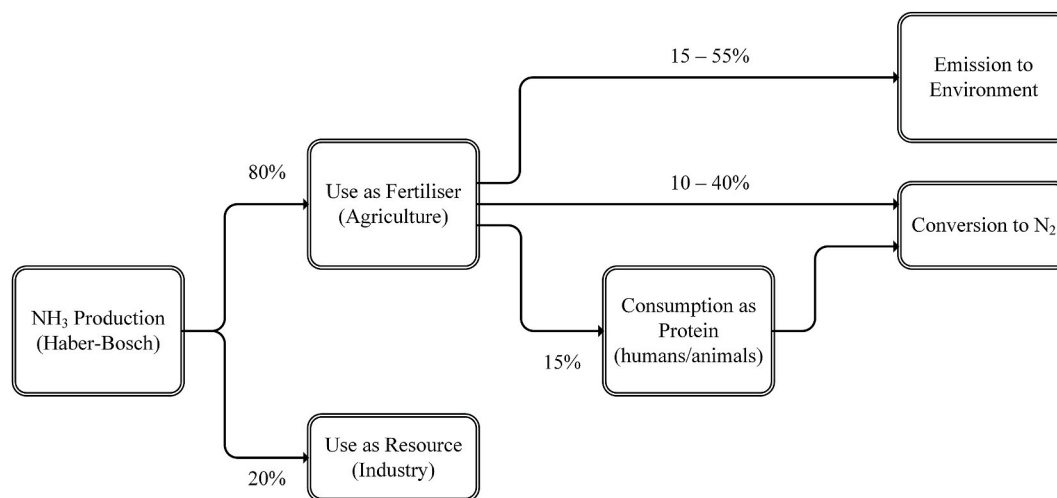


Fig. 1. Fate of produced NH_3 from Haber-Bosch (Erisman et al., 2008; Galloway et al., 2004).

society. Nowadays, more than 189 Million ton of NH_3 are synthesised annually (FAO, 2019). However, the overuse of N-fertilizers is creating an environmental and human health problem. For example, N can directly evaporate into the atmosphere or be flushed away from the soil by rainfall and end up in water bodies in the form of nitrates and ammoniacal nitrogen or TAN (the sum of dissolved ammonium NH_4^+ and ammonia NH_3), causing eutrophication (Erisman et al., 2007). Statistics show that only 16% of the total amount of annually applied fertilizers are consumed as vegetable and animal proteins (Fig. 1), the rest is lost into water bodies and the atmosphere. In the EU, 11 Million ton-year⁻¹ of N is currently applied to the crops, and 18 – 46% of the N are not being recovered from agriculture, sewage and food chain (especially slaughterhouses) waste streams (Afif et al., 2016; Buckwell and Nadeu, 2016).

About 10 – 40% of the lost N-fertilizers is denitrified (converted to N_2) either by vegetation or in wastewater treatment plants (WWTPs) (Erisman et al., 2007; Galloway et al., 2004; Matassa et al., 2015). They may also contribute to global warming and atmospheric pollution when transformed into gaseous oxidised-N species (Ju et al., 2016), such as nitrogen oxide (NO), nitrogen dioxide (NO_2), nitrous oxide (N_2O). In fact, some scientists suggest that the planetary limit for tolerable anthropogenic changes in the N-cycle has already been crossed (Rockström et al., 2009). To avoid environmental damage, stricter disposal regulations for N-containing waste (or residuals) should be met, while the sustainability of the NH_3 -cycle must be improved and removal and recovery of TAN is required (Galloway et al., 2008).

1.2. Treatment of residual waters that contain ammoniacal nitrogen

Conventionally, the ammonium (NH_4^+) is removed from residual waters by nitrification in combination with denitrification (N/DN). To achieve sufficient N removal, N/DN reactors require high hydraulic and solids retention times, which results in large footprints. Besides, nitrification needs abundant supply of oxygen (O_2) by addition of air, attaining liquid dissolved O_2 concentrations of $>2 \text{ mg L}^{-1}$, which represents up to 80% of the total energy consumption of sewage treatment plants (Siegrist et al., 2008).

Removal of NH_4^+ can also be achieved by the combination of partial nitrification and anammox (PN/A), which is typically applied to treat residual streams with relatively high NH_4^+ concentrations ($0.5\text{--}2.5 \text{ g L}^{-1}$), such as reject water of digested manure, waste activated sludge or landfill leachate (Gonzalez-Martinez et al., 2018; Lackner et al., 2014; Magrí et al., 2013). Because during PN/A, NH_4^+ is only partially oxidised, less energy for aeration is required than for N/DN. According to Lackner et al. (2014) and Schaubroeck et al. (2015), the energy consumption of NH_4^+ removal from reject water by N/DN is reported to be $57 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$,

whereas the application of PN/A to remove NH_4^+ from reject water requires $3\text{--}15 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$. Despite the energetic advantage of PN/A over N/DN, the application of PN/A is currently limited to (warm) side streams with low carbon to N ratios (C/N), as the preferred operating temperature of anammox bacteria is about 35°C and the growth of these bacteria is outcompeted by other bacteria species in the presence of high concentrations of organic carbon in the feed water (Gonzalez-Martinez et al., 2018). Besides, Lackner et al. (2014) reported that the stable operation of PN/A systems can be challenging due to the accumulation of solids, insufficient retention of the biomass and the accumulation of nitrite and nitrate.

Another challenge for the application of N/DN and PN/A is the generation and emission of gaseous oxidised N species, such as N_2O , NO and NO_2 . Especially the emission of N_2O is undesirable, because it is a potent greenhouse gas, having a 296 times higher global warming potential than CO_2 (Prather et al., 2001). According to the review of Desloover et al. (2012), the emission of N_2O during biochemical processes can contribute to 80% of the total greenhouse gas emissions of water treatment plants that process sewage, manure, landfill leachate or industrial effluents. The fraction of $\text{N}_2\text{O}\text{--N}$ emission relative to the total N load of full-scale biochemical N removal systems is reported to be 0 – 14.6% (Kampschreur et al., 2009; Vasilaki et al., 2019). In general, currently available literature shows that biochemical removal of NH_4^+ consumes energy and results in the emission of strong greenhouse gases.

1.3. Aim of the review

The biochemical conversion of NH_4^+ into N_2 at the expense of energy, excludes the potential to recover and reuse TAN. To this end, more focus has been brought onto the recovery of TAN from residual waters by both mature technologies such as chemical precipitation and stripping, as well as novel technologies such as reverse osmosis (RO) and electro-dialysis (ED) (Mehta et al., 2015a; Xie et al., 2016). However, recent reviews discuss the ongoing research or the different available technologies from the perspective of one specific TAN application, i.e., as a fertilizer (Mehta et al., 2015a), or as energy carrier (Valera-Medina et al., 2018).

Moreover, published reviews overlook or do not explore one important aspect of the TAN recovery potential, which is the existing available N-loaded streams and their composition, especially the N-speciation. N is mainly present in residual streams in two forms: organic and inorganic. Organic N refers to organic nitrogenous compounds such as proteins, whereas inorganic N refers to nitrite, nitrate and TAN.

In general, TAN concentration is the only considered parameter for TAN recovery potential assessment. According to a study by Mulder

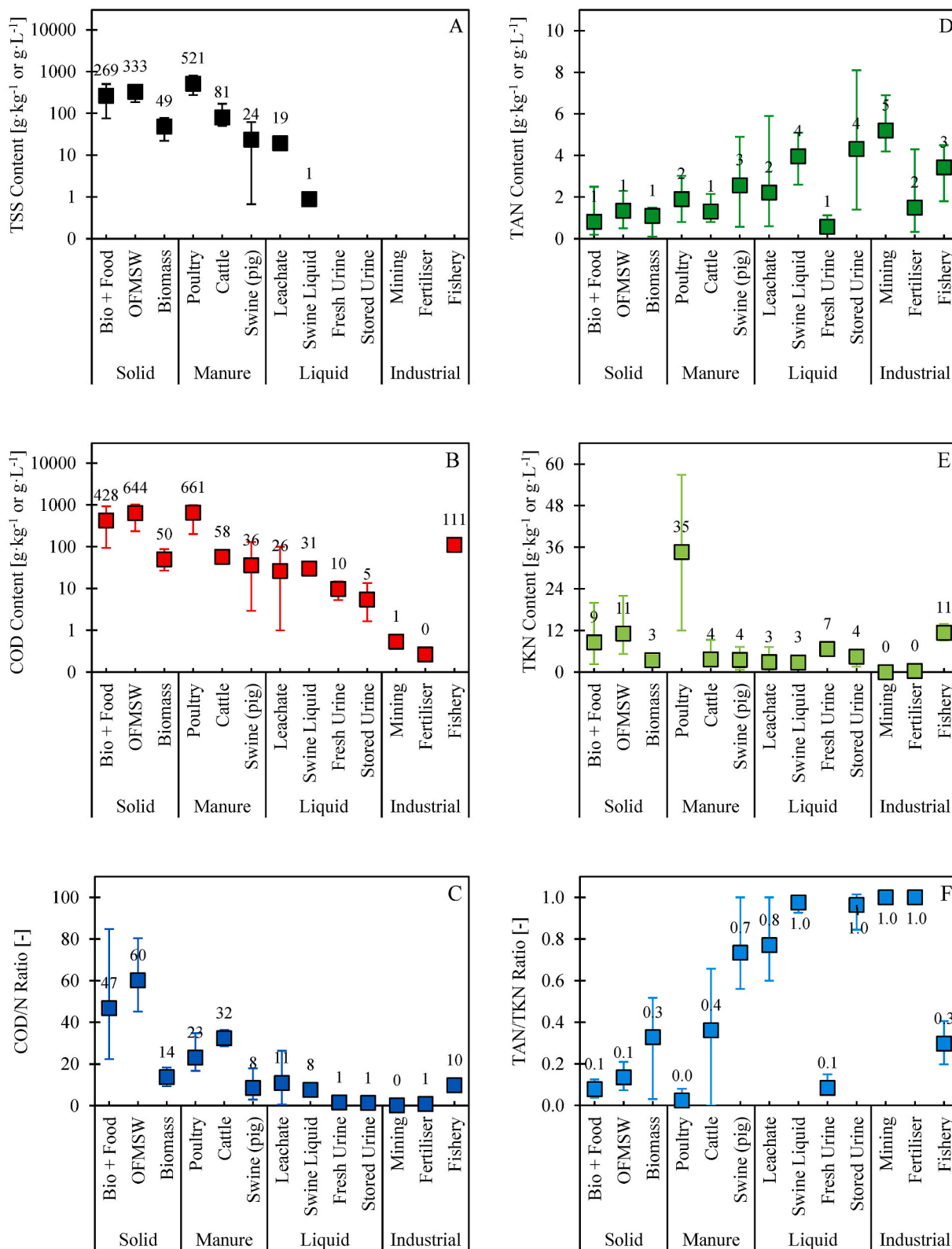


Fig. 2. An overview of the identified N-loaded residual streams and their characteristics in terms of TSS (A), COD (B), TKN (D) and TAN (E) content, and the respective calculated COD/N (C) and TAN/TKN (F) ratios. The presented values and error bars represent the averages and minimum and maximum values of at least three independently consulted references. The consulted references are extensively presented and referred to in the Supporting Information.

(2003), the treatment of residual streams with TAN concentrations < 0.1 g L⁻¹ is only cost-effective if biochemical TAN removal technologies are used. TAN recovery from streams with concentrations between 0.1 and 5 g L⁻¹ is technically possible but not cost-effective and only TAN

recovery from streams with concentrations > 5 g L⁻¹ is economically feasible. However, this study was published in 2003 and ever since, water treatment and TAN recovery technologies made great progress, not to mention the changes in the regulatory framework with regard to N

emissions (Lymeratou et al., 2015; Pikaar et al., 2018).

Currently, there is neither a clear identification of potential streams, nor related indicators to classify them according to their treatment requirements proposed or discussed so far. For example, residual streams rich in organic N could also be considered for TAN recovery if anaerobic digestion (AD) is applied first. Therefore, the presence of organic N and TAN in residual streams, quantified by the total Kjeldahl nitrogen (TKN, including organic N and TAN) content, should be considered as an important indicator for TAN potential classification of streams.

This review:

- identifies existing and potential N-loaded streams for TAN recovery;
- proposes a parameter-based categorisation for their treatment;
- discusses the available TAN recovery technologies based on their principle (concentrate TAN as NH_4^+ , NH_3 or separate TAN as NH_3 from the liquid), main energy input, end product and challenges;
- summarises the possible uses of the recovered TAN including specific requirements in terms of quantity and quality, and challenges for the TAN reuse technologies.

During the identification of suitable residual streams, we came across a wide range of descriptions of the term “nitrogen-loaded”: “nitrogen rich”, “high nitrogen content” and “high strength nitrogen”. In this review, the term “nitrogen-loaded” (hereafter N-loaded) refers to residual streams containing TKN concentrations of at least 0.5 g L^{-1} or $\text{g}\cdot\text{kg}^{-1}$. The overall aim of this review was to provide an objective and complete overview, without judgement on the preferable recovery techniques and application. Additionally, economic aspects were not discussed due to lack of information from collected literature preventing a fair and objective comparison.

2. Characterisation and categorisation of N-loaded residual streams

2.1. Parameters for characterisation of N-loaded residual streams

To characterise the N-loaded residual streams, we collected data on various key parameters to be considered for their treatment to allow for TAN recovery. These parameters are: the total suspended solids (TSS), the chemical oxygen demand (COD), the TKN and the TAN. To report consistently, we normalised the TSS, COD, TKN and TAN to parts per thousand, which corresponds to $\text{g}\cdot\text{kg}^{-1}$ and $\text{g}\cdot\text{L}^{-1}$ for solid and liquid streams (assuming a liquid density of $1000 \text{ g}\cdot\text{L}^{-1}$), respectively. The TSS indicates the feasibility to use directly physicochemical technologies for the recovery of TAN or the need for pre-treatment (e.g. filtration). The COD is an indication of the presence of organic matter, which must be decreased before discharge to receiving water bodies. Also, a high COD is likely to induce fouling in the physicochemical technologies for TAN recovery, indicating the need for pre-treatment.

The absolute TKN content is an indication of the amount of N that is present as both organic N and TAN. The TAN/TKN ratio indicates the dominant present form of N. Data collected in this review suggests that physicochemical technologies, such as stripping and precipitation, can be used for direct TAN recovery, i.e., pre-treatment steps such as conversion of organic N to TAN or solids removal is not needed, at a TAN/TKN ratio higher than 0.5; TAN/TKN ratios lower than 0.5 suggest that the organic N must be first converted to TAN to allow for recovery of TAN.

When it is necessary to convert organic N to TAN, the COD/N ratio must also be considered. Typically, when the COD/N ratio of residual streams falls within a certain range (i.e., 20 – 30), biochemical technologies such as AD are suitable to decrease the organic matter content, without potential problems of N shortage or inhibition (Rajagopal et al., 2013). Besides, the COD/N ratio is an indicator of fouling proneness in membrane operations (Feng et al., 2012). Although the C/N and COD/N are both used (Khalid et al., 2011; Mata-Alvarez et al., 2014; Rajagopal

et al., 2013), there are some differences. Compared to the COD/N ratio, the C/N ratio is less convenient to use because it cannot be directly related to CH_4 production (van Lier et al., 2020). In contrast, the COD can be used to estimate the CH_4 production, design and calculate the conversion efficiency of anaerobic systems (André et al., 2017). Regarding the N in the COD/N ratio, this could refer to either the TN (including both organic N and inorganic N) or the TKN. It must be noted that under anaerobic conditions the TKN is assumed to be equal to the TN so the COD/N ratio can be calculated with the TKN or TN.

2.2. Identification of N-loaded residual streams

We collected data on N-loaded residual streams obtained from approximately 150 studies, all using real (not synthetic) residual streams either for analytical or experimental research purposes. For each identified N-loaded residual stream, we used at least three independent references and report the average, minimum and maximum values of the characteristic parameters. We divided the N-loaded residual streams in four different groups, based on their origins: solid residual streams, manure, liquid residual streams (all domestic) and residual streams reported to originate from industrial processes. The obtained average, minimum and maximum values are presented in Fig. 2. In the text, we only refer to the average values. More details on the consulted references, such as the TSS, COD, TKN and TAN content, the respective units, used treatment technologies and reference details can be found in the Supplementary material.

2.2.1. Solid residual streams

The first group includes: bio- and food waste, the organic fraction of municipal solid waste (OFMSW) and spent biomass, such as the waste activated sludge from wastewater treatment plants (WWTPs) and algal sludge (Mata-Alvarez et al., 2000). Typical TSS values for bio- and food waste and OFMSW are 269 and $333 \text{ g}\cdot\text{kg}^{-1}$ (ranges can be consulted in Fig. 2), respectively, while the COD content is 428 and $644 \text{ g}\cdot\text{kg}^{-1}$, respectively. For the spent biomass streams, the TSS and COD are considerably lower than for bio- and food waste and OFMSW: 49 and $50 \text{ g}\cdot\text{kg}^{-1}$, respectively. The typical TAN content of the solid residual streams is $1 \text{ g}\cdot\text{kg}^{-1}$. Furthermore, the TKN ranges between 3 and $12 \text{ g}\cdot\text{kg}^{-1}$ and is mainly represented by the presence of proteins (Braguglia et al., 2018; Ganesh Saratale et al., 2018). The relatively low TAN/TKN ratios (ranging 0 – 0.3) indicate that direct TAN recovery will be challenging. To allow for effective TAN recovery, the TAN/TKN ratio must be increased by converting organic N to TAN. For bio- and food waste and OFMSW, the COD/N ratio is 47 and 60, respectively, whereas for spent biomass the COD/N ratio is 14, because of the lower COD content. Anaerobic (co-)digestion is a widely applied technology to treat these solid residual streams, due to the relatively high COD ($> 10 \text{ g}\cdot\text{kg}^{-1}$) and N contents ($> 0.5 \text{ g}\cdot\text{kg}^{-1}$) (Hartmann and Ahring, 2005; Keucken et al., 2018). Anaerobic (co-)digestion allows for the simultaneous decrease of the solids and COD content of the residual streams, while the organic N is converted to TAN, increasing the TAN/TKN ratio. More information on the use of AD to convert organic N to TAN is presented in Section 3.

2.2.2. Manure

The second group concerns manure, which is frequently reported to contain high levels of TAN and TSS, and is often considered to be problematic for its treatment via AD (Massé et al., 2014; Rodriguez-Verde et al., 2018). Manure is divided into poultry, cattle and swine manure. Poultry manure has the highest TSS, COD and TKN: 521, 661 and $35 \text{ g}\cdot\text{kg}^{-1}$, respectively. Despite the low TAN/TKN ratio (TAN/TKN \approx 0) of poultry manure, its absolute TAN content is high, i.e., $2 \text{ g}\cdot\text{kg}^{-1}$, practically 92% N is present as organic N in poultry manure. Cattle and swine manure have a much lower content of TSS, i.e., 81 and $24 \text{ g}\cdot\text{kg}^{-1}$, respectively, COD, i.e., 58 and $36 \text{ g}\cdot\text{kg}^{-1}$, respectively, and TKN, i.e., $4 \text{ g}\cdot\text{kg}^{-1}$. The TAN of cattle manure is $1 \text{ g}\cdot\text{kg}^{-1}$, whereas swine manure has a TAN of $4 \text{ g}\cdot\text{kg}^{-1}$. The N in cattle manure is predominantly

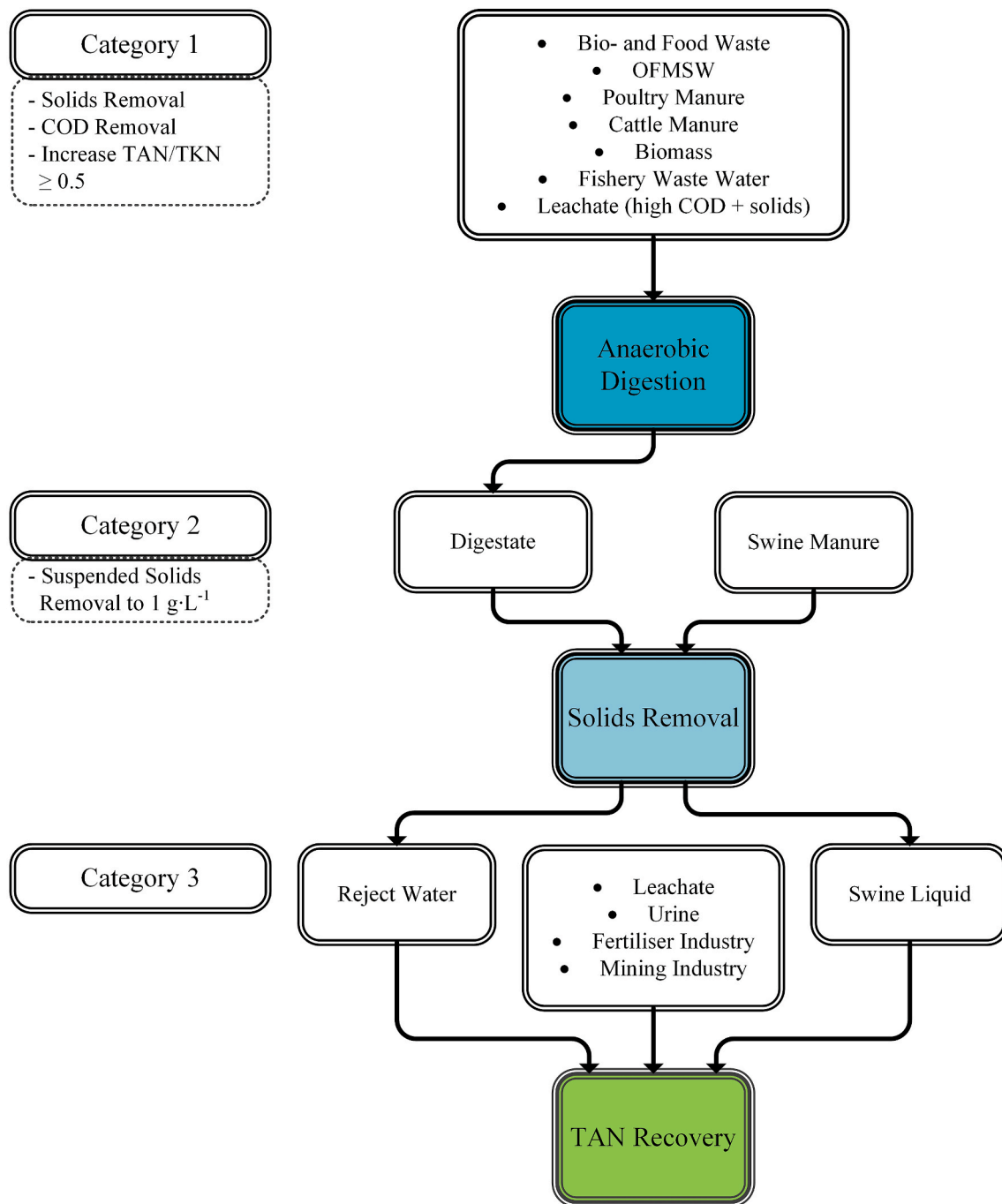


Fig. 3. A strategic categorisation of the various N-loaded residual streams, based on their characteristics and required (pre-)treatment before TAN recovery.

present as organic N (TAN/TKN ratio is 0.4), whereas in swine manure N is already present predominantly as TAN (TAN/TKN ratio is 0.7). The COD/N ratio of the various types of manure ranges between 8 and 32. According to the consulted studies, manure is mainly treated by AD but, due to the high level of TAN, is often co-digested with other organic residues to suppress the negative effects of the presence of TAN during the AD processes (Hartmann and Ahring, 2005; Mata-Alvarez et al., 2014).

2.2.3. Liquid residual streams

The third group includes: leachate, the liquid fraction of raw swine manure (swine liquid) and human (source-separated) urine. The TSS of swine liquid and urine streams is below $1 \text{ g}\cdot\text{L}^{-1}$, whereas conversely, leachates can contain high amounts of suspended solids ($19 \text{ g}\cdot\text{L}^{-1}$).

Regarding COD, leachates and swine liquid contain 26 and $31 \text{ g}\cdot\text{L}^{-1}$, respectively, whereas human urine ranges between 5 and $10 \text{ g}\cdot\text{L}^{-1}$. The TKN content for all the liquid N-loaded residual streams ranges between 3 and $7 \text{ g}\cdot\text{L}^{-1}$. For leachate, swine liquid and stored human urine, the TAN/TKN is at least 0.8 . Fresh human urine, however, has a TAN/TKN ratio of 0.0 , because N is still present as urea. When urine is stored, urea is hydrolysed to TAN, increasing the TAN/TKN ratio (Udert et al., 2003). When leachates contain high TSS and COD, and have a high COD/N ratio, anaerobic (co-)digestion can be applied for the treatment of the organic fraction (Lei et al., 2018; Montusiewicz et al., 2018).

2.2.4. Industrial residual streams

The fourth group concerns those N-loaded residual streams that have an industrial origin, such as mining and fertilizer industry and fish/

fishmeal processing. Amongst these industrial N-loaded residual streams, fishery residual water has the highest COD content ($110 \text{ g}\cdot\text{L}^{-1}$) and TKN content ($3 \text{ g}\cdot\text{L}^{-1}$), and the TKN is mostly present as organic N (TAN/TKN ratio of 0.3). Residual streams originating from mining and fertilizer industries have a much lower COD content (1 and $0 \text{ g}\cdot\text{L}^{-1}$, respectively), while all N is present as TAN (TAN/TKN is 1.0). The TAN content of mining and fertilizer industry residual streams is 5 and $2 \text{ g}\cdot\text{L}^{-1}$, respectively. For the treatment of fishery residual streams, AD has been used (Guerrero et al., 1999), whereas physicochemical TAN recovery technologies and biological oxidation processes were used to treat mining and fertilizer wastewater (Huang et al., 2011; Noworyta et al., 2003). Finally, there are also specific (industrial) residual streams that are not represented in Fig. 2, but are considered to be N-loaded and therefore potentially interesting for recovery. For example, TAN content of glutamate wastewater ranges between 16 and $19 \text{ g}\cdot\text{L}^{-1}$ (Wang et al., 2011; Yang et al., 2005); pectin wastewater can contain around $1.4 \text{ g}\cdot\text{L}^{-1}$ (Degn Pedersen et al., 2003); slaughterhouse wastewater $\sim 0.7 \text{ g}\cdot\text{L}^{-1}$ (Kundu et al., 2013); nuclear wastewater $\sim 35 \text{ g}\cdot\text{L}^{-1}$ (Gain et al., 2002); coking wastewater can contain between 0.2 and $0.6 \text{ g}\cdot\text{L}^{-1}$ (Jin et al., 2013; Lin et al., 2018) and ion exchange brine up to $3.9 \text{ g}\cdot\text{L}^{-1}$ (Vecino et al., 2019). These residual streams also have a high potential

for TAN recovery, but insufficient information on their composition and current treatment is available to consider them in this review.

2.3. Discussion on categorisation of N-loaded residual streams

Based on their characteristics related to the required (pre-)treatment before TAN recovery, the N-loaded residual streams are divided into three categories:

1. Residual streams with a TAN/TKN ratio lower than 0.5;
2. Residual streams with a TAN/TKN higher than 0.5 and TSS higher than $1 \text{ g}\cdot\text{L}^{-1}$;
3. Residual streams with a TAN/TKN higher than 0.5 and TSS lower than $1 \text{ g}\cdot\text{L}^{-1}$, suitable for direct TAN recovery.

Fig. 3 presents a strategic distribution of these various categories, based on the need of (pre-)treatment before subsequent technologies can be applied for the recovery of TAN.

2.3.1. Category 1: TAN/TKN < 0.5

Category 1 contains N-loaded residual streams with a TAN/TKN ratio

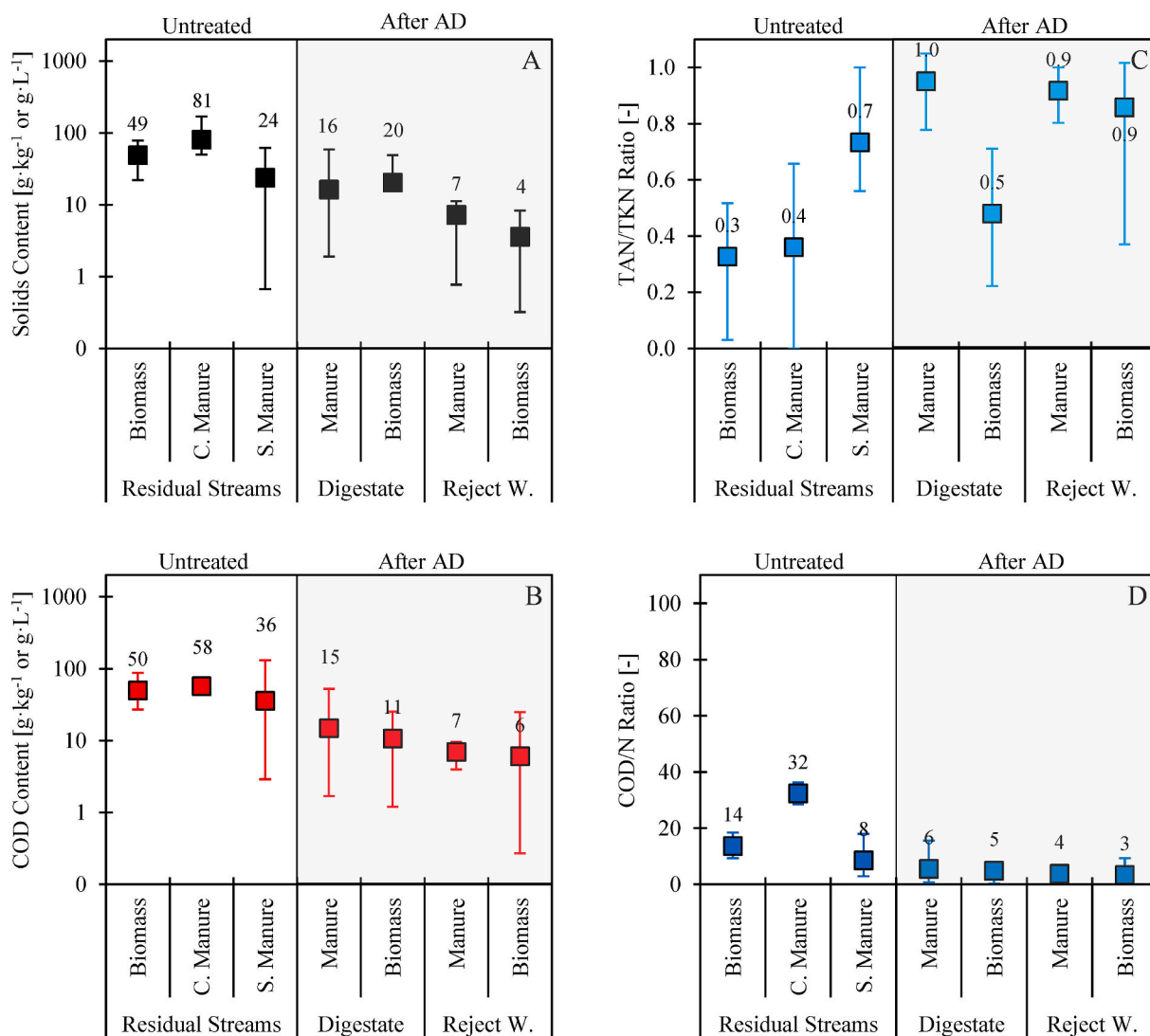


Fig. 4. Change in the solids content (A), COD content (B), TAN/TKN ratio (C), and COD/N content (D) in the obtained digestates and reject waters resulting from AD applied to biomass and manure. Note the exponential y-axis in figures A and B. The presented values and error bars represent the averages and minimum and maximum values of at least three independently consulted references. The consulted references are extensively presented and referred to in the Supporting Information.

< 0.5 and a TSS and COD content both higher than 24 and 36 g·kg⁻¹, respectively. For these streams, the TAN/TKN must be increased to at least 0.5 to allow for subsequent effective TAN recovery; Christiaens et al. (2019) reported 0.5 as the lowest TAN/TKN ratio for which TAN recovery is applied from biomass digestate. The N-loaded residual streams that require this organic N to TAN conversion step are bio- and food waste, OFMSW, spent biomass, poultry and cattle manure. Various biochemical and physicochemical processes, such as AD, can be used to increase the TAN/TKN ratio by conversion of organic N to TAN, while simultaneously the TSS and COD content is decreased. The conversion of organic N to TAN by AD is more extensively discussed in Section 3.

2.3.2. Category 2: TAN/TKN ≥ 0.5, TSS > 1 g·L⁻¹

Category 2 contains N-loaded residual streams with a TAN/TKN ratio greater than 0.5 and TSS concentrations higher than 1 g·L⁻¹. The application of AD to treat various organic N-loaded residual streams from category 1 leads to the generation of digestate (which falls into this category), having a TAN/TKN ratio greater than 0.5. Direct TAN recovery is possible by solids-tolerant recovery technologies, such as struvite precipitation and air stripping (see Section 4) for digestate with TSS up to 1 g·L⁻¹. However, for recovery technologies that are prone to fouling (mostly membrane-based technologies), the feed stream must be made free from solids by using solid-liquid separation (centrifugation or belt-press filtration), sedimentation, sand filtration, micro- or ultrafiltration.

2.3.3. Category 3: TAN/TKN ≥ 0.5, TSS ≤ 1 g·L⁻¹

Category 3 contains N-loaded residual streams with a TAN concentration higher than 0.5 g·L⁻¹, a TAN/TKN ratio greater than 0.5 and TSS ≤ 1 g·L⁻¹. Within category 3, the liquid residual streams, such as solids free leachate, filtered swine liquid and urine, and various industrial N-loaded residual streams originating from mining and fertilizer industry are placed. According to the obtained data with respect to applied treatment technologies, these residual streams are considered suitable for direct TAN recovery by technologies discussed in Section 4.

2.3.4. Categorisation of N-loaded residual streams

The proposed categories indicate the suitability and the path for TAN recovery. If we take category 1 (rich in organic N and low TAN/TKN ratio) as an example, the organic N needs to be converted firstly to TAN to increase the TAN/TKN ratio, usually carried out by AD, then the TAN in the effluent has to be separated from the solids, and finally the liquid stream is suitable for TAN recovery. Therefore, category 1 streams require the most (pre-)treatment steps, whereas category 3 streams, on the other hand, require the least. The streams in category 3 can be found in specific industries, e.g. mining and chemical industries, and have a high potential for TAN recovery because of their high TAN concentrations and low TSS content. However, the access to this information is usually limited. Therefore, the streams from category 2 are commonly considered as having the greatest potential for TAN recovery.

3. Conversion of organic N to TAN by AD

3.1. Conversion of organic N to TAN

Both biochemical and physicochemical processes can be used to convert organic N to TAN. Biochemical processes, in particular AD, is predominantly used in commercial applications. Besides the conversion of organic N to TAN, AD allows for resource recovery from rather diluted streams, i.e., biogas production and nutrient recovery during post-treatment, while having low operational costs (Oladejo et al., 2018). On the other hand, biochemical processes have slow kinetics resulting in long residence times, ranging between one to five weeks for AD digesters, and consequently large reactor footprints.

When dealing with poorly biodegradable substrates, e.g. spent biomass, physicochemical processes such as pyrolysis, gasification and

wet air oxidation might be preferred over AD (Oladejo et al., 2018). The conversion time of these processes ranges from seconds to minutes and 80 – 90% conversion efficiencies can be achieved (Oladejo et al., 2018). However, physicochemical processes require extensive pre-treatment (e.g. exhaustive drying) and complex reactor designs to handle high temperatures and pressures. For example, hydrothermal liquefaction of algae requires temperatures in the range of 250 – 350 °C and pressures in the range of 40 – 250 bar (Jazrawi et al., 2015). Furthermore, large chemical and/or energy inputs are required and sometimes expensive catalyst must be used (Elliott et al., 2013; Oliviero et al., 2003). Therefore, even though physicochemical processes allow for higher organic N to NH₃ conversion rates and efficiencies, their practical application is rather limited.

Besides the mentioned treatment methods, hydrothermal pre-treatment and alkali dosage are also frequently discussed (Ahmad et al., 2018; Carrere et al., 2016). These methods are proven to be suitable for enhancing the hydrolysis of various complex substrates, especially biomass, and therefore can be used to enhance the organic N conversion. However, no data is found on organic N or protein conversion efficiency.

Fig. 4 shows the effect of the application of AD by comparing the characteristics of the residual streams (raw substrates) before and after AD, referred to as digestate and reject water, the latter being the liquid fraction of the digestate obtained after solid-liquid separation by centrifuges, screw presses, etc. When AD is used, the TSS and COD of the residual streams are reduced by 30 – 85% depending on the streams. A high degree of removal is especially desirable when membrane-based TAN recovery technologies are considered, because of reduced fouling risks (Zarebska et al., 2015). The TSS of the untreated streams, which ranges between 24 and 81 g·kg⁻¹ or g·L⁻¹, is reduced to 16–20 g·L⁻¹ in the digestate and to a range of 4 – 11 g·L⁻¹ in the reject water (see Fig. 4A). Regarding COD, the initial content of the residual streams is reduced from 36 to 58 to 11 – 15 g·kg⁻¹ or g·L⁻¹ in the digestate and to 6 – 7 g·L⁻¹ in the reject water after AD (see Fig. 4B). The decrease in COD content ultimately results in a decrease of the overall COD/N ratio, as presented in Fig. 4D, since the total N content remains unaffected (only transformed into TAN) by AD.

Furthermore, Fig. 4C shows that the application of AD leads to a substantial increase in the TAN/TKN ratio, up to 0.9 – 1 in both digestate and reject water, for most of the residual streams here considered, indicating the effective conversion of organic N to TAN. Only biomass appears to be a substrate for which limited conversion can take place, as the TAN/TKN ratio of its digestate is only 0.5. Note that swine manure possesses already a high TAN/TKN ratio (0.7) before AD, whereas the TAN/TKN ratio for biomass and cattle manure does not exceed 0.5.

Because AD is widely applied commercially and as seen here, it can be considered an effective method to convert organic N to TAN. The next subsection will further elaborate on its mechanisms and limitations in the conversion of organic N to TAN.

3.2. Mechanisms for biochemical conversion of organic N to TAN during AD

Proteins are considered to be the most common organic N compound present in wastewaters. Except proteins, organic N can also exist as urea, quaternary ammonium salts (such as betaine), and melanoidins. The amount of urea present in animal manure is prominent, but it is easily hydrolysed into TAN without additional conversion steps. Betaine is present in low concentrations (0.8 – 1.6% dry weight of sugar-beet) (Thalasso et al., 1999) and melanoidins have a poor biodegradability (6 – 7%) (Pazouki et al., 2008). Both groups of compounds have a low potential for TAN recovery and were not further considered in this review. In fact, only protein-rich residual streams have been considered as the main organic N-loaded streams that need a conversion step before TAN recovery.

The conversion of proteins to TAN under anaerobic conditions has

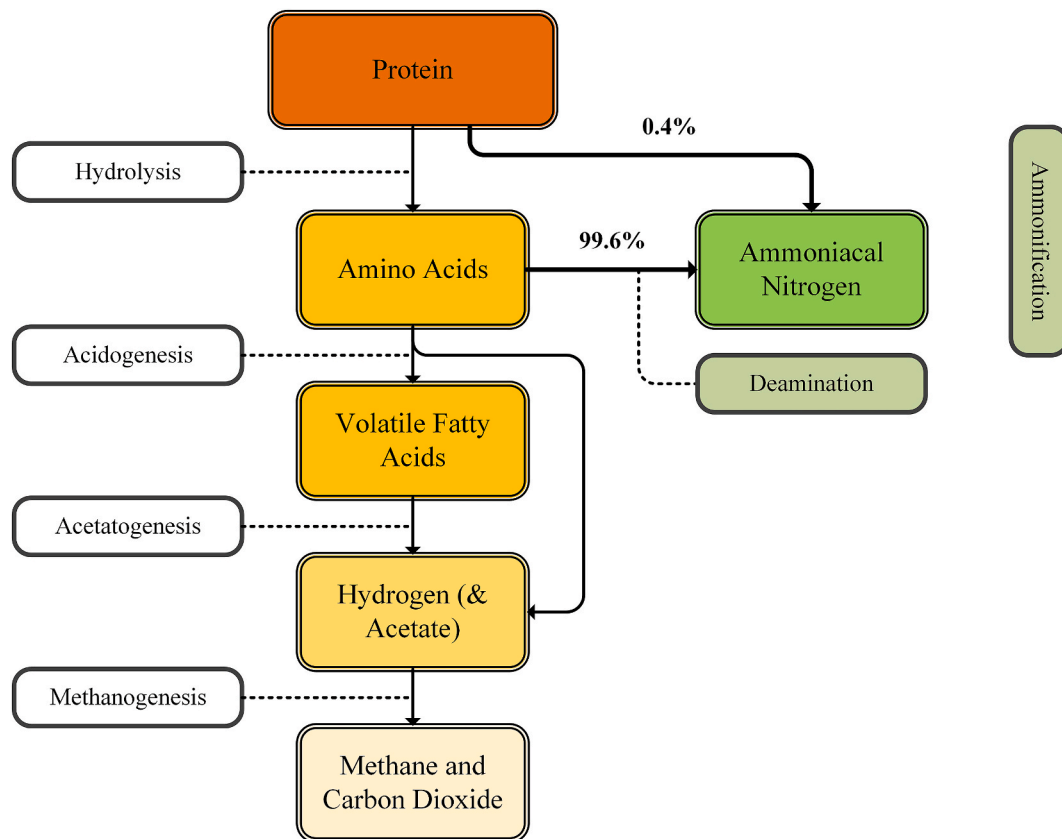


Fig. 5. - Schematic representation of protein degradation during AD. Formation of TAN particularly occurs during the first stages of AD (assuming 100% protein to TAN conversion efficiency), adapted from (Kayhanian, 1999; McInerney, 1988).

been widely studied (Kayhanian, 1999; McInerney, 1988) and is schematically represented in Fig. 5. The full conversion of protein into TAN is called ammonification. The ammonification process includes two main steps: the hydrolysis of the proteins to peptides and subsequently to amino acids by proteases and peptidases (i.e., proteinases); and the deamination of the amino acids to TAN, volatile fatty acids (VFAs), and H_2 (McInerney, 1988). The deamination process is included in the acidogenesis process, in which the amino groups ($-NH_2$) are removed from the amino acids (Ladd and Jackson, 1982). TAN is partly produced (0.4%) during hydrolysis, while the major part (99.6%) is produced during deamination assuming a 100% protein to TAN conversion efficiency (Dakin, 1920). Therefore, the conversion of organic N to TAN in AD can be simplified as proteins (hydrolysis) → amino acids (deamination) → TAN (Bareha et al., 2019; Batstone et al., 2002).

3.3. Limiting factors of conversion of organic N to TAN by AD

The conversion of organic N to TAN mostly is not fully completed during AD of non-synthetic residual streams; its efficiency has been reported to be in the range of 18 – 77% and depends largely on the substrate and the operational conditions (Bareha et al., 2018). According to literature, the rate-limiting step of the conversion is hydrolysis, specifically for complex substrates, e.g. food waste and algal biomass (Kobayashi et al., 2015; Zhang et al., 2014). When hydrolysis is impeded, the availability of amino acids is limited and the subsequent deamination can only take place to a certain degree.

Proteins are complex molecules that can contain more than 100 amino acids and can roughly be divided into insoluble fibroid and soluble globular proteins (Powar and Chatwal, 2007). Fibroid proteins, e.g. in manure fibres, are more resistant to hydrolysis because they are less soluble than globular proteins (Sanders, 2001). Besides, humic acids can decrease the solubility of proteins when binding to them, forming larger

and more complex molecules. In fact, Sanders (2001) reported that the hydrolysis of protein is inhibited at humic acids concentrations higher than $1 \text{ g} \cdot \text{L}^{-1}$.

A recent study revealed that the N biodegradability (understood as organic N conversion potential) varies before and after storage (Bareha et al., 2018). The potential can be roughly indicated by TAN/TKN. If the TAN/TKN is higher than 0.3 before AD (e.g. during storage of manure or food wastes), the organic N left in the substrate is more resistant to degradation. This could be attributed to certain uncontrolled storage conditions (e.g. decreasing pH) that can promote partial conversion of the organic N to TAN, while the unconverted remaining organic N can form more stable compounds, which impedes the hydrolysis (Bareha et al., 2018). Uncontrolled storage of complex substrates seems to be less preferable in terms of organic N conversion.

Besides the impeded hydrolysis of proteins, also the produced TAN from proteins is regarded as a problem for AD. In earlier studies, TAN is believed to inhibit mainly the methanogenesis phase (Angelidaki and Ahring, 1993). Whereas in later studies, TAN is also found to affect the overall AD processes. For example, Niu et al. (2014) found similar IC_{50} values (i.e., half-maximal inhibitory concentrations, in this case TAN, for which the conversion efficiency is reduced by 50%) for methanogenesis, acidogenesis and hydrolysis, which were 5.1, 5.3 and 5.7 $\text{g} \cdot \text{N} \cdot \text{L}^{-1}$, respectively. According to Nakakubo et al. (2008), the addition of TAN caused a decrease in CH_4 production, but no immediate VFA accumulation was observed, indicating that hydrolysis and acidogenesis were also inhibited. Protein conversion rates and efficiencies were found inhibited by TAN at concentrations above 2 $\text{g} \cdot \text{N} \cdot \text{L}^{-1}$ (Chen et al., 2018; Gallert et al., 1998; Krylova et al., 1997). Krylova et al. (1997) reported that TAN concentrations ranging between 2.6 and 8 $\text{g} \cdot \text{N} \cdot \text{L}^{-1}$ significantly reduce the population of the proteolytic group (i.e., microbial group producing protease). Gallert et al. (1998) found 50% inhibition of deamination at TAN concentrations of 3.0 $\text{g} \cdot \text{N} \cdot \text{L}^{-1}$ and NH_3

Table 1
Effect of TAN and NH₃ concentrations (under certain pH and temperature) on organic N conversion by AD.

pH	Temp.	TAN	NH ₃	Impact on conversion of organic N	Reference
–	°C	g-N·L ⁻¹	g-N·L ⁻¹		
7.4–8.3	55	3.0	0.3–1.3	0% inhibition	Niu et al. (2014)
7.4–8.3	55	6.0	0.5–2.6	75% inhibition	
<7.0	37 or 55	7.9	0.1–0.2	10% reduction of proteolytic group population	Krylova et al. (1997)
<7.0	37 or 55	13.2	0.2–0.5	40% reduction of proteolytic group population	
8.0–8.2	35	3.0–5.0	0.3–0.5	40% inhibition	Niu et al. (2013)
8.0–8.2	35	5.0–15.0	0.5–1.5	80% inhibition	
7.3	37	2.0	0.1	50% reduction of proteobacteria	Chen et al. (2018)
7.3	37	7.8	0.2	100% inhibition	
7.2	37	3.0	0.1	50% inhibition	Gallert et al. (1998)
7.2	55	2.0	0.3	50% inhibition	

concentration of 95 mg-N·L⁻¹ under mesophilic conditions, and at TAN concentrations of 2.0 g-N·L⁻¹ and NH₃ concentration of 274 mg-N·L⁻¹ under thermophilic conditions. Similarly, Niu et al. (2013) reported a rapid decrease in protein conversion efficiency when TAN was above 5.0 g-N·L⁻¹. Table 1 presents an overview of the TAN and NH₃ concentrations, as well as the operational conditions (i.e., pH and temperature), and their effect on the conversion of organic N to TAN found in literature.

3.4. Methods to enhance the conversion of organic N to TAN

To deal with the protein-rich streams, many methods have been applied, such as co-digestion, pre-/side-treatments and adaption, to stabilize the AD system and promote CH₄ production. In general, protein is only considered part of the total COD and few studies were found that especially focused on protein conversion. Therefore, we have summarized the effect of the most often applied or studied methods for protein conversion.

Co-digestion with carbohydrate-rich streams, such as starch or glycerine, is applied to increase the C/N ratio to avoid TAN inhibition. Based on the collected data, co-digestion is detrimental to protein conversion rate and efficiency. The presence of carbohydrates suppress the production of protease and the increased C/N ratio induces a lower protein solubilisation rate (Glenn, 1976; Wang et al., 2014). Besides, increasing the C/N by co-digestion promotes bacterial growth and TAN concentration might be reduced by the N consumption during this growth (Verma, 2002).

Pre-treatment methods are also applied, to improve the CH₄ production during anaerobic treatment of protein-rich streams. Guerrero et al. (1999) found that 80% protein conversion can be achieved by the acidification of fishmeal wastewater. Microwave pre-treated sewage sludge showed a 10% higher solubility of organic N compared to the untreated sludge, but still the effect on its conversion efficiency is not clear (Gil et al., 2018). Side-treatments (e.g. side-stripping, side-adsorption) are used to reduce the NH₃ accumulation in AD systems, especially when TAN/TKN ratio is above 0.5 (Kabdasi et al., 2000; Resch et al., 2011). The effect of these side-treatments on the organic N conversion has not been studied. Only one study reported that increased temperature and alkalinity of the side-stripping stream improved the organic N solubility, resulting in a 20–33% increase in the organic N conversion efficiency (Serna-Maza et al., 2014).

In the past decade, adaption of AD biomass towards inhibitive TAN concentrations has been studied, aiming at restoring the CH₄ production after an episode of TAN inhibition. The activity of the proteolytic group is found closely related to the protein conversion rate, therefore, adaption is considered to promote the restoration of the protein conversion

rate of TAN inhibited biomass (Chen et al., 2018). The successful adaption of methanogens to a TAN concentration of 5 g-N·L⁻¹ in an AD reactor was reported by Fotidis et al. (2014). Without interrupting the process or replacing the substrate, they observed a 30% higher CH₄ production in the bio-augmentation reactor (with addition of methanogenic culture) than the control reactor within 41 days. Chen et al. (2018) reported that protein degradation decreased noticeably when TAN increased to 5.5 g-N·L⁻¹, but 90% protein conversion rate (relative to the conversion rate of the non-inhibited biomass) can be restored after adaption. Likewise, Kovács et al. (2013) found 3.5–5 times higher protease activity in the biomass after adapted to increasing protein concentrations.

3.5. Discussion on AD to convert organic N to TAN

AD is used to convert the organic N to TAN, while reducing COD and TSS simultaneously. However, the N contained in the protein was not always completely converted to TAN. For complex substrates like spent biomass, the maximum conversion of TKN to TAN is only 50% (Christiaens et al., 2019). Recently, studies are trying to predict the fate of N during anaerobic digestion, Bareha et al. (2018) estimated the accessibility of organic N in complex substrates by chemical extraction methods (Van Soest fraction and EPS fraction). Their study reports that the organic N conversion efficiency cannot be accurately predicted by the biodegradability or chemical accessibility of the substrate. Therefore, for evaluating the potential recovery of TAN from organic N-loaded streams, there is a need for estimating the actual TAN potential of these streams.

The wide range in the reported organic N conversion efficiencies (18–77%) might be caused by limited hydrolysis of complex proteins, but can also be attributed to the inhibitory effects of accumulating TAN. Although extensive work was done to study TAN inhibition effects and mechanisms, there is no agreement on either the ‘intrinsic’ TAN inhibitory concentration, nor the inhibitory form of TAN (NH₄⁺ or NH₃). Probably because these studies were done with various substrates and under various operational conditions (pH and/or temperature), which make it difficult to compare the results. Moreover, studies reporting on the effect of TAN on organic N or protein conversion are rather limited, increasing the difficulties for a systematic analysis or review.

Unsurprisingly, the currently applied or studied methods to relieve the stress that protein-rich streams pose on AD, such as co-digestion, pre-/side-treatments and adaption, are all focusing on the CH₄ yield. Based on the collected data, no firm conclusion can be made. Generally, studies show that increasing C/N (or COD/N) ratio by co-digestion can negatively affect the organic N conversion efficiency, and more importantly, this method dilutes the TAN concentration in the AD, which has an additional impact on the potential TAN recovery efficiency downstream. Side-stripping removes NH₃ from the AD system, and therefore is supposed to improve the organic N conversion: on the one hand it reduces the inhibition by NH₃, and on the other hand it enhances the protein hydrolysis efficiency by applying higher temperatures and pH than in the AD reactor. However, the improvement (10–33%) reported might be less attractive considering the required energy input (further elaborated in Section 4). Direct NH₃ stripping in AD reactors is also being studied, however, AD reactors are rather unstable at high temperatures (> 55 °C) and high pH (> 8) that is needed for the stripping process (Gebreyessus and Jenicek, 2016). Combining the two technologies can either result in AD reactor failure or low concentration NH₃-containing end products. Adaption does not improve the conversion efficiency but allows for restoration of the conversion after TAN inhibition. Although the process takes longer, adaption might be advantageous for maintaining both the conversion efficiency and keeping the concentrated TAN in the effluent, which might be advantageous for TAN recovery after AD.

4. Technologies to recover TAN from residual streams

4.1. Technologies to concentrate TAN

Solid–liquid separation or complete solids removal is suggested prior to TAN recovery; mature technologies such as centrifugation, media (sand) filtration or microfiltration and ultrafiltration, can be applied to reduce an initial TSS to below $1 \text{ g}\cdot\text{L}^{-1}$ (2%) (Masse et al., 2007; Zarebska et al., 2015). The technologies discussed below are considered to be used at a TSS concentration lower than $1 \text{ g}\cdot\text{L}^{-1}$ or else after solids removal technologies.

4.1.1. Reverse and forward osmosis

In reverse osmosis (RO), the residual water is pressurised to allow for water permeation through a membrane that rejects practically all dissolved substances. To effectively use RO as concentration technology by rejecting TAN and decrease the volume of the residual, TAN must be present as NH_4^+ , because uncharged NH_3 can easily permeate through the membrane (Masse et al., 2008). For example, Mondor et al. (2008) and Gong (2013) reported final TAN concentrations of 12.8 and $12 \text{ g}\cdot\text{L}^{-1}$ in the concentrate after using RO to treat filtered swine manure reject water, respectively. The concentration factor (CF), relating the final achieved concentration to the ingoing concentration of the respective residual water, was 1.5 and 3.6 , respectively. Ledda et al. (2013) achieved TAN concentrations of 5.7 and $7.3 \text{ g}\cdot\text{L}^{-1}$, with a CF of 4 , after treating cow and swine reject water pre-treated by ultrafiltration, respectively. In addition, Schoeman and Strachan (2009b) obtained $1.8 \text{ g}\cdot\text{L}^{-1}$, CF of 2 , after concentrating solid waste leachate by RO. Finally, Fu et al. (2011) used RO to concentrate TAN in simulated acid scrubber effluent and reported a final TAN concentration of $12.6 \text{ g}\cdot\text{L}^{-1}$ (CF of 3 , based on the reported volume reduction and NH_4^+ rejection) and Noworyta et al. (2003) produced an RO concentrate with a TAN concentration of $11 \text{ g}\cdot\text{L}^{-1}$ (CF of 8) after treating NH_4NO_3 condensate from fertilizer industry.

Forward osmosis (FO) uses a saline draw solution to force water permeation from the feed water by osmosis, while most (except volatile) dissolved substances are rejected by a membrane. Holloway et al. (2007) reported a water recovery of 70% and a NH_4^+ rejection of 92% for the use of FO to concentrate nutrients in filtered sludge reject water, resulting a final TAN concentration of $4 \text{ g}\cdot\text{L}^{-1}$ (CF of 3). Interestingly, even though the same water recovery was achieved on manure digestate by Li et al. (2020), the authors did not succeed to concentrate TAN by FO as the rejection of NH_4^+ was less than 40% .

According to the consulted studies, the maximum CF that can be achieved by RO and FO for TAN in residual streams is about 6 and 3 , respectively. The CF for RO and FO is mainly limited by the water recovery. Furthermore, as a result of the effective rejection (and thus concentration) of substances such as humic acids and multivalent ions by RO and FO, membrane fouling was observed in many studies (Holloway et al., 2007; Li et al., 2020; Masse et al., 2007; Xie et al., 2016), even when pre-treatment by (membrane) filtration was applied. Hence, extensive membrane cleaning is required, to allow for stable operation to concentrate TAN by RO and FO when substances in the feed stream are present that induce particulate fouling, scaling or organic fouling. Finally, because the membranes reject practically everything, also other concentrated compounds (organics and salts) are present in the concentrated NH_4^+ solution.

4.1.2. (Bio-)electrochemical cells

In (bio-)electrochemical cells ((B)ECs), TAN is transported as NH_4^+ from the anode compartment, through a cation exchange membrane, to the cathode compartment when an electric current is applied. In the last decade, these technologies have been widely applied to recover TAN from residual streams as urine and reject water, containing NH_4^+ feed concentrations up to $4 \text{ g}\cdot\text{L}^{-1}$ (Kuntke et al., 2018; Rodríguez Arredondo et al., 2015). The various types of (B)ECs comprise microbial fuel cells

(MFCs), microbial electrolysis cells (MECs) and electrochemical cells (ECs). While MFCs are actually able to recover TAN and simultaneously produce energy, the highest reported NH_4^+ fluxes ($0.08 \text{ kg}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) are 6.5 times lower than for MECs ($0.52 \text{ kg}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and 4.8 times lower than for ECs ($0.38 \text{ kg}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Kuntke et al., 2018). The higher NH_4^+ fluxes in MECs and ECs are attained at the expense of external electricity supply, which results in a higher energy consumption for MECs ($4 - 22 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$) and ECs ($18 - 94 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$) compared to the energy producing MFCs ($- 10 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$) (Kuntke et al., 2018). The wide range of reported energy consumptions by the (B) ECs can be explained by the very wide range of achieved TAN removal efficiencies ($1 - 100\%$). Based on the reported (B)ECs data, the efforts to decrease the electrode and membrane areas, for which increased NH_4^+ fluxes are required, led to higher energy consumptions.

(B)ECs are actually used to concentrate TAN, and are mostly combined with stripping and scrubbing of NH_3 , allowing for actual TAN recovery (Kuntke et al., 2018). Hence, very little attention is paid to the concentrations of TAN obtained in the cathode compartment. The study of Ledezma et al. (2017) reported a final concentration of $26.2 \text{ g}\cdot\text{N}\cdot\text{L}^{-1}$ (CF of 4.5) in the cathode during the recovery of TAN from synthetic urine by a novel MEC, while Kuntke et al. (2014) achieved a concentration of $7 \text{ g}\cdot\text{N}\cdot\text{L}^{-1}$ when concentrating TAN in an MEC (CF of 10). A convenient aspect of (B)ECs is the reduction of water at the cathode side, resulting in the generation of OH^- , allowing for an in-situ pH increase while no chemicals are needed. Hence, the concentrated TAN solution produced by (B)ECs contains dissolved NH_3 . Interestingly, no limitations by fouling were reported in the reviews of Kuntke et al. (2018) and Rodríguez Arredondo et al. (2015) while urine, (pig) digestate, reject water leachate were used as feed streams. The apparent tolerance of (B) ECs to blockage by solids and fouling is possibly explained by the relatively wide anode compartments and the fact that the feed water is not pressurised and forced through the membrane.

4.1.3. (Bipolar membrane) electro dialysis

Similar as in (B)ECs, TAN is transported as NH_4^+ from the residual water when an electric current is applied in electro dialysis (ED). However, because ED contains alternating cation and anion exchange membranes, alternating feed water and concentrate channels are formed. Eventually, the transported NH_4^+ ends up as concentrated TAN in the ED concentrate. Pronk et al. (2006a) applied ED to concentrate 93% of the TAN from source-separated urine and achieved a final concentration of $14.2 \text{ g}\cdot\text{L}^{-1}$ (CF of 2.9). Studies performed by Mondor et al. (2008) and Ippersiel et al. (2012) showed that ED can be used to remove NH_4^+ by $75 - 85\%$ from filtered manure reject water and that final TAN concentrations between 14 and $21 \text{ g}\cdot\text{L}^{-1}$ can be achieved (CF up to 5.6) for an energy consumption ranging $66 - 71 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$. Furthermore, Ward et al. (2018) achieved a CF of 8.5 in the ED concentrate of 120 L by removing 23% of the NH_4^+ from 5400 L sludge reject water at an energy consumption of $18 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$, leading to a final TAN concentration of $7 \text{ g}\cdot\text{L}^{-1}$. Furthermore, by optimising the applied current density (which will minimise osmotic water transport and ion back-diffusion), the TAN concentration can be increased from 1.5 to $10 \text{ g}\cdot\text{L}^{-1}$ (CF of 6.7) for 90% TAN removal at energy consumption of $5 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$ (van Linden et al., 2019).

By using bipolar membrane electro dialysis (BPMED), TAN can be transported from the residual stream as NH_4^+ and simultaneously be concentrated as dissolved NH_3 due to the production of OH^- by bipolar membranes, which only requires electricity (van Linden et al., 2020). According to the study of van Linden et al. (2020), at least 85% TAN removal can be achieved by BPMED for the production of $5 \text{ g}\cdot\text{L}^{-1}$ of NH_3 at the expense of $19 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$. Dissolved NH_3 concentrations of 46 and $54 \text{ g}\cdot\text{NH}_3\cdot\text{L}^{-1}$ starting from synthetic NH_4Cl and NH_4NO_3 solutions containing 37 and $45 \text{ g}\cdot\text{NH}_4^+\cdot\text{L}^{-1}$, respectively, were achieved by Li et al. (2016) and Gain et al. (2002), respectively. A study performed by Pronk et al. (2006b) resulted in the production of a solution containing $2.5 \text{ g}\cdot\text{NH}_3\cdot\text{L}^{-1}$ after treating diluted urine with an initial TAN

Table 2

An overview of the various technologies that can be used to concentrate or recover TAN from residual waters.

	Technology	TAN Feed Conc.	Required Input	Solids Tolerance	End products	TAN Product Conc.	Energy Consumption	Reference
		$\text{g}\cdot\text{L}^{-1}$	–	–	–	m% TAN	$\text{MJ}\cdot\text{kg}\cdot\text{N}^{-1}$	
Technologies to concentrate TAN	RO	0.9–8.5	Electricity	Low	NH_4^+ (aq)	1.3	n.r.	(Mondor et al., 2008; Schoeman and Strachan, 2009a)
	FO	1.3	Elec. + salt solution	Low	NH_4^+ (aq)	0.4	n.r.	Holloway et al. (2007)
	(B)ES ED	0.7–5.8 0.8–4.9	Electricity Electricity	Low Low	NH_3 (aq) NH_4^+ (aq)	0.7–2.6 0.7–2.1	–10 - 94*	Kuntke et al. (2018) (Ippersiel et al., 2012; Mondor et al., 2008; Ward et al., 2018)
Technologies to recover TAN	BP MED	4.9–45	Electricity	Low	NH_3 (aq)	0.5–1.4	58	Shi et al. (2018)
	Precipitation	n.r.	Base and salt	High	$\text{MgNH}_4\text{PO}_4\cdot(6\text{H}_2\text{O})$ (s)	7	59	Magrí et al. (2013)
	AS (+Scrub)	0.5–6.7	Electricity, heat and base (acid)	High	NH_3 -air (g) (NH_4^+) (aq)	0.9 (7–14)	14–50	(Mehta et al., 2015b; Zarebska et al., 2015)
	V(M)S	1–12	Electricity, heat and base	High	NH_3 -water (g)	1.0–6.8	215	Beckinghausen et al. (2020)

n.r. – not reported; * - negative energy consumption means energy production.

concentration of $4.9 \text{ g}\cdot\text{L}^{-1}$ by BP MED, while Shi et al. (2018) used BP MED to completely remove NH_4^+ from synthetic pig manure reject water at the expense of $58 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$, reaching a final concentration of $13.8 \text{ g}\cdot\text{NH}_3\cdot\text{L}^{-1}$.

Similar to FO and RO, feed waters with low solids concentrations are desired for ED and BP MED, to avoid particulate fouling between the spacers and membranes. Besides, in available studies on ED to concentrate TAN, organic fouling and scaling on the membranes was reported (Mondor et al., 2009; Shi et al., 2019). Fouling in ED can be reversed and limited by chemical cleaning, reversing the electrode polarity (Shi et al., 2019) or by avoiding the transport of scaling substances (multivalent ions) and humic acids by using selective membranes (Kim et al., 2002). In the few published studies on BP MED to recover TAN from residual streams, no information on fouling was found.

4.2. Technologies to recover TAN

4.2.1. Struvite precipitation

The addition of magnesium to residual waters containing both TAN and phosphate within the optimum pH range ($\text{pH} = 8 - 9$) leads to the precipitation of struvite crystals ($\text{MgNH}_4\text{PO}_4\cdot 6\text{H}_2\text{O}$, having an NH_4^+ content of 7 m%), which can be used as fertilizer (Mehta et al., 2015a; Zarebska et al., 2015). Struvite precipitation is widely applied to avoid undesired scaling in pipelines during the transport of digestate and for the recovery of phosphorus. Moreover, struvite formation can directly be achieved in manure reject water, suggesting that struvite precipitation has a high tolerance to the presence of solids in the residual water (Mehta et al., 2015a; Zarebska et al., 2015). However, in N-loaded residual waters, TAN is present in excess molar concentrations with respect to phosphate (equal molar concentrations required to form struvite) resulting in a TAN removal efficiency of struvite precipitation limited to 15 – 30% (Mehta et al., 2015a; Zarebska et al., 2015). The energy consumption of chemical precipitation for the removal and recovery of TAN was reported by Magrí et al. (2013) to be $59 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$, taking the use of chemicals into account.

4.2.2. (Air) stripping and acid scrubbing

TAN can also be recovered as NH_3 by air stripping (AS), for example from manure and sludge reject water (Magrí et al., 2013; Zarebska et al., 2015) and recovery of TAN from cathode solutions produced by (B)ECs (Kuntke et al., 2018). Because the vapour pressure of NH_3 in fresh air is negligibly low, NH_3 transport from the residual water to the air takes place. TAN recovery by AS in stripping towers has a high tolerance of solids as studies reported that no pre-treatment of digestate was required

(Mehta et al., 2015a; Zarebska et al., 2015), while it should be noted that accumulation and scaling of minerals requires cleaning. However, before NH_3 effectively can be stripped, the pH of the residual water must be increased to convert NH_4^+ to NH_3 , by means of chemical addition, CO_2 stripping or electrochemical reactions (water reduction or water dissociation).

The actual concentrations of NH_3 in the air after NH_3 stripping according to the study of Wang et al. (2010) are below 9000 ppm (corresponding to 0.9 m%). Besides, based on the reported NH_3 mass flows and the used air flow rates, the concentration of NH_3 in the air is well below 1 m% according to the studies of Bonmati and Flotats (2003) and Lei et al. (2007). Hence, by using AS, only diluted gaseous NH_3 is obtained. By subsequent scrubbing of the NH_3 gas-containing air with acid, dissolved NH_4^+ solutions or even solid NH_4^+ salts such as ammonium sulphate ($(\text{NH}_4)_2\text{SO}_4$), ammonium bicarbonate (NH_4HCO_3) or ammonium nitrate (NH_4NO_3) can be obtained (Bonmati and Flotats, 2003; Kuntke et al., 2018; Ukwuani and Tao, 2016). The energy consumption of AS and subsequent scrubbing in acid ranges from 14 to $50 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$ and depends strongly on the TAN concentration and temperature of the residual water (Mehta et al., 2015a; Zarebska et al., 2015). However, not all reported values consistently consider the energy for the addition of heat and the addition of chemicals.

By using hydrophobic membranes, which are impermeable for liquids, but permeable for vapours and gases, to separate the liquid and gas phase, small installation footprints can be realised by providing a large contact area per unit of volume between the feed water and the permeate. Moreover, the pressure of the liquid can be controlled independently from the pressure of the gas. When an acidic solution is recirculated in the permeate side and the feed water contains dissolved NH_3 , stripping and direct scrubbing takes place, resulting in the direct production of solution containing NH_4^+ . This configuration of NH_3 stripping and scrubbing is called direct membrane contactors (DMCS) or transmembrane chemisorption (TMCS).

According to the review studies of Zarebska et al. (2015), Mehta et al. (2015a) and Kuntke et al. (2018), the DMCS or TMCS was widely applied to directly scrub the stripped NH_3 from digestates, reject waters, stored urine and from the cathode compartment solutions from (B)ECs, even though TSS concentrations of up to $20 \text{ g}\cdot\text{L}^{-1}$ were present. The review of Beckinghausen et al. (2020) reported that the energy consumption was about $4 \text{ MJ}\cdot\text{kg}\cdot\text{N}^{-1}$, but it remains unclear whether this includes the use of heat and chemicals such as H_2SO_4 .

4.2.3. Vacuum (membrane) stripping

Finally, stripping of NH_3 also can be achieved by applying vacuum,

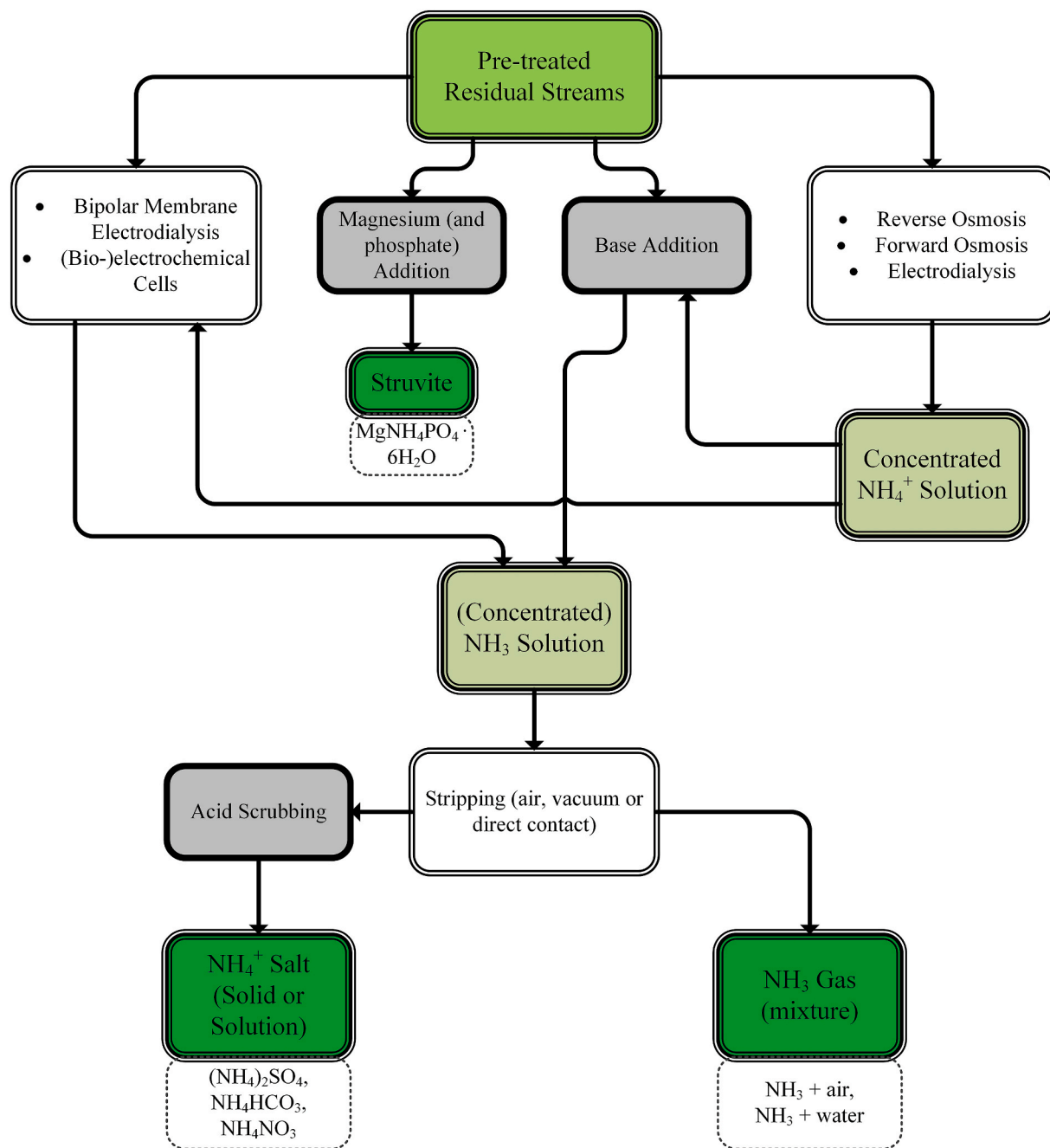


Fig. 6. A schematic overview of the various TAN recovery technologies to obtain various TAN products, such as concentrated NH_4^+ solutions, NH_3 solutions, struvite, solid or dissolved NH_4^+ salts and gaseous NH_3 .

which avoids the presence of air in the vapour that contains the stripped NH_3 . Ukwuani and Tao (2016) successfully used vacuum stripping (VS) in combination with acid scrubbing to recover NH_3 from water at various feed water temperatures and vacuum pressures from manure, food waste, sludge digestate and landfill leachate (containing 1.0 – 6.4 $\text{g}\cdot\text{L}^{-1}$ of NH_3). According to the review of Beckinghausen et al. (2020), the required energy for TAN recovery by VS was 215 $\text{MJ}\cdot\text{kg-N}^{-1}$, which is mainly required for increasing the feed water temperature. However, besides the stripping of NH_3 , also water is evaporated during VS and vacuum membrane stripping (VMS) resulting in a gaseous NH_3 and water vapour mixture (He et al., 2018). In fact, the ratio of the NH_3 flux to the total flux (water and NH_3) during VMS to recover at NH_3 at TAN feed concentrations ranging 1 – 4 $\text{g}\cdot\text{L}^{-1}$ was only 1% (NH_3 concentration of 1 m%) in unfiltered digestate in the studies of He et al. (2018);

(2017a). However, according to the study of El-Bourawi et al. (2007), the NH_3 in the recovered gas increases from 1.2 m% to 6.8 m% when the concentration of NH_3 in the liquid feed is increased from 4.9 to 20 $\text{g}\cdot\text{NH}_3\cdot\text{L}^{-1}$, respectively.

4.3. Discussion on TAN concentration and recovery technologies

Table 2 presents an overview of all discussed TAN recovery technologies, including the tolerance to the presence of solids in the feed/source water, the required inputs (i.e., energy, chemicals, etc.), the obtained outputs (i.e., TAN chemical form, concentration, etc.). In addition, Fig. 6 presents an overview of the applications of the various TAN recovery technologies to obtain the various TAN products. The most suitable approach to recover TAN is very location-specific, because

the local situation determines the characteristics of the residual stream, discharge restrictions, the available resources (energy, financial, etc.) and the possibilities for reuse/recovery. Therefore, this review presents the technology sequence in an objective manner by only providing the information for decision making, which relates to input, product, and qualities, as shown in Table 2.

For almost all technologies (except for struvite precipitation), the actual energy consumption depends heavily on the feed water characteristics, the operational conditions and the actual performance. In current available literature, the energy consumption for RO and FO is not directly reported. Hence, to concentrate TAN by RO and FO, high TAN rejections and water recoveries must be achieved, leading to an increase in osmotic pressure throughout the operation, which will ultimately translate to a higher energy consumption. Also, the required information to determine the energy consumption to concentrate TAN is lacking. Therefore, there is a need to assess and normalise the energy consumption to concentrate TAN (in $\text{MJ}\cdot\text{kg}\cdot\text{N}^{-1}$) by RO and FO, which will be a function of the TAN feed concentration and rejection, the water recovery and flow rate, transmembrane membrane pressure and pump efficiency. The same holds for (B)ECs and (BPM)ED, for which the energy consumption to concentrate TAN strongly depends on the feed concentration, the amount of TAN transported, the efficiency of using electric charge and the resistance of the cell and membrane stacks. When sufficient data is available, there is potential to normalise the data and derive technology-specific energy values to concentrate TAN. Eventually, the normalisation of the required energy input for the recovery of TAN can be based on the respective feed concentration and the removal or recovery efficiency.

Finally, to actually recover TAN as gaseous NH_3 , NH_4^+ solution or solid NH_4^+ crystals, the energy consumption must be expressed including the required amount of heat and energy to produce chemicals and to increase the pH and scrub the NH_3 . Only when normalised information is available on the various strategies and technologies to recover TAN, fair comparisons between technologies can be made. Eventually, the choice to use a certain technology or combination of technologies will depend on the availability of local resources, the potential to use the recovered products and the financial implications.

5. Use of the recovered TAN

5.1. N-fertilizer production

Among the plant nutrients, N is the most important one for crop yield and plant growth and accounts for the majority (> 50%) of the total crop fertilizer consumption. N-fertilizers can be produced in various chemical forms such as: urea, $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , and urea ammonium nitrate. A higher crop growth rate is forecasted for urea, since it is has the highest N content (~46 m% of N) (Chien et al., 2011). The potentially recovered TAN can be used to produce any of these N-fertilizers, shortcutting the artificial N-cycle and partially meeting the fertilizer demand.

Although there is an increasing interest in the production of bio-based fertilizers in the emerging circular economy (Sigurnjak et al., 2016), there is also a lack of information about the resulting end-products and their performance. In fact, the European fertilizer legislation (Regulation (EC) No. 2003/2003) still identifies these products as animal manure, hindering their market potential. One of the few and most recent studies on the production and testing of ammonia-based fertilizers from agricultural waste is that of Sigurnjak et al. (2016), where the authors found no performance differences between the bio-based produced NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ and the synthetic fertilizers when applied in crops of *Lactuca sativa* L. However, a high N-content variability for bio-based $(\text{NH}_4)_2\text{SO}_4$ (30–86 $\text{g}\cdot\text{N}\cdot\text{kg}^{-1}$) and NH_4NO_3 (132–198 $\text{g}\cdot\text{N}\cdot\text{kg}^{-1}$) recovered at different installations was found. Similar conclusions were reached by Szymańska et al. (2019) in a very similar study for maize and grass. The work of Sigurnjak et al. (2016) is

part of the EU funded project SYSTEMIC that aims at recovery and recycle of valuable mineral components from organic waste streams into new products including fertilizers at large scale. During the project, extensive characterisation and performance assessment was done for the end-products ($(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3) of several ammonia stripping installations from anaerobic digesters in Europe.

The most common approach for production of fertilizers from recovered TAN is the stripping-scrubbing approach, where either $(\text{NH}_4)_2\text{SO}_4$ or NH_4NO_3 are produced. Both end-products have a high N-concentration, yet, most implemented stripping units produce $(\text{NH}_4)_2\text{SO}_4$. Worldwide, $(\text{NH}_4)_2\text{SO}_4$ represents only 4% of the N-fertilizer production because of its relatively low N-concentration as compared to urea (21 m% and 45 m%, respectively) (Vaneckhaute et al., 2017). However, the demand for $(\text{NH}_4)_2\text{SO}_4$ has raised recently because of an increased necessity for S-nutrition, due to that better air quality results in less S deposition on agricultural land (Chien et al., 2011). Typically, commercial ammonia stripping systems can achieve $(\text{NH}_4)_2\text{SO}_4$ concentrations between 25 and 40 m% (equivalent to 6 – 10% of N), which is often marketed as a liquid fertilizer (Vaneckhaute et al., 2017). Although granular (> 1.8 mm crystals) $(\text{NH}_4)_2\text{SO}_4$ is preferred because of its higher market price, the techno-economic feasibility of producing it from residual water requires more investigation (Hofmann et al., 2009; Vaneckhaute et al., 2017). According to the former European regulation (Regulation (EU) 2003/2003), fertilizers should have a minimum N-concentration of 15 m%, which is more easily reachable by NH_4NO_3 due to the additional presence of N as NO_3^- . In the new EU regulation for fertilizer products (Regulation (EU) 2019/1009) lower N-concentration concentrations (1.5 – 3 m%) are required for liquid inorganic macronutrient fertilisers, which can easily be produced with $(\text{NH}_4)_2\text{SO}_4$ solutions. In any case, if both $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 are obtained from residual streams, their use in crops is limited and regulated by the current European nitrates' directive (Directive 91/676/EEC, 1991).

Besides the recovery of $(\text{NH}_4)_2\text{SO}_4$, TAN is also recovered in full-scale applications as struvite, which can be used as a slow-release fertilizer. However, the struvite precipitation only recovers a limited fraction of TAN in the residual streams due to the off-balance in molar concentrations of TAN and phosphate (as discussed in 4.2.1). Regarding urea, no literature or projects have been found, probably because of the technical difficulties for its synthesis entailing high temperatures (i.e., 160–180 °C) and pressures (i.e., 110 atm). A very interesting work is presented by He et al. (2017b), who described ammonia water recovery from biogas digestate via VMS, which is subsequently used for the upgrade of biogas (i.e., CO_2 capture). The by-product of the process is aqueous NH_4HCO_3 , an inexpensive N-fertilizer that is commonly used as a substitute of urea in China.

Finally, an important aspect for the commercialisation of bio-based fertilizers is the presence of impurities and their potential long-term environmental impact. Impurities caused by organics, metals, pathogens and other contaminants may occur and must be, therefore, assessed (Vaneckhaute et al., 2017). To this end, Laurenzi et al. (2013) assessed the influence of the COD content of the feed streams (pig manure and reject water) on the organic contamination (TOC and VOCs) of the recovered $(\text{NH}_4)_2\text{SO}_4$ via ammonia stripping. The degree of impurity depended greatly on the initial organic matter content of the streams and to some extent on the pH used to strip NH_3 . In general, reject water (i.e., low COD) and high pH is preferred. The authors also propose the use of basic traps downstream to increase the organics abatement. Besides, a few researchers investigated the properties of recovered struvite (Rahman et al., 2014) and $(\text{NH}_4)_2\text{SO}_4$ (El Diwani et al., 2007; Möller and Müller, 2012; Vaneckhaute et al., 2014) as fertilizers.

5.2. Energy carrier

NH_3 was recently identified as a suitable energy carrier, as an alternative to carbon-based fuels such as oil and natural gas, because it is

Table 3

Various direct NH₃ fuel cells and their operational characteristics and peak power densities according to the review studies of (Afif et al., 2016); Lan and Tao (2014) (Ni et al., 2009);

Type	Operating Temperature	Electrolyte	Mobile ion	Peak Power Density
AMFC	25 °C	Anion Exchange Membrane	OH ⁻	16 mW·cm ⁻²
AFC	50–450 °C	Dissolved/molten OH ⁻	OH ⁻	40 mW·cm ⁻²
SOFC-H	450–750 °C	Ceramic Membrane	H ⁺	580 mW·cm ⁻²
SOFC-O	500–1000 °C	Ceramic Membrane	O ²⁻	1190 mW·cm ⁻²

carbon-free and the storage and transportation systems are already established (Valera-Medina et al., 2018). Therefore, NH₃ can also be used as a fuel to produce both heat and power. Based on collected data, a general overview of direct NH₃ fuel cells and their operational characteristics and peak power densities is provided in Table 3 (Afif et al., 2016; Lan and Tao, 2014; Ni et al., 2009).

5.2.1. Combustion technologies

NH₃ can be used as fuel in thermal combustion and propulsion technologies. This is normally done in combination with other fuels, such as H₂, CH₄ or other carbon-based fuels (Valera-Medina et al., 2018). The inherent efficiency of combustion-based technologies is limited. Moreover, according to the review studies of Kobayashi et al. (2019) and Dimitriou and Javaid (2020), there are additional challenges when using NH₃ regarding low flammability, emission of unprocessed NH₃ and oxidised nitrogen species (NO_x, N₂O, etc.). No studies on the use of recovered NH₃ in energy generation with combustion-based technologies could be found.

Recent developments in the maritime shipping industry are focusing on using ammonia in internal combustion engines (Lesmana et al., 2019). In fact, ammonia is regarded as a key carbon neutral player, particularly in the retrofitting of the existing fleet. According to a recent market outlook (DNV-GL, 2020) in order to achieve the UN-IMO (International Maritime Association) targets, ammonia (dual-fuel) internal combustion engines could have a share between 30 and 60% of the total maritime fuel mix by 2050.

5.2.2. Alkaline (membrane) fuel cells

Alkaline fuel cells (AFCs) use dissolved or molten hydroxide for the transport of hydroxide (OH⁻) from the cathode to the anode, whereas alkaline membrane fuel cells (AMFCs) use an anion exchange membrane. In both AFCs and AMFCs, NH₃ is directly electrochemically oxidised by OH⁻ at the anode, while at the cathode O₂ and water react together with the supplied electrons to OH⁻ (Lan and Tao, 2014). However, Lan and Tao (2010) mentioned that the low operational temperature of 25 °C resulted in a long stabilisation time of the established electric potential difference between the anode and cathode of their AMFC, indicating slow kinetics of the processes. Furthermore, research conducted by Suzuki et al. (2012) showed that the performance of AMFCs is limited by fuel cross-over, caused by diffusion of NH₃ from the anode to the cathode. Moreover, Suzuki et al. (2012) also showed that poisoning of the metal catalysts with adsorbed N species takes place at the anode. Finally, the reported maximum power densities for AFCs and AMFCs are only 40 and 16 mW·cm⁻², respectively, (Ganley, 2008; Lan and Tao, 2010), which is an order of magnitude lower than high temperature fuel cells.

5.2.3. Solid oxide fuel cells

SOFCs can be divided, based on their ability, to either conduct protons (SOFC-H) or oxygen ions (SOFC-O) through the solid ceramic electrolyte. Because in both SOFC types the operational temperature is

well above 500 °C, NH₃ is spontaneously cracked at the anode in the presence of a nickel catalyst, resulting in the production of H₂ and N₂. Initially, nickel was used as a catalyst for H₂-fueled SOFCs (Mahato et al., 2015) and later also appeared to be a good catalyst to crack NH₃ (Fournier et al., 2006). In SOFC-Hs, the electrolyte is proton-conducting, while in SOFC-Os, the electrolyte is oxygen-conducting, implying different reactions to take place. According to the study of Ni et al. (2008), the application of SOFC-Hs could lead to higher electrical efficiencies than SOFC-Os when using NH₃ as fuel due to the place where the oxidation reaction takes place, which affects the activity of the reactants. However, available literature reports higher power densities for SOFC-Os than for SOFC-Hs (Afif et al., 2016; Ni et al., 2009), which is mainly attributed to the low resistance of the oxygen-conducting electrolytes. A maximum power density of 1190 mW·cm⁻² for SOFC-Os was reported by Meng et al. (2007), compared to 580 mW·cm⁻² for SOFC-Hs (Aoki et al., 2018), both using NH₃ directly (without external cracking) as fuel. The higher power density reported for SOFC-O is probably a result of more intensive research activities. Based on review papers of (Ni et al., 2009) and (Afif et al., 2016), it can be concluded that also the design of the cell is important, as the use of anode-supported planar cells results in superior power densities, compared to the use of tubular-supported or tubular cells. Unfortunately, most research on NH₃-fueled SOFCs only report the achieved power density. In addition to the maximum power density, it is also important how efficient the fuel is used, to determine the actual electrical efficiency. Only a limited number of studies reported the actual electrical efficiency (i.e., conversion of chemical energy to electrical energy), which can go up to 70% (Dekker and Rietveld, 2006). A more general review paper on SOFCs by Stambouli and Traversa (2002) also reported that electrical efficiencies of 60% are feasible, while an additional 30% of the chemical energy from the fuels can be used as a high-grade heat. Therefore, SOFCs can potentially effectively use 90% of the total energy content of NH₃, making the SOFC the most efficient technology to reclaim energy from NH₃.

Another advantage of SOFCs over combustion-based technologies and A(M)FCs is the negligible production of oxidised N species (NO_x). Staniforth and Ormerod (2003), Ma et al. (2006) and Okanishi et al. (2017) analysed the anode off-gas, and concluded that the concentration of oxidised N-species is below the detection limit, and 0.5 ppm by Dekker and Rietveld (2006). Moreover, the application of SOFC-Hs using NH₃ as fuel will even less likely produce oxidised N species, because N₂ and O₂ will not be in direct contact, as only H⁺ is transported through the proton-conducting electrolyte (Ni et al., 2009). However, a potential challenge for the use of SOFCs is nickel nitridation, which is the formation of nickel-nitrogen (Ni₃N) species at the anode. Nitridation of nickel at the nickel/yttria-stabilised zirconia anode was observed by Yang et al. (2015), who linked this to a decrease in electric potential over the operational run time at an operational temperature ranging 600–700 °C. These findings were confirmed by Stoeckl et al. (2019b), who also observed a decrease in electric potential at 700 °C, while using a nickel/yttria-stabilised zirconia anode. However, interestingly, Stoeckl et al. (2019b) also observed a stable electric potential at an operational temperature of 800 °C. In a subsequent study of Stoeckl et al. (2020), the electric potential decreased only by 1% over an operational run time of 1000 h, when an operational temperature ranging 815–845 °C was maintained. The authors reported that no structural damage was observed at the anode. However, in this study, a nickel/gadolinium-doped ceria anode was used. Therefore, it remains unclear under what operational conditions and for what anode materials nickel nitridation threatens the stable operation of NH₃ in SOFC-Os. For SOFC-Hs, we found no studies reporting on nitridation or the production of oxidised N species.

To the best of our knowledge, there are no studies that use NH₃ actually recovered from residual streams as a fuel in a fuel cell. In case of recovery from residual water, NH₃ will be accompanied by water vapour and potentially other contaminants. Interestingly, the main components

of biogas (CH₄ and CO₂), can also be fed to the SOFC–Os, because after CH₄ reforming with steam or CO₂, the produced H₂ and CO also serve as fuel (Gür, 2016; Saadabadi et al., 2019). However, research conducted by Papadias et al. (2012) showed that SOFCs are especially sensitive to contaminants such as H₂S (typically present in biogas), HCl and siloxanes, which deactivate the nickel catalyst and decrease the effective surface of the anode, suggesting that gas cleaning is required before using recovered gases as fuels. Finally, studies on SOFC–Os conducted by Wojcik et al. (2003), Cinti et al. (2016), Stoeckl et al. (2019a) and Stoeckl et al. (2020) showed that it is actually feasible to use mixtures of NH₃ and water vapour as fuel for SOFC–Os. However, the minimum concentration of NH₃ in the fuel was 17% (Wojcik et al., 2003) and it remains unclear whether NH₃ in this concentration can (directly) be recovered from water. Therefore, more research is required to determine what concentrations of NH₃ can be realised when NH₃ is recovered as a gas from (residual) water and whether SOFCs can work with these concentrations.

5.3. Resource for chemical and biochemical processes

Besides its use as (resource for) fertilizers, NH₃ is also widely used as a refrigerant gas, for purification of water supplies, and in the manufacture of plastics, explosives, textiles, pesticides, dyes and other chemicals. Besides, NH₃ is found in household and many industrial-strength cleaning solutions. In general, NH₃ is the precursor of most of the N-containing compounds synthesized by the industry. Important derivatives of NH₃ are nitric acid (for the fabrication of explosives and fertilizers), hydrazine (used as foaming agent, for pharmaceuticals, insecticides and also as rocket fuel) and hydroxylamine (used in the preparation of oximes, precursors of nylon). Industrial NH₃ for chemical application is normally sold as NH₃ liquor or sometimes called ammonium hydroxide (an aqueous solution with a 25 – 33% concentration of NH₃) or as pressurised/refrigerated anhydrous liquid NH₃.

Low concentrated (4 – 28 m%) aqueous ammonia solutions are also used for CO₂ capture. Compared to the amine-based traditional capture solvents, aqueous ammonia is cheaper, not easily decomposed by other flue gases and less corrosive. However, aqueous ammonia has a lower CO₂ absorption rate and its higher vapour pressure leads to ammonia losses in the flue gas (Yang et al., 2014). Recent studies have proposed process modifications (e.g., Lean vapour compression, Rich solvent split, etc.) to overcome these drawbacks, showing promising results (Obek et al., 2019), even at pilot plant level (Yu et al., 2018). Additionally, selective reduction (Catalytic and Non-Catalytic) is a well-known chemical process for NO_x abatement in flue or exhaust gas streams in both vehicles and industrial processes. Numerous commercial products use an ammonia or a urea liquid solution as reducing agents. Ammonium sulphate solutions from recovered NH₃ also can be used for this purpose (Ellersdorfer, 2018; Lubensky et al., 2019).

Recently, the possibility of shortcutting the anthropogenic N cycle via direct production of edible proteins with NH₃ recovered from residual streams is being explored (Pikaar et al., 2018). The concept involves the production of high-value protein (single-cell protein) via biosynthesis, using lithotrophic H₂-oxidising bacteria in a reactor. For the bacteria to synthesise proteins, NH₃, H₂ and CO₂ are required as raw materials. According to Matassa et al. (2015), the physical footprint of such a system can surpass that of the plant-based one by several orders of magnitude. The authors theorised a potential yield, assuming wind energy-powered H₂ production at an average power of 2 W·m⁻² and a recovery of 1235 ton NH₃-N per year from Amsterdam-West WWTP, up to 3120 ton·Ha⁻¹·y⁻¹ of microbial protein, which would be 3 orders of magnitude more efficient than actual soy productivity (currently around 3 ton·Ha⁻¹·y⁻¹). The results of the first phase (proof-of-concept in a 5 L reactor operating in batch and continuous mode) showed H₂ conversion efficiencies ranging between 65 and 81% and protein concentrations between 78 and 375 g cell dry weight·m⁻³·h⁻¹ with very good nutritional properties comparable to those of high-quality fishmeal, showing

the potential of this concept (Matassa et al., 2015).

Finally, recovered aqueous NH₃ can also be used as biomass feedstock (e.g. wheat straw) pre-treatment prior AD in biogas plants (Wang et al., 2019), or to enhance the production of functionalised carbonaceous materials (biochar) via hydrothermal carbonisation (Latham et al., 2018).

5.4. Discussion on the use of recovered TAN

As above discussed, TAN is a very versatile commodity that has many applications. Moreover, all the mentioned applications have or will have important roles in the future CO₂-neutral and circular economy scenario. It seems logical to pursue closing the N-cycle by increasing the recovery of the TAN and eventually reducing the industrial NH₃ production. However, the amount and quality standards of the current and future ammonia applications are challenging.

In this review, two main challenges hindering the practical use of the recovered TAN were identified: 1) the maximum amount of TAN that the combination of the recovery technologies and the TAN concentrations in the identified sources can provide and 2) the quality and/or concentration of the recovered TAN. In marketable terms, fertilizers are the main share (i.e., 80%) as mentioned in previous sections (Galloway et al., 2008). However, there is an important lack of information regarding impurities/pollutant content and agronomic performance. Although the recovery technologies here mentioned showed very high selectivity towards TAN, most of the studies lack detailed chemical analysis and/or field trials. Parameters such as acidity and COD are likely of interest to the agronomic community, and other parameters such as heavy metal content (e.g., Cd, Cr, Hg, Ni, Pb, etc.), microbiological parameters (e.g., Salmonella, *E. coli*), etc. have maximum limits established by law. This is probably the reason why the legislators and the consumers are hesitant towards the application in the fields of fertilizers produced from recovered TAN. In fact, the current fertilizer regulation framework in the EU, although moving towards a circular economy scenario, shows some incongruences between the classification and applicability criteria and reduces the financial value of these products. Moreover, in most of the studies the final product was liquid, whereas a solid fertilizer is mostly preferred. A liquid product has much higher transportation and storage costs and may have stability problems over time.

If used as a fuel, the obtained NH₃ concentration is a key factor. The presence of water vapour or any other additional non-oxidizable gas stream will reduce the performance of the technology. Gas upgrading by reducing the water content needs to be explored. Additionally, for the fuel cells and in particular for the SOFC technology, certain chemical compounds such as H₂S might deactivate the catalysts (catalyst poisoning) and may necessitate gas cleaning before it enters the cell (Wasajja H. et al., 2021). In the case of the ammonia fueled combustion engines, NO_x formation and low performance related to the low ammonia combustion rate are the main challenges. The use of combustion promoters (i.e., a second fuel) or partial ammonia cracking prior the combustion are the approaches being explored for the latter challenge (Lesmana et al., 2019). Direct electrification will always be the most energy efficient option, however in the case of transportation and specifically in long-haul or freight trips, ammonia combustion engines can offer a better alternative.

The application of ammonia solutions to capture CO₂ and eventually produce ammonium carbonate or even urea is gaining interest, however, NH₃ slips in the flue gas and its relatively lower absorption capacity are the main development barriers. Additionally, the influence of other impurities or gases in the recovered TAN solutions is unknown and could be considerable, given the delicate thermodynamic equilibrium of the NH₃-CO₂-H₂O system.

The application of ammonia, urea or ammonium sulphate solutions to NO_x abatement is also very interesting but NH₃ slip is an important issue. The same issue applies regarding the unknown effects of other

Table 4

An overview of the various applications for recovered TAN, including the respective ammonia forms, concentrations, and main product challenges.

Application	TAN Form	Concentration Required	Challenges/opportunities	Reference
Fertilizer	NH ₄ ⁺ (aq.) or NH ₄ ⁺ (s) (urea, ammonium sulphate, struvite, etc.)	6–10 m% (15 m% required by EU regulation)	Possible presence of pollutants and lack of end product performance assesment Use in crops is limited and regulated by the European nitrates' directive Big market (Million ton-year ⁻¹)	(Vaneekhaute et al., 2017); (Regulation (EU) 2003/2003; Directive 91/676/EEC, 1991)
CO ₂ capture	NH ₃ (aq.)	4 - 18 g-N·L ⁻¹	Market dominated by amines Lower CO ₂ absorption capacity Ammonia gas leaks	He et al. (2017b)
Chemical industry (chemical precursor)	NH ₃ (aq.) NH ₃ (g.)	25–33 m%	High purity and concentration needed Big market (Million ton-year ⁻¹)	Mysoreammonia (2020); Steelmangas (2020)
Biomass pre-treatment processes (bio refineries)	NH ₃ (aq.)	0.7 m%	Small market	Wang et al. (2019)
NOx abatement (flue gas)	NH ₃ (aq.) NH ₄ ⁺ (aq.) urea or ammonium sulphate	20–32 m%	High purity and concentration needed Big market (ktons-year ⁻¹)	Lubensky et al. (2019)
Energy Generation	NH ₃ (-H ₂ O mixture) (g)	>17 m%	Possible presence of poisoning pollutants for fuel cells (S)	Wojcik et al. (2003)

impurities or gases present in the recovered TAN solutions, especially in the case of the catalytic selective reduction, where a solid catalyst is employed.

In order to gain the trust of the end-users and the authorities and advance towards a real circular economy, an effort from the scientific community towards a detailed and application-oriented characterisation of the quantity and the quality of the recovered TAN and its performance is needed. We have summarized in Table 4 the various applications discussed here, as well as the form of TAN, the (minimum) required concentrations for each application and their main challenges.

6. Conclusions

- In this review, residual streams with a minimum TKN of 0.5 g-N·L⁻¹ are identified as N-loaded streams and categorized by their TAN recovery potential. Three different categories are established according to the TAN/TKN ratio and TSS content. Category 1 as streams with TAN/TKN < 0.5, category 2 as streams with TAN/TKN ≥ 0.5 and TSS > 1 g·L⁻¹ and category 3 as streams with TAN/TKN ≥ 0.5 and TSS ≤ 1 g·L⁻¹. Category 1 streams usually need a conversion of organic N to TAN before TAN recovery, whereas category 2 streams require the removal of solids to enhance the TAN recovery efficiency and category 3 streams are suitable for direct TAN recovery.
- AD is an advantageous pre-treatment for TAN recovery especially for category 1 streams, because it transforms the organic N into TAN while reducing COD and TSS. However, AD sometimes faces slow organic-N conversion rates and limited efficiencies. Incomplete hydrolysis and TAN inhibition are generally identified as the main causes. In general, further research is needed to identify methods for enhancing the organic-N conversion during AD.
- TAN recovery can be done via several technologies requiring different inputs (i.e., electricity, heat, chemicals, etc.), resulting in several end-products (i.e., TAN solutions, NH₃ gas, solid NH₄⁺ salts, etc.). Complete information about quality and TAN concentration of end-products, energy and chemical inputs and operational costs is limited and therefore reliable comparisons are difficult to make. Therefore, the energy consumption must be normalised to the respective TAN concentration in the feed and the removal or recovery efficiency.
- Various applications to use the recovered TAN have been identified and discussed in this review, including: N-fertilizer production, energy generation, industrial processes usage and some novel ones like microbial protein production. Two main factors constraining the reuse of TAN were identified: the actual TAN that can be recovered

from the residual streams and the quality of this recovered TAN. Especially in the case of N-fertilizer production, which use is linked to its quality, a lack of information on product quality and performance has been reported.

Credit author statement

Zhe Deng: Conceptualization, Investigation, Data curation, Writing-Original draft preparation. Niels van Linden: Conceptualization, Data curation, Visualization, Writing- Original draft preparation. Elena Guillen: Conceptualization, Writing - Review & Editing. Henri Spanjers: Conceptualization, Writing - Review & Editing, Supervision. Jules van Lier: Writing - Review & Editing, Supervision.

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

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.jenvman.2021.113096>.

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