

## Nitrogen recovery from reject water for improved sustainability of wastewater treatment

Hakan Jönsson <sup>a,\*</sup>, Andriy Malovanyy<sup>b</sup> and Susanne Tumlin <sup>c</sup>

<sup>a</sup> Department of Energy and Technology, Swedish University of Agricultural Sciences (SLU), P.O. Box 7032, SE-750 07 Uppsala, Sweden

<sup>b</sup> IVL Swedish Environmental Research Institute, Valhallavägen 81, SE-114 28 Stockholm, Sweden

<sup>c</sup> Gryaab AB, Norra Fågelrovägen 3, SE-418 34 Gothenburg, Sweden

\*Corresponding author. E-mail: hakan.jonsson@slu.se

 HJ, 0000-0002-8141-8171

### ABSTRACT

Flows of reactive nitrogen ( $N_r$ ) and greenhouse gas emissions from society are exceeding planetary boundaries, posing a serious risk to the stability of living conditions on Earth. Wastewater contains the largest flows of  $N_r$  in urban society, so recycling  $N_r$  from wastewater treatment plants (WWTPs) could reduce the climate impact and the need for new  $N_r$ . The reject water from dewatering anaerobically digested sludge contains high concentrations of  $N_r$  and recovery of this  $N_r$  would decrease the load on biological nitrogen removal processes, and thus nitrous oxide emissions. Simultaneously, the need for external carbon sources and energy for aeration will decrease. In a case study at Rya WWTP in Gothenburg, Sweden, three  $N_r$  recovery technologies were investigated: (1) conventional ammonia stripping to ammonium sulphate; (2) thermal stripping to ammonium sulphate and (3) distillation of ammonia from reject water to ammonia water. All three technologies were found to decrease the climate impact compared with the removal processes currently used at Rya WWTP for the removal of  $N_r$ . Recovery by distillation to ammonia water had the lowest climate impact, while conventional stripping minimised the energy requirement.

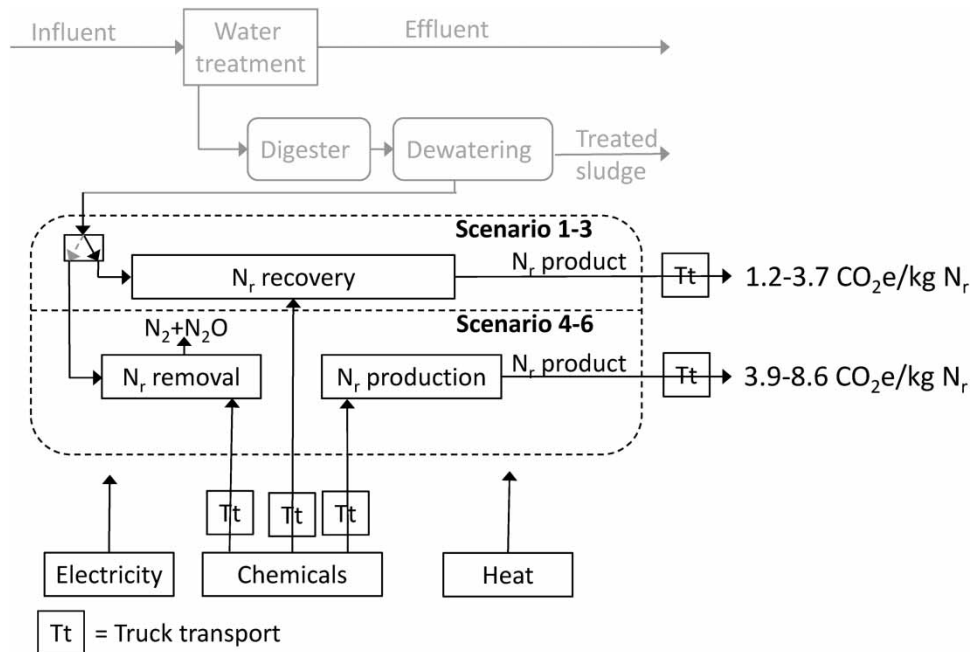
**Key words:** energy, greenhouse gas, nitrogen, recover, reject water, wastewater

### HIGHLIGHTS

- Recovery of reactive nitrogen ( $N_r$ ) from reject water in WWTP lowered GHG emissions.
- Recovery of  $N_r$  lowers GHG emissions also when fossil fuels have been phased out.
- The climate impact of recovery depended on emission factors from electricity and heat use.
- Recovery of  $N_r$  increased truck transport at the WWTP and in society.
- For Rya WWTP, recovery of  $N_r$  as ammonia water gave the lowest climate impact and WWTP transport.

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY 4.0), which permits copying, adaptation and redistribution, provided the original work is properly cited (<http://creativecommons.org/licenses/by/4.0/>).

## GRAPHICAL ABSTRACT



## INTRODUCTION

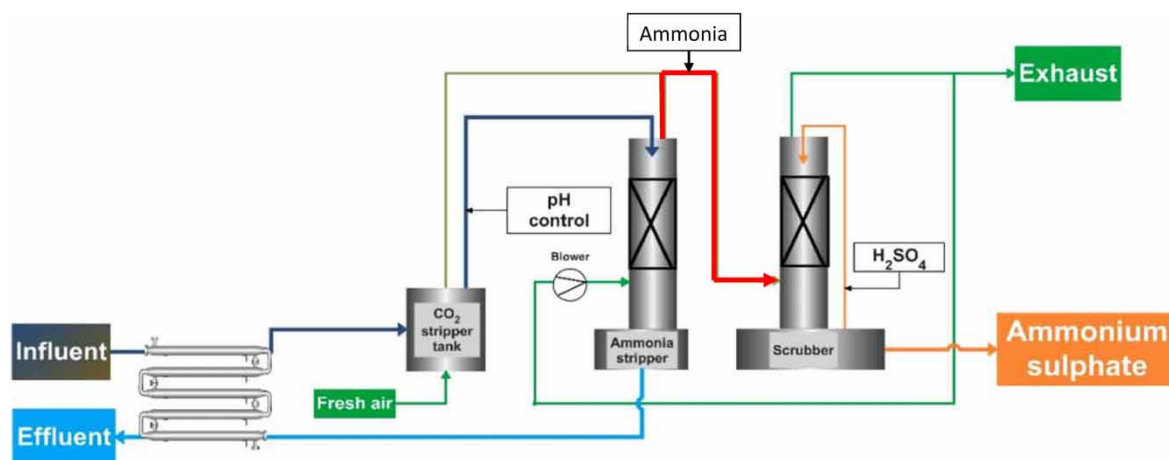
Flows of reactive nitrogen ( $N_r$ ) and greenhouse gas (GHG) emissions from society are exceeding planetary boundaries, posing a major risk to maintaining stable living conditions on Earth (Richardson *et al.* 2023). Wastewater contains the largest flow of  $N_r$  in urban society. In wastewater treatment plants (WWTPs) with high requirements for nitrogen removal, this  $N_r$  is largely removed by converting it into nitrogen gas ( $N_2$ ), usually by biological nitrification and denitrification or deammonification. Nitrous oxide ( $N_2O$ ) is an intermediate product of both nitrification, denitrification and deammonification. The biological conversion of  $N_r$  into  $N_2$  leads to some production of  $N_2O$ , and the amounts produced can be especially high when the system is not well balanced (Kanders *et al.* 2019). Since  $N_2O$  is a very potent GHG, these emissions often represent the single largest climate impact of WWTPs that are using low  $CO_2$ -emission electricity (Delre *et al.* 2019).

Nitrogen is the plant nutrient used in the largest amounts in crop production. Industrial production of  $N_r$  fertilisers by the Haber-Bosch process emits large amounts of GHGs, both  $N_2O$  and fossil carbon dioxide ( $CO_2$ ) (Brentrup & Pallière 2008). Recovering and recycling  $N_r$  from WWTPs has the potential to decrease total climate impact by lowering the  $N_2O$  emissions, the demand for external carbon sources and energy for aeration at the WWTP. Recovery of  $N_r$  can also decrease the need for the production of new  $N_r$  and thus the GHG emission from the production plants. In addition, recovery can increase the robustness of food production systems, as the recycled  $N_r$  product will be locally available.

Recovery of  $N_r$  from wastewater is augmented if the concentration of  $N_r$  is high and the wastewater is warm, as many recovery processes involve the conversion of ammonium ( $NH_4^+$ ) to dissolved ammonia ( $NH_3$ ) and transfer to the gas phase, both of which are facilitated by elevated temperature. In WWTPs, the digested sewage sludge and the reject water from sludge dewatering are warm (25–45 °C) and the concentration of  $N_r$  is high, often 800–1,300 mg  $N_r$ /L (Malovanyy *et al.* 2020). These properties are valuable for all three technologies for recovery of  $N_r$  compared in this study: conventional ammonia stripping, thermal stripping and distillation of ammonia water. These technologies were chosen based on the results of Malovanyy *et al.* (2020) and the Rya WWTP being interested in them. Since the  $N_r$  recovery depends on  $NH_3$  gas being stripped from reject water and then reacting to form an  $N_r$  product, only contaminants that evaporate can appear in the  $N_r$  product, resulting in a low contamination level. However, since the recovery of nitrogen is resource-intensive and the end product sometimes has a relatively low nitrogen content, it is important to consider the demand for truck transportation when assessing nitrogen recovery from wastewater, especially as WWTPs are often located close to the city centre and far from agricultural land.

### Conventional ammonia stripping

In conventional ammonia stripping,  $\text{NH}_3$  is driven off from the liquid phase in a scrubber where the liquid meets a gas (Figure 1). The  $\text{NH}_3$ -rich gas is transferred to a second scrubber, where it meets an acid, often sulphuric acid, that absorbs the  $\text{NH}_3$ , forming ammonium sulphate ( $(\text{NH}_4)_2\text{SO}_4$ ). To increase the efficiency of ammonia stripping, the pH of the liquid should be high (10–11) in order to convert most of the ammonium ( $\text{NH}_4^+$ ) into  $\text{NH}_3$ , which can be driven off in the first scrubber. Therefore, sodium hydroxide ( $\text{NaOH}$ ) is often added to increase the pH. Prior to this,  $\text{CO}_2$  is normally stripped from the liquid in order to decrease its buffering capacity and thus reduce the need for  $\text{NaOH}$  (Malovanyy *et al.* 2022). The temperature of the reject water is often high enough for running the process without any additional heating, especially if the retention time in buffer tanks is minimised. Conventional ammonia stripping without heating is used at some WWTPs, e.g. at Veas WWTP in Oslo (Eskicioglu *et al.* 2018). In most cases, an ammonium solution is produced and used as a fertiliser directly or for the production of mineral fertiliser (Stenström *et al.* 2017). Efforts have been made to produce crystalline ammonium sulphate directly in the scrubbing process (Andersson 2020).



**Figure 1** | General layout of a conventional ammonia stripping process (Changed after Menkveld & Broeders (2018)).

### Thermal ammonia stripping

Another way to strip ammonia from digested sewage sludge is by thermal stripping from digestate or reject water (Malovanyy *et al.* 2022). The substrate is then heated to about 80 °C in a large tank, during which both  $\text{NH}_3$  and  $\text{CO}_2$  are driven off without the need for pH adjustment. In one commercially available system, ANAStrip from GNS (Germany), the gases are led to a tank where the  $\text{NH}_3$  and  $\text{CO}_2$  react with gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ), forming a product consisting of dissolved  $(\text{NH}_4)_2\text{SO}_4$  (containing approximately 5% N) in a slurry of calcium carbonate ( $\text{CaCO}_3$ ). This process has been evaluated in a pilot study at a WWTP in Austria and installed at a number of organic waste digestion plants in Germany (Malovanyy *et al.* 2022).

### Distillation of ammonia

In the ammonia distillation process, reject water is evaporated using mechanical vapour recompression technology to about 5–30% of the original volume, but unlike in conventional evaporation, the pH is not decreased but left uncontrolled or increased slightly by adding a relatively small dose of  $\text{NaOH}$  (EPCON 2021). An elevated temperature of about 80 °C is used for evaporation, resulting in  $\text{NH}_3$  gas evaporating and being transferred together with the condensate. After further distillation steps, ammonia water containing around 16% N is produced (EPCON 2021). A few digestion plants in Finland use the process (Laanti & Karjala 2023). The concentrate left after evaporation, which is rich in other nutrients, can be used separately as a fertiliser.

### OBJECTIVE

The objective of this study was to estimate the consequences for society (climate impact, energy use and truck transport) of recovery of  $\text{N}_r$  in reject water from dewatering of digested sewage sludge. To fulfil this objective,

a case study was carried out at Rya WWTP in Gothenburg, Sweden. The hypothesis was that recovering and recycling  $N_r$  from reject water and hence lowering the demand for conventional  $N_r$  production in society, would lower the total climate impact compared with the current system of removing  $N_r$  by conversion to  $N_2$  emitted to the atmosphere, combined with conventional  $N_r$  production. The hypothesis was that energy use would decrease despite increased truck transport.

## METHODS

A life cycle assessment (LCA) inspired system analysis approach was used in the study, deviating from a full LCA in that only climate impact, energy use and truck transport were evaluated, that energy use and not primary energy were evaluated and that changes in infrastructure were not included. The functional unit was the removal of 90% of  $N_r$  in reject water at Rya WWTP during 2022 and the delivery of the same amount of  $N_r$  in a product to society. In  $N_r$  recovery scenarios, both parts of the functional unit were achieved by recovery of  $N_r$  from reject water. In  $N_r$  removal scenarios, the removal part of the functional unit was achieved by converting  $N_r$  in reject water into  $N_2$  (with some loss of  $N_2O$ ) in an  $N_r$  removal process, while the delivery of  $N_r$  product to society was achieved by industrial production of the same type and amount of  $N_r$  product as was produced in the recovery scenarios.

Emissions of GHG, energy use and mass of truck-transported chemicals required for  $N_r$  recovery or removal as well as for industrial production of  $N_r$  and the  $N_r$  product itself, were calculated for the different scenarios. The system boundaries for GHG emissions were cradle to  $N_r$  products and  $N_2$  leaving the gate of the WWTP or the  $N_r$  production site. In the energy analysis, the system boundaries included energy for the production of chemicals and energy needed at the WWTP, while energy for extraction of the input materials used for the production of the chemicals was not included. Emissions from, and energy for, the transport of chemicals and products were not included. The analysis was limited to the operational phase of the scenarios, i.e. construction of infrastructure was not included.

All scenarios studied were based on the layout and  $N_r$  removal processes at Rya WWTP, one of the largest WWTPs in Sweden (813,000 connected persons in 2022), located in Gothenburg. It uses an activated sludge process for the reduction of organic matter, nitrate and phosphate, followed by post-nitrification trickling filters and post-nitrification and post-denitrification moving bed biofilm reactors (MBBRs). The reject water is either treated in an MBBR-based deammonification process and then conveyed to the mainstream nitrogen removal process for polishing treatment, or pumped directly to the mainstream nitrogen removal process. Calculations were made mainly based on resource consumption for the nitrogen recovery technologies as summarised by Malovanyy *et al.* (2022), the Ecoinvent database and for removal scenarios measured data from Rya WWTP including its flow and composition of reject water as a yearly average during 2022 (Table 1).

**Table 1** | Measured<sup>a</sup> nitrous oxide ( $N_2O$ ) emissions, use of methanol and use of electricity in existing nitrogen removal processes at Rya WWTP, where during 2022, the total flow of reject water was 530,000 m<sup>3</sup> with a mean  $NH_4-N$  concentration of 0.864 g/L

Nitrogen removal process	No. of measurements; Date	Min kg $N_2O$ /kg $N^b$	Mean kg $N_2O$ /kg $N^b$	Max kg $N_2O$ /kg $N^b$	Methanol kg/kg $N^b$	Electricity kWh/kg $N^b$
Deammonification MBBR	2; 14 and 16 Jan 2020	0.0032	0.0062	0.0092	0	1.34
Post-nitrification trickling filter	4; 19 Feb and 28 Aug 2015	0.0077	0.0117	0.0180	0	5.41
Post-nitrification MBBR	3; 15–16 Jan 2020	0.0010	0.0014	0.0022	0	5.01
Post-denitrification MBBR	2; 30 June 2020	0.0001	0.0003	0.0004	3.16	0.59

<sup>a</sup>The  $N_2O$  emissions from deammonification and the MBBRs were measured using a floating chamber and from the trickling filter by upwind and downwind measurements using the tracer gas dispersion method.

<sup>b</sup>Per kg N transformed, i.e. for nitrification to  $NO_3^-$  and  $N_2O$ , for denitrification to  $N_2$  and  $N_2O$  and for deammonification to  $N_2$ ,  $N_2O$  and  $NO_3^-$ .

## SYSTEM AND SCENARIOS STUDIED

The system studied was the treatment of reject water from dewatering of mesophilic digested sewage sludge and a supply of  $N_r$  to society either by recovery of  $N_r$  from reject water or by industrial production of new  $N_r$ . The three recovery scenarios were:

- (1) Conventional NH<sub>3</sub> stripping, recycling the reject N<sub>r</sub> as solid (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (21% N).
- (2) Thermal stripping of NH<sub>3</sub>, recycling the reject N<sub>r</sub> as dissolved (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (5% N).
- (3) Distillation of NH<sub>3</sub>, recycling the reject N<sub>r</sub> as ammonia water (16% N).

The three scenarios with the removal of N<sub>r</sub> combined with the production of N<sub>r</sub> were:

- (4) Removal of N<sub>r</sub> in reject water by deammonification, combined with production of (a) (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or (b) NH<sub>3</sub>.
- (5) Removal of N<sub>r</sub> in reject water by post-nitrification in trickling filter and post-MBBR denitrification, combined with the production of (a) (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or (b) NH<sub>3</sub>.
- (6) Removal of N<sub>r</sub> in reject water by post nitrification and post-MBBR denitrification, combined with the production of (a) (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or (b) NH<sub>3</sub>.

It was assumed that 90% of total ammonium/ammonia nitrogen (TAN, NH<sub>4</sub><sup>+</sup>-N + NH<sub>3</sub>-N) in the reject water was recovered in the recovery scenarios (1–3) and that the same amount of TAN was released to the atmosphere, as nitrogen gas or nitrous oxide, in the removal scenarios (4–6). Thus, after recovery or removal, the reject water was assumed to be sufficiently similar between the scenarios to have a similar impact on the WWTP.

The calculated amounts of chemicals and energy used in the recovery processes are shown in Table 2. The calculations on the mass of chemicals and the mass of products needing truck transport were based on the amount of N<sub>r</sub> recycled and the corresponding mass of N<sub>r</sub> product, the amounts of methanol and other chemicals required (according to Tables 1 and 2) and the main chemicals needed for industrial production of N<sub>r</sub> product. The fossil natural gas used for industrial production of N<sub>r</sub> products was assumed not to require truck transport as the production units are connected to gas networks.

**Table 2** | Amounts of sodium hydroxide (NaOH), sulphuric acid (H<sub>2</sub>SO<sub>4</sub>), hydrochloric acid (HCl), gypsum (CaSO<sub>4</sub>·2 H<sub>2</sub>O), electricity and heat used in the three recovery scenarios (Malovanyy *et al.* 2022)

Nitrogen recovery	NaOH kg/kg N	H <sub>2</sub> SO <sub>4</sub> kg/kg N	HCl kg/kg N	Gypsum kg/kg N	Electricity kWh/kg N	Heat kWh/kg N
1. Conventional stripping	5.7	3.5	0.07	0	5.0	0
2. Thermal stripping	0	0	0	6.1	6.5	35
3. Ammonia distillation	1.5	0	0	0	30.0	0

During 2022, Rya WWTP used electricity from wind power emitting 0.011 kg CO<sub>2</sub>e/kWh (Energimyndigheten 2022) and heat from the Gothenburg district heating system emitting 0.065 kg CO<sub>2</sub>e/kWh (Göteborg Energi 2023). The energy required for the production of relevant chemicals (Tables 1 and 2) and the industrially produced N<sub>r</sub> products NH<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in scenarios (4–6), and the GHG emissions from production, are listed in Supplementary Table S1.

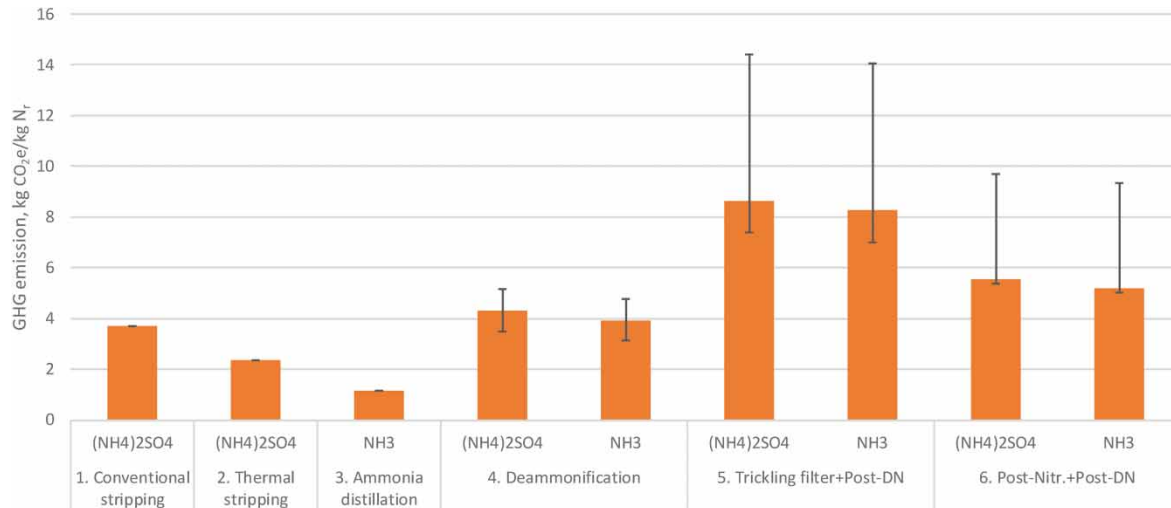
## RESULTS AND DISCUSSION

### GHG emissions

The difference in GHG emissions within each N<sub>r</sub> removal scenarios (4–6) between producing (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>3</sub> was small (Figure 2). For those scenarios, min, max and mean climate impact were calculated based on measured N<sub>2</sub>O emissions and carbon source for denitrification, where the min and mean impacts were calculated assuming the use of non-fossil methanol as the carbon source, while for the max impact use of methanol from fossil resources was assumed. The resulting min–max values were therefore asymmetrical in relation to the mean, especially for scenarios 5 and 6, which used more methanol. This should be borne in mind when comparing the recovery of N<sub>r</sub> to the use of only non-fossil methanol as different options for decreasing climate impact.

Moreover, measured N<sub>2</sub>O emissions from the N<sub>r</sub> removal processes at Rya were based on just a few (2–4) point measurements spread out over a very limited period (1 day to 6 months) (see Table 1). This means that the min–max bars in Figure 2 probably only represent part of the true range of climate impact from the processes during a year.

All N<sub>r</sub> recovery scenarios (1–3) emitted less GHGs than the mean of each removal scenario (4–6) (Figure 2), confirming the hypothesis. Among the N<sub>r</sub> removal scenarios, deammonification (scenario 4) had the lowest GHG



**Figure 2** | Greenhouse gas emissions (kg CO<sub>2</sub>e/kg N<sub>r</sub> treated) in scenarios 1–6. Mean value is represented by the coloured bar, while the min and max values are represented by the lines around the mean. Min and mean values in scenarios 4–6 were calculated assuming non-fossil methanol, while fossil methanol was assumed for the max values.

emissions, due to low N<sub>2</sub>O emissions from the deammonification process itself combined with low N<sub>2</sub>O emissions from the polishing treatment for removal of nitrate (NO<sub>3</sub><sup>-</sup>) from reject water in the post-denitrification MBBR (Table 1).

Removal of N<sub>r</sub> by post-nitrification and post-denitrification in MBBRs (scenario 6) gave lower GHG emissions than nitrification in trickling filters combined with MBBR post-denitrification (scenario 5) (Figure 2). This difference was due completely to greater N<sub>2</sub>O emissions for nitrification in the trickling filter, as the MBBR post-denitrification was the same in both scenarios. The main reason for this may be that the airflow and the area of the air–liquid interface are greater in the trickling filter than in the aerobic MBBR, allowing for greater mass transfer of N<sub>2</sub>O from the water phase in the trickling filter (Metcalf *et al.* 1991). Another possible reason is that process conditions are probably more uniform and better controlled in MBBR post-nitrification than in the trickling filter.

There were large differences between the three recovery technologies. Recovering N<sub>r</sub> as distilled NH<sub>3</sub> water (16% N) (scenario 3) gave by far the lowest GHG emissions (1.2 kg CO<sub>2</sub>e/kg N), less than one-third of the mean emissions from deammonification combined with production of ammonia (scenario 4b; 4.1 kg CO<sub>2</sub>e/kg N) (Figure 2). Thermal stripping (scenario 2) also gave low GHG emissions, around 55% of the mean climate impact from deammonification combined with the production of ammonium sulphate (scenario 4a). The climate impact of conventional stripping (scenario 1) was calculated to be 87% of that of deammonification with production of ammonium sulphate (scenario 4a).

Had N<sub>r</sub> been recovered from reject water at Rya via scenario 3, distillation to ammonia water, during 2022 the GHG emissions would have been 1,135 tons CO<sub>2</sub>e lower than N<sub>r</sub> removal via scenario 4 deammonification, 2,923 tons CO<sub>2</sub>e lower than N<sub>r</sub> removal via scenario 5 trickling filter and 1,656 tons CO<sub>2</sub>e lower than N<sub>r</sub> removal via scenario 6 post-nitrification and post-denitrification. These decreases represent 7, 19 and 11% decrease in the net GHG emissions from Rya during 2022 as reported in the environmental report (Videbris 2023). The distribution of N<sub>r</sub> removal in Rya during 2022 between the different removal scenarios (4, 5 and 6) is not known, thus the GHG decrease that would have resulted if the N<sub>r</sub> from reject water had been recovered can not be calculated.

In further analysis, it was assessed whether the GHG emissions would still be lower for the recovery scenarios (1–3) than for the removal scenarios if a carbon source without any GHG emissions was used, such as waste glycol from de-icing the wings of airplanes. When comparing the contribution of methanol use (Table 3) with the average values in Figure 2, which were calculated using non-fossil methanol, all recovery scenarios emitted less GHG than removal scenarios 4 and 5. However, only recovery scenarios 2 (thermal stripping) and 3 (ammonia distillation) emitted less GHG than scenario 6, while scenario 1 (conventional stripping) emitted slightly more GHG than scenario 6 (MBBR post-nitrification).



**Table 3** | Greenhouse gas (GHG) emissions from the use of non-fossil methanol used for calculation of min and average values in Figure 2 and fossil methanol used for calculation of max values for the different removal scenarios (4–6)

Emission/Scenario	Units	Methanol – non-fossil	Methanol – fossil
GHG emissions from:	kg CO <sub>2</sub> e/kg	0.76	1.98
4. Deammonification	kg CO <sub>2</sub> e/kg N <sub>r</sub>	0.03	0.08
5. Trickling filter	kg CO <sub>2</sub> e/kg N <sub>r</sub>	2.40	6.26
6. MBBR post-nitrification	kg CO <sub>2</sub> e/kg N <sub>r</sub>	2.40	6.26

In a conventional activated sludge system with pre-denitrification and no need for any external carbon source; recovery of N<sub>r</sub> from reject water will decrease the load on the N<sub>r</sub> removal processes, and thus might affect parameters such as recirculation flow, sludge retention time and biogas yield. Hence, it is difficult to fully identify implications for such WWTPs based on the results of this study.

The GHG emissions from electricity use at Rya WWTP in 2022 were low (0.011 kg CO<sub>2</sub>e/kWh) compared with those from the European Attribute Mix (0.531 kg CO<sub>2</sub>e/kWh) (AIB 2023). Had this mix been assumed, ammonia distillation (scenario 3) would have emitted most GHGs and deammonification (scenario 4) least, while GHG emissions from recovery scenarios 1 and 2 would still have been lower than those from removal scenarios 5 and 6. Further analysis of the sensitivity to GHG emissions from electricity showed that at 0.061 kg CO<sub>2</sub>e/kWh the lowest-emitting scenario changed from scenario 3, ammonia distillation, to scenario 2, thermal stripping. At 0.312 kg CO<sub>2</sub>e/kWh, the removal scenario 4 + NH<sub>3</sub> became the lowest-emitting one. The EU goal of climate neutrality (EU 2021) means that European electricity production needs to phase out fossil fuels and reach low levels of emissions, such as that of e.g. wind power which is what Rya WWTP used in 2022.

Emissions from heat energy were 0.065 CO<sub>2</sub>e/kWh (Göteborg Energi 2023). Of this, 0.044 CO<sub>2</sub>e/kWh were due to plastic and other fossil material in the waste used as fuel and 0.011 CO<sub>2</sub>e/kWh were due to fossil fuels used for district heating. The goal of climate neutrality (EU 2021) means that all fossils, i.e. both plastics and fossil fuels, should be eliminated. This would reduce emissions from district heating to 0.010 CO<sub>2</sub>e/kWh. In that case, thermal stripping (scenario 2) would have the lowest emissions, only 36% of those from ammonia distillation (scenario 3).

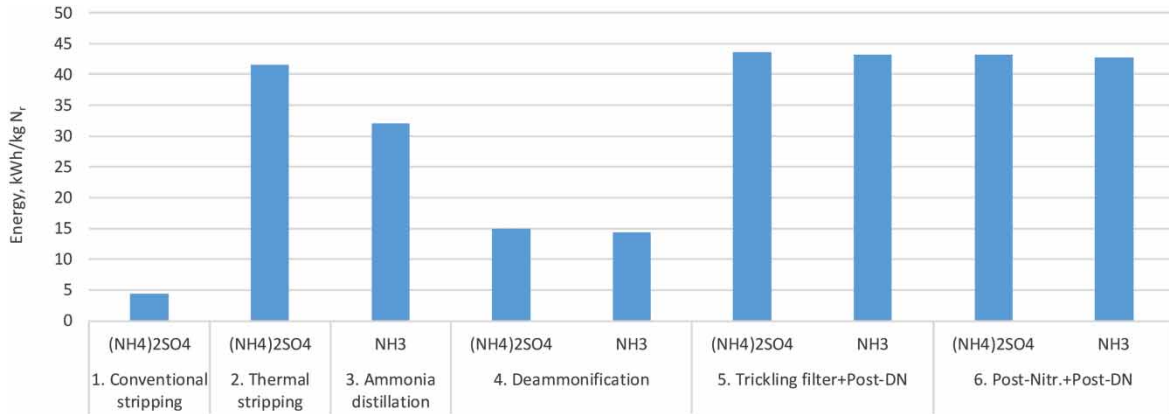
Also, fertiliser production needs to turn to renewable energy. According to Ahlgren *et al.* (2015), producing NH<sub>3</sub> with wind energy emits 0.12 kg CO<sub>2</sub>e/kg N<sub>r</sub>. In this future, the GHG emissions would still be lowest from scenario 3, ammonia distillation. However, the second lowest emission will be from 4b, deammonification. Scenarios 5a and 5b trickling filter nitrification and post-denitrification will have the highest emission. Thus, the emission from the N<sub>r</sub> production influences the results greatly. Conversely, in such a future society, not only fertiliser production but also the energy system and the production of NaOH and HCl will use renewable resources, which was assumed could lower the GHG emissions from these chemicals by 90%. In such a future, the three recovery scenarios will have the lowest emissions, with scenario 3, ammonia distillation, still having the lowest GHG emission. The three removing scenarios will all have larger emissions, with scenario 4 having the lowest in this group and scenario 5 trickling filter + post-denitrification having the highest GHG emission. Therefore, also in the future based on renewable energy, N<sub>r</sub> recovery in WWTP decreases the GHG emission.

### Energy use

Conventional stripping (scenario 1) used the smallest amount of energy for recycling N<sub>r</sub> (Figure 3). The reason is that the production of H<sub>2</sub>SO<sub>4</sub> from sulphur (S) is an exothermal process and the energy released when producing the H<sub>2</sub>SO<sub>4</sub> was credited to scenario 1, as this energy is usually recovered as industrial steam and used in other processes (Ecoinvent 3.9.1). The second-lowest energy use was seen for deammonification (scenario 4), due to this process being very energy-efficient in removing N<sub>r</sub>.

For the removal scenarios (4–6), the energy requirement for producing NH<sub>3</sub> was almost the same as that for producing (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (Figure 3). All recovery scenarios (1–3) used less energy than removal scenarios 5 and 6, in which all nitrogen was removed by post-nitrification and denitrification.

Recycling N<sub>r</sub> by ammonia distillation (scenario 3) required approximately twice as much energy as removing it by deammonification and producing new N<sub>r</sub> as (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or NH<sub>3</sub> (scenario 4a, 4b). However, the energy used



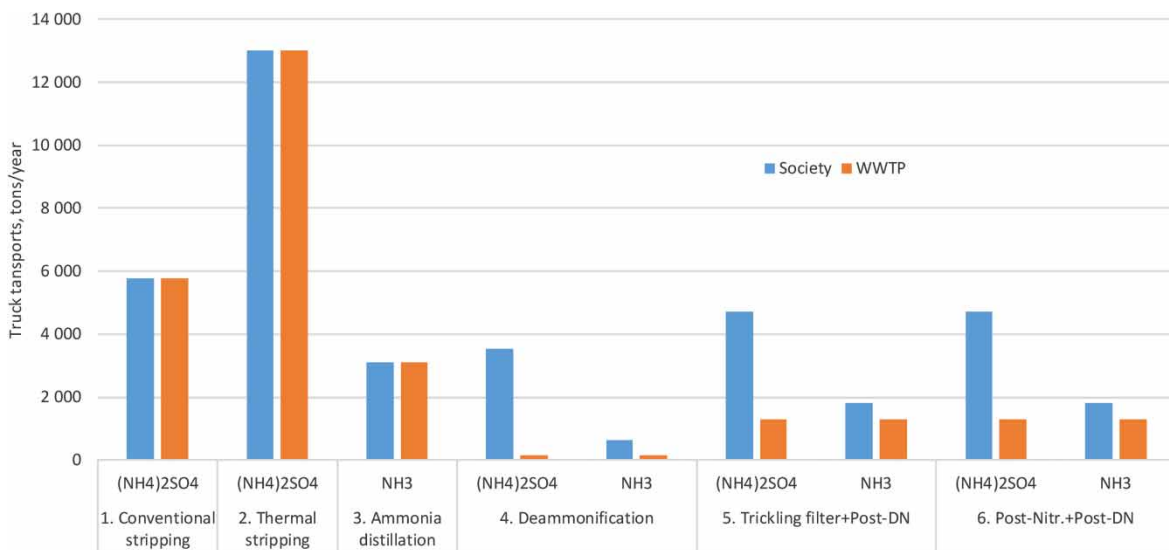
**Figure 3** | Use of energy for recycling reactive nitrogen (N<sub>r</sub>) (scenarios 1–3) or for removing and producing N<sub>r</sub> (scenarios 4–6), including producing the chemicals used by these processes.

in ammonia distillation was mainly electricity and can thus be renewable, while the energy used for the production of NH<sub>3</sub> was fossil natural gas.

**Transport of chemicals and products**

Deammonification combined with the production of NH<sub>3</sub> (scenario 4b) gave by far the lowest requirement for truck transport, both at Rya WWTP and in society. At the WWTP, the N<sub>r</sub>-relevant mass flow consisted only of 143 tons of methanol in that scenario. The flow in society was the methanol plus 500 tons/year of NH<sub>3</sub>, as the N<sub>r</sub> content in ammonia is very high (82%). Compared with the removal scenarios involving the production of NH<sub>3</sub> (4b, 5b, 6b), the flow in recovered ammonia water in scenario 3, ammonia distillation, was larger due to the lower N<sub>r</sub> content (16%, not 82% as in ammonia) and also because of use of 1.5 kg NaOH per kg N<sub>r</sub> recovered in the distillation process at the WWTP.

For all N<sub>r</sub> removal scenarios (4–6), there was a greater need for truck transport in society than at the WWTP, as the N<sub>r</sub> product was produced at a fertiliser factory and transported to a farm and the sulphuric acid needed for production of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> was truck-transported to the fertiliser factory. In these scenarios, only the carbon source was transported to the WWTP (Figure 4). The difference was far larger for the scenarios with (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (4a, 5a, 6a) than for those with NH<sub>3</sub> (4b, 5b, 6b), as the N concentration in ammonia (82%) is far larger than that in ammonium sulphate (21%) and since a large flow of H<sub>2</sub>SO<sub>4</sub> is needed as input material to produce ammonium



**Figure 4** | Flows of nitrogen-relevant chemicals and products for scenarios 1–6 in society (i.e. total flows including those to and from the WWTP) and the flows starting or ending at Rya wastewater treatment plant (WWTP).



sulphate. In the  $N_r$  recovery scenarios (1–3), the  $N_r$  product was produced at the WWTP and thus the need for truck transport was the same at the WWTP as in society (Figure 4). The need for truck transport was by far the largest in thermal stripping (scenario 2) since it used a lot of gypsum and produced a wet slurry containing low-strength ammonium sulphate and calcium carbonate.

Truck transport at the WWTP when recycling  $N_r$  (scenarios 1–3) was 3,100–13,000 tons, which was large especially when compared with the transport requirement for deammonification (scenario 4; 143 tons in 2022). Scenarios 1–3 recycled 412 tons  $N_r$ /year with low concentrations of pollutants. The added need for truck transport in the recovery scenarios can be compared with the need for transport of phosphorus-relevant chemicals and products (precipitants and sewage sludge) at Rya WWTP, which amounted to 55,100 tons in 2022. These transports led to the recycling of 424 tons of phosphorus, i.e. almost the same amount in tons as the amount of  $N_r$  that could be recycled as ammonia water with a much smaller need for transport of chemicals and products, 3,100 tons. Sludge also contains a lot of valuable organic substances and nitrogen, but the nitrogen present is mainly in organic form, which means that its availability to plants is limited (Delin *et al.* 2012), as is the immediate availability of phosphorus in sludge (Krogstad *et al.* 2005; Delin 2016), even though the long-term availability is sufficiently good to provide the requirements of cereals, but not potatoes (Jönsson 2019). Furthermore, sludge contains both organic and inorganic contaminants of concern. The N in recycled  $N_r$  is fully mineral and thus plant-available if correctly applied and the recycling process is substance-specific, meaning that the concentrations of contaminants are very low (Ghosh *et al.* 2019; Yee *et al.* 2019).

## CONCLUSIONS

- For Rya WWTP, where reject water is currently treated by deammonification and polishing post-denitrification, all three  $N_r$  recovery scenarios (conventional stripping, thermal stripping and distillation to ammonia water) decreased GHG emissions compared with the current system of removal of  $N_r$  and industrial production of  $N_r$  product.
- The three recovery scenarios will have lower GHG emissions than the removal scenarios also in a future society when fertiliser and chemical production and the energy system are based on renewable energy.
- Among the three removal scenarios studied, the removal of  $N_r$  via deammonification combined with the production of ammonia gave the lowest GHG emissions.
- With electricity emitting 0.312 kg CO<sub>2</sub>e/kWh recovery of  $N_r$  via distillation to ammonia water and removal via deammonification were calculated to have similar total GHG emissions.
- Recovery of  $N_r$  with distilled ammonia water emitted less than one-third of the emissions from deammonification combined with the production of ammonia, corresponding to 1,135 tons CO<sub>2</sub>e, or 7% less than the declared net GHG emission during 2022 from Rya WWTP.
- Recovery of  $N_r$  with distilled ammonia water emitted 1,656 tons CO<sub>2</sub>e less than post-nitrification and denitrification combined with the production of ammonia, corresponding to 11% less than the declared net GHG emission during 2022 from Rya WWTP.
- Recovery of  $N_r$  by conventional stripping had the lowest energy demand, followed by  $N_r$  removal via deammonification combined with the production of ammonia.
- All  $N_r$  recovery scenarios increased the need for truck transport of  $N_r$ -relevant chemicals, in society and especially at the WWTP, but the increase was small compared with other truck transport to and from the WWTP.

## ACKNOWLEDGEMENTS

This study was performed within the ordinary activities of the authors. H.J. worked on this paper during his retirement. A.M., IVL, and S.T., Gryaab, did their work within their employments. We are grateful to researcher Priscila De Moraes Lima, SLU for help with Ecoinvent and to Mary McFee for revising and improving the English language.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

## CONFLICT OF INTEREST

The authors declare there is no conflict.

## REFERENCES

- Ahlgren, S., Bauer, F. & Hulteberg, C. 2015 *Produktion av kvävegödsel baserad på förnybar energi – En översikt av teknik, miljöeffekter och ekonomi för några alternativ (Nitrogen Fertilizer Production Based on Renewable Energy – An Overview of Technology, Environmental Effects and Economics for Some Options)*. Rapport 82, Institutionen för energi och teknik, Sveriges Lantbruksuniversitet, Uppsala, Sweden.
- AIB 2023 *European Residual Mixes – Version 1.0 2023-06-01*. Association of Issuing Bodies. Available from: [https://www.aib-net.org/sites/default/files/assets/facts/residual-mix/2022/AIB\\_2022\\_Residual\\_Mix\\_Results\\_inclAnnex.pdf](https://www.aib-net.org/sites/default/files/assets/facts/residual-mix/2022/AIB_2022_Residual_Mix_Results_inclAnnex.pdf).
- Andersson, S. L. 2020 *Kväveåtervinning genom strippning och kristallisation (Nitrogen Recovery Through Stripping and Crystallization)*. Rapport B2412, IVL Svenska Miljöinstitutet, Stockholm, Sweden.
- Brentrup, F. & Pallière, C. 2008 Energy efficiency and greenhouse gas emissions in European nitrogen fertilizer production and use. In *International Fertiliser Society, Proceedings* 639.
- Delin, S. 2016 *Fertilizer value of phosphorus in different residues. Soil Use and Management* **32**(1), 17–26.
- Delin, S., Stenberg, B., Nyberg, A. & Brohede, L. 2012 *Potential methods for estimating nitrogen fertilizer value of organic residues. Soil Use and Management* **28**(3), 283–291.
- Delre, A., ten Hoeve, M. & Scheutz, C. 2019 *Site-specific carbon footprints of Scandinavian wastewater treatment plants, using the life cycle assessment approach. Journal of Cleaner Production* **211**, 1001–1014.
- Energimyndigheten 2022 *Växthusgasutsläpp från vindkraft (Green House Emissions from Wind Power)*. Swedish Energy Agency, Eskilstuna, Sweden. ISSN 1404-3343, ISBN-978-91-7993-063-9.
- EPCON 2021 *Biogas Digestate Treatment by MVR Evaporation*. Produktdatablad.
- Eskicioglu, C., Galvagno, G. & Cimon, C. 2018 *Approaches and processes for ammonia removal from side-streams of municipal effluent treatment plants. Bioresource Technology* **268**, 797–810.
- EU 2021 Regulation (EU) 2021/1119 of the European Parliament and of the Council of 30 June 2021 establishing the framework for achieving climate neutrality and amending Regulations (EC) No 401/2009 and (EU) 2018/1999 ('European Climate Law'). *Official Journal of the European Union* 9.7.2021.
- Ghosh, S., Lobanov, S. & Lo, V. K. 2019 *An overview of technologies to recover phosphorus as struvite from wastewater: Advantages and shortcomings. Environmental Science and Pollution Research* **26**, 19063–19077.
- Göteborg energi 2023 *Miljövärden för levererad fjärrvärme 2022 - Slutliga miljövärden 2023-04-30 (Environmental Values for Delivered District Heating 2022 – Final Environmental Values 2023-04-30)*. Gothenburg Energy, Gothenburg, Sweden.
- Jönsson, H. 2019 *Fosfor, kväve, kalium och svavel-tillgång, sårbarhet och återvinning från avlopp (Phosphorus, Nitrogen, Potassium and Sulfur – Availability, Vulnerability and Recycling from Wastewater)*. Rapport 105, Institutionen för energi och teknik, SLU, Sweden.
- Kanders, L., Yang, J. J., Baresel, C. & Zambrano, J. 2019 *Full-scale comparison of N<sub>2</sub>O emissions from SBR N/DN operation versus one-stage deammonification MBBR treating reject water – and optimization with pH set-point. Water Science and Technology* **79**(8), 1616–1625.
- Krogstad, T., Sogn, T. A., Asdal, Å. & Sæbø, A. 2005 *Influence of chemically and biologically stabilized sewage sludge on plant-available phosphorous in soil. Ecological Engineering* **25**(1), 51–60.
- Laanti, J. & Karjala, R. 2023 *Recovering nitrogen from municipal sludge – Advanced procurement and process development. In Presentation at NORDIWA 2023 Nordic Wastewater Conference, Gothenburg, Sweden.*
- Malovany, A., Johannesdottir, S., Schwede, S., Ahlgren, S., Flodin, E. & Shanmugam, K. 2022 *Återvinning av näringsämnen från avlopp (Recycling of Nutrients from Wastewater)*. Rapport 2022-6, Svenskt Vatten Utveckling.
- Menkveld, H. W. H. & Broeders, E. 2018 *Recovery of ammonia from digestate as fertilizer. Water Practice & Technology* **13**(2), 382–387. Fig 1 is reprinted from this paper with permission from the copyright holders, IWA Publishing.
- Metcalf, L., Eddy, H. P. & Tchobanoglous, G. 1991 *Wastewater Engineering: Treatment, Disposal, and Reuse*, Vol. 4. McGraw-Hill, New York.
- Richardson, K., Willem Lucht, W., Bendtsen, J., Cornell, S. E., Donges, J. F., Drüke, M., Fetzer, I., Bala, G., von Bloh, W., Feulner, G., Fiedler, S., Gerten, D., Gleeson, T., Hofmann, M., Huiskamp, W., Kumm, M., Mohan, C., Nogués-Bravo, D., Petri, S., Porkka, M., Rahmstorf, S., Schaphoff, S., Thonicke, K., Tobian, A., Virkki, V., Wang-Erlandsson, L., Weber, L. & Rockström, J. 2023 *Earth beyond six of nine planetary boundaries. Science Advances* **9**(37).
- Stenström, F., la Cour Jansen, J., Andersson Chan, A., Eliasson, M., Eriksson, Y., Marsteng, A.-K., Sehlén, R. & Thelin, G. 2017 *Rejektvattenbehandling – en kunskapssammanställning (Reject Water Treatment – A Knowledge Compilation)*. Rapport 2017-11, Svenskt Vatten Utveckling, Stockholm, Sweden.
- Videbris, K.-E. 2023 *Miljörapport Gryaab, Ryaverket 2022 (Environmental Report Gryaab, Rya WWTP 2022)*. Gryaab Rapport 2023:2, Göteborg, Sweden.
- Yee, R. A., Leifels, M., Scott, C., Ashbolt, N. J. & Liu, Y. 2019 *Evaluating microbial and chemical hazards in commercial struvite recovered from wastewater. Environmental Science & Technology* **53**(9), 5378–5386.

First received 8 November 2023; accepted in revised form 26 April 2024. Available online 13 June 2024