Blue-Green Systems

IWA PUBLISHING

Blue-Green Systems Vol 6 No 1, 90 doi: 10.2166/bgs.2024.054

Life cycle assessment of ammonium sulfate recovery from urban wastewater

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ABSTRACT

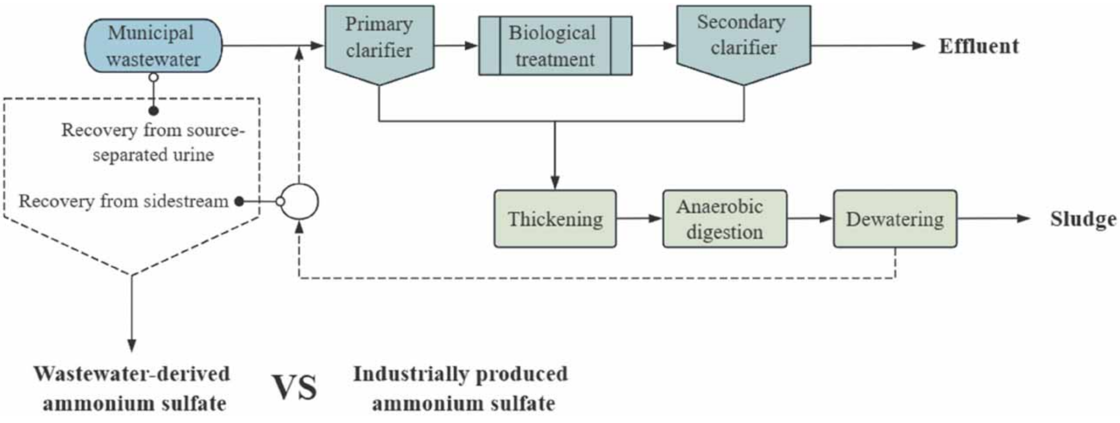
Anthropogenic nitrogen fluxes are profoundly altering the global biogeochemical nitrogen cycle. Better management of these nitrogen fluxes is essential. Recovering nitrogen from urban wastewater reduces both the energy and resources required to produce nitrogen-based fertilizer and to remove nitrogen from wastewater collected. Nitrogen can be recovered from wastewater in the form of ammonium sulfate, a common nitrogen-based fertilizer. In the urban setting, the technology can be applied to target source-separated urine or municipal wastewater. To assess the environmental sustainability of this approach, this study compared the life cycle environmental impacts of ammonium sulfate recovered from urban wastewater (through eight different recovery technology trains) and ammonium sulfate produced by six different industrial processes. The results show that wastewater-derived ammonium sulfate generally has lower potential environmental impacts than industrially produced ammonium sulfate in most of the impact categories assessed. The impact for the source-separated urine centralized recovery train is the smallest. The contribution analysis shows that energy, sulfuric acid and sodium hydroxide use are the major contributors, while the background inventory analysis shows that the results can be sensitive to the choice of region-specific background inventory. In the future, nitrogen recovery from urban wastewater is promising for the circular economy in cities.

Key words: ammonium sulfate, life cycle assessment, nitrogen recovery, wastewater

HIGHLIGHTS

- We compared wastewater-derived and industrially produced ammonium sulfate.
- Wastewater-derived ones generally have lower potential environmental impacts.
- The impact of the source-separated urine centralized recovery is the lowest.
- We also assessed the impacts of region-specific background life cycle inventory.
- Nitrogen recovery from wastewater is promising for the circular economy in cities.

GRAPHICAL ABSTRACT



ABBREVIATIONS

LCA	Life cycle assessment
LCI	Life cycle inventory
WWTP	Wastewater treatment plant
GLO	Global
RER	European subcontinent
ROW	Rest of the World

1. INTRODUCTION

Nitrogen is an important resource that plays a key role in areas such as agriculture and industry. Since the beginning of the 20th century, about 48% of the global population has been dependent on the industrial conversion of atmospheric nitrogen to ammonia and its derivatives such as urea and nitrates made through the Haber–Bosch process (Erisman *et al.* 2008). This process involves the substantial use of fossil energy and the emission of large quantities of greenhouse gases. Given the rapid growth of the world's population, the global demand for nitrogen resources is expected to increase further (Steen 2006; Sørensen *et al.* 2015). The significant increases in anthropogenic nitrogen fluxes and their release into the environment are profoundly altering the global biogeochemical nitrogen cycle (Spiller *et al.* 2022).

In view of this, better management of these nitrogen fluxes is essential. Wastewater has been cited as a potential source of supply in recent decades, and nitrogen is one of the key nutrients and pollutants in a variety of wastewater (Zhai *et al.* 2023). Nitrogen discharge from wastewater treatment plants (WWTPs) is a challenging issue worldwide as it leads to a variety of environmental impacts, including acidification, eutrophication, and secondary air pollution (Wu *et al.* 2016; Zhang *et al.* 2017). Various biological nitrogen removal processes (e.g., nitrification–denitrification, partial nitrification–denitrification, and autotrophic anaerobic ammonia oxidation) are applied to municipal wastewater; however, the removal of nutrients does not fit into the current context of sustainable development as defined by the United Nations (Winkler & Straka 2019). Therefore, the recovery of nitrogen from wastewater is a win-win decision, both in terms of reducing the amount of energy needed to produce nitrogen-based fertilizers through the traditional Haber–Bosch process and in terms of reducing the resources consumed by WWTPs to remove nitrogen (Kogler *et al.* 2021).

The fundamental processes and products of nitrogen recovery technologies from waste streams vary widely. For example, they include ammonia stripping, membrane contactors, electrochemistry, electrodialysis, and chemical precipitation (Cruz *et al.* 2019). Recovered nitrogen can be in the forms of struvite, ammonium sulfate, and ammonium nitrate. An unsolved question is whether these products recovered from wastewater can sustainably serve as substitutes for industrially produced nitrogen products. To understand the environmental sustainability of different recovery technologies, life cycle assessment (LCA) has been widely used (Lam *et al.* 2020). For instance, Igos *et al.* (2017) assessed the environmental impacts of recovering nitrogen from urine. van Zelm *et al.* (2020) evaluated the environmental impacts of sidestream ammonia recovery with the Nijhuis Ammonia Recovery system and compared them with that of the SHARON-anammox sidestream ammonia removal system. Most studies assessed the environmental impacts of one or multiple recovery processes without comparing the recovered products with the conventional products to be substituted.

The study aims to compare the potential environmental impacts between wastewater-derived and industrially produced ammonium sulfate. It also assesses the influence of region-specific background inventories on the LCA results.

2. MATERIAL AND METHODS

LCA is a standardized approach to assess the potential environmental impacts of a product or a process over its entire life cycle from raw material extraction to end-of-life disposal. This study follows the general LCA framework – goal and scope definition, inventory analysis, impact assessment, and interpretation of results.

2.1. Goal and scope definition

This study focused on assessing the potential environmental impacts of ammonium sulfate recovered from urban wastewater versus industrially produced ammonium sulfate from different processes. The main function of the studied systems is the production of ammonium sulfate, so the functional unit is defined as 'the production of

1 kg of ammonium sulfate'. The study uses a cradle-to-gate system boundary. An attributional LCA approach is used (European Commission – Joint Research Centre – Institute for Environment and Sustainability 2010). The foreground system of the nitrogen recovery processes considers only the recovery line but not the water line and the sludge line.

2.2. Inventory sources

2.2.1. Wastewater-derived ammonium sulfate

Inventory data for the wastewater-derived ammonium sulfate were obtained from the literature (Gong *et al.* 2024). The search was conducted using the Web of Science database with the keywords 'nitrogen recovery', 'life cycle assessment', 'municipal wastewater', and 'ammonium sulfate'. Because many nitrogen recovery processes are in the early stages of technological development, they have not yet been widely applied. In order to improve data quality and assessment accuracy, we selected five LCA studies that evaluated the recovery of ammonium sulfate from wastewater, covering eight different recovery trains such as source-separated urine, air-stripping, and membrane contactors (see Table 1 for details; specific data are in the Supplementary Material). Only one of the studies was using a functional unit of ammonium sulfate.

2.2.2. Industrially produced ammonium sulfate

The inventory data for the industrially produced ammonium sulfate are derived from the Ecoinvent v3.8 database, encompassing a total of six production processes (see Table 2 for details; specific data can be found in the Supplementary Material). The industrial production process is further divided into those where ammonium sulfate is the primary product (IP1-2, a reaction between sulfuric acid and ammonia) and those where ammonium

Table 1 | Summary of inventory sources for wastewater-derived ammonium sulfate

ID		Functional unit in the original study	Recovery technology trains	Scale	Recovery point	References
Foreground	Background					
N-1	NG-1 NR-1 NW-1	The treatment of 1 m^3 of wastewater	Decentralized recovery from source-separated urine using the microbial electrolysis cell	50,000 PE	Urine	Igos *et al.* (2017)
N-2	NG-2 NR-2 NW-2	The treatment of 1 m^3 of wastewater	Centralized recovery from source-separated urine using the microbial electrolysis cell	50,000 PE	Urine	Igos *et al.* (2017)
N-3	NG-3 NR-3 NW-3	The treatment of 1 kg of total dissolved nitrogen inflow	Nijhuis Ammonia Recovery system	–	Sidestream	van Zelm *et al.* (2020)
N-4	NG-4 NR-4 NW-4	1 kg of ammonium sulfate	Anaerobic digested sidestream and ammonia air-stripping	–	Sidestream	Kar *et al.* (2023)
N-5	NG-5 NR-5 NW-5	Treatment of 1 m^3 of incoming reject water from dewatered, digested sludge	Membrane contactor	–	Sidestream	Hogstrand *et al.* (2023)
N-6	NG-6 NR-6 NW-6	1 m^3 of the incoming anaerobic digestion supernatant	Ultrafiltration and liquid–liquid membrane contactor	500,000 PE	Sidestream	Vinardell *et al.* (2023)
N-7	NG-7 NR-7 NW-7	1 m^3 of the incoming anaerobic digestion supernatant	Ultrafiltration and reverse osmosis and liquid–liquid membrane contactor	500,000 PE	Sidestream	Vinardell *et al.* (2023)
N-8	NG-8 NR-8 NW-8	1 m^3 of the incoming anaerobic digestion supernatant	Ultrafiltration and reverse osmosis and forward osmosis and liquid–liquid membrane contactor	500,000 PE	Sidestream	Vinardell *et al.* (2023)

Table 2 | Summary of inventory sources for industrially produced ammonium sulfate

Category	ID	Inventory name	Recovery technology trains	Main raw materials	Source
Industrial production (byproduct)	IB-1	Ammonium sulfate {CN}| smelting and refining of nickel concentrate, 7% Ni | cut-off, U	Pyrometallurgical smelting process	Nickel concentrate	Ecoinvent v3.8
Industrial production (byproduct)	IB-2	Ammonium sulfate {GLO}| cobalt production | cut-off, U	Metal smelting process	Ore	Ecoinvent v3.8
Industrial production (byproduct)	IB-3	Ammonium sulfate {GLO}| smelting and refining of nickel concentrate, 16% Ni | cut-off, U	Combination of pyrometallurgical and hydrometallurgical processes	Nickel concentrate	Ecoinvent v3.8
Industrial production (byproduct)	IB-4	Ammonium sulfate {ROW}| primary zinc production from concentrate | cut-off, U	Electrometallurgical smelting process	Zinc concentrate	Ecoinvent v3.8
Industrial production (primary product)	IP-1	Ammonium sulfate {RER}| ammonium sulfate production | cut-off, U	Reaction of sulfuric acid and ammonia, formula: $2NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4$	Sulfuric acid, ammonia	Ecoinvent v3.8
Industrial production (primary product)	IP-2	Ammonium sulfate {ROW}| ammonium sulfate production | cut-off, U	Reaction of sulfuric acid and ammonia, formula: $2NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4$	Sulfuric acid, ammonia	Ecoinvent v3.8

sulfate is a byproduct (IB1-4, all involving ore smelting processes for the production of heavy metals such as nickel, cobalt, and zinc).

2.3. Life cycle inventory

The life cycle inventory (LCI) consists of a foreground inventory and a background inventory. Foreground inventory data for wastewater-derived ammonium sulfate were obtained from the selected literature (Table 1) and linked to the background inventory in the Ecoinvent v3.8 database. The background inventory is region-specific such as European subcontinent (RER), Rest of the World (ROW), and Global (GLO). The background inventory selects these three types of datasets. Together with the foreground inventory, it constitutes three versions of the LCI. The GLO version of the LCI is the most complete, and chemicals missing from both the RER and ROW versions of the LCI are replaced with datasets from the GLO. Both foreground and background data for the industrial production of ammonium sulfate were obtained from the Ecoinvent v3.8 database.

2.4. Impact assessment

After the LCI was developed, the ReCiPe 2016 midpoint (H) method was selected as the impact assessment method (Huijbregts *et al.* 2017). The method is problem-oriented, linking inventory results to 18 midpoint indicators to assess potential impacts from a variety of perspectives, including human health, water, air quality, and soil. The midpoint impact indicators used in this study include global warming (kg CO_2 eq), stratospheric ozone depletion (kgCFC_{11}eq), fine particulate matter formation (kg$PM_{2.5}$eq), freshwater eutrophication (kg P eq), terrestrial ecotoxicity (kg 1,4-DCB), human carcinogenic toxicity (kg 1,4-DCB), mineral resource scarcity (kg Cu eq), and fossil resource scarcity (kg oil eq). These impact categories were selected on the basis of the most relevant environmental issues related to wastewater treatment, which have been used previously to assess wastewater treatment and resource recovery (Corominas *et al.* 2013; Fang *et al.* 2016; Garfí *et al.* 2017; Lam *et al.* 2022).

3. RESULTS AND DISCUSSION

3.1. Overall results and contribution analysis

3.1.1. Overall results

The life cycle impact assessment results cover eight different midpoint impact categories (Figure 1) (other results can be found in the Supplementary Material). Wastewater-derived ammonium sulfate (wastewater recovery in Figure 1) was compared with the industrial production of ammonium sulfate. In most impact categories, the variations of wastewater-derived ammonium sulfate are very high because of the very different recovery technologies employed. Industrial production processes with ammonium sulfate as the byproduct are also highly dispersed, far exceeding other processes in the categories of terrestrial ecotoxicity and mineral resource scarcity. Industrial production processes with ammonium sulfate as the primary product are low in most impact categories but the highest for global warming and fossil resource scarcity. As far as the averages are concerned, wastewater-derived ammonium sulfate has clear environmental advantages in terms of global warming, fine particulate matter formation, terrestrial ecotoxicity, and mineral resource scarcity, and is slightly better than industrial production processes where ammonium sulfate is the primary product in terms of freshwater eutrophication and human carcinogenic toxicity.

3.1.2. Contribution analysis

Based on the contribution analysis (Figure 2), it can be concluded that the largest contributor to global warming and fossil resource scarcity in the industrial production of ammonium sulfate as the primary product is ammonia. This is due to the fact that the conventional industrial conversion of atmospheric nitrogen to ammonia using the Haber–Bosch process is very energy-intensive. In industrial production with ammonium sulfate as a byproduct, emissions and other material inputs are the main contributors to fine particulate matter formation, terrestrial ecotoxicity, human carcinogenic toxicity, and mineral resource scarcity. Because the primary purpose of these industrial productions is metal smelting, many materials other than sulfuric acid are used in the production process, and a large amount of gases and particulates are emitted, as well as toxic wastewater-containing heavy metals.

For wastewater-derived ammonium sulfate, the contribution varies considerably between recovery technologies. For membrane technology-based recovery trains, such as membrane contactors (N-5), ultrafiltration (N-6), reverse osmosis (N-7), and forward osmosis (N-8), sodium hydroxide and energy use are the main contributors to environmental impacts. This is because membrane technology requires sodium hydroxide to adjust the pH and to clean the sediment from the membrane. For the remaining wastewater recovery processes (N1–N5), energy use and sulfuric acid are the main contributors. Overall, the significant contribution of sulfuric acid, sodium hydroxide, and energy use in the recovery of ammonium sulfate from wastewater highlights the need to explore potentially more sustainable alternatives.

3.2. Impact of background inventories

Three background inventories (GLO, RER, and ROW) were selected to investigate their impacts on the results of impact assessments (Figure 3). Among the assessed impact categories, GLO exhibits the highest environmental

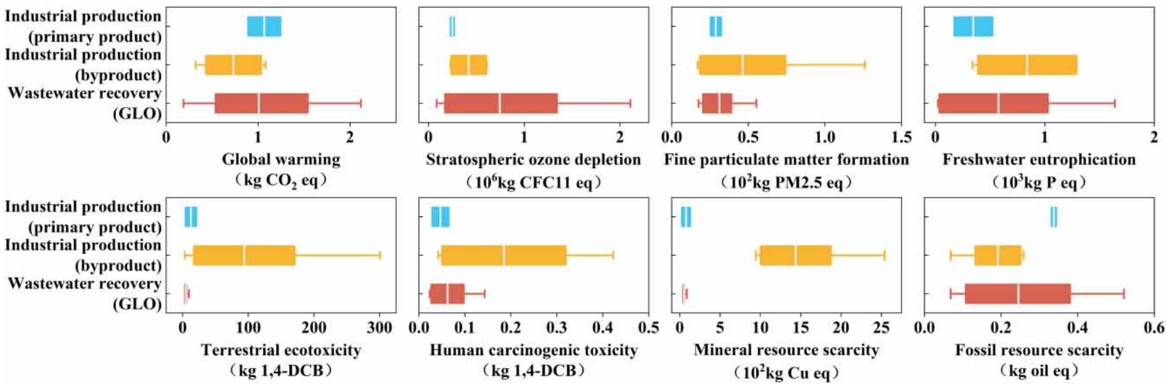


Figure 1 | Midpoint impact categories of wastewater-derived ammonium sulfate and industrially produced ammonium sulfate.

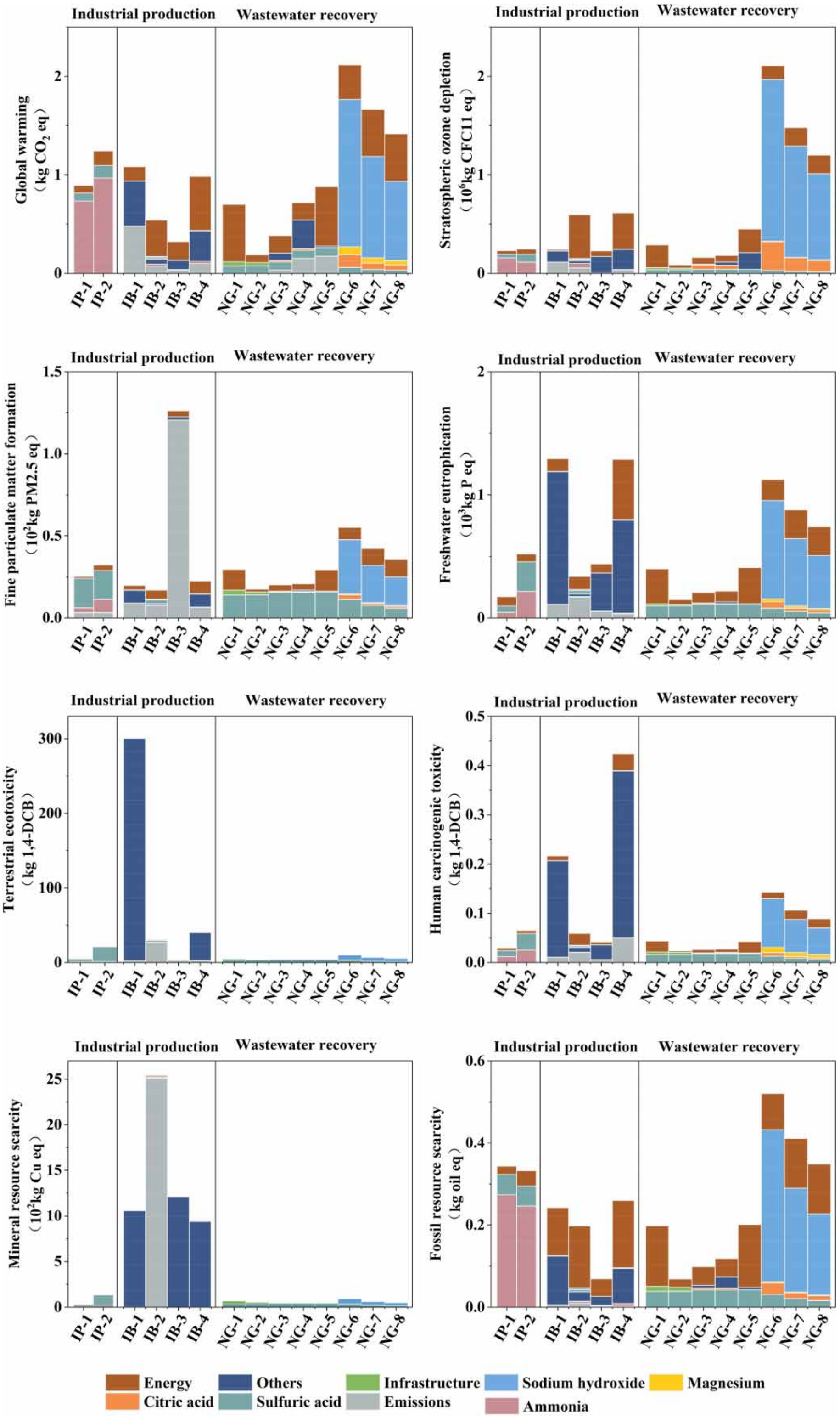


Figure 2 | Contribution analysis of wastewater-derived ammonium sulfate based on GLO background inventory and industrially produced ammonium sulfate.

impacts, with RER and ROW being relatively close. However, in specific categories such as global warming, fine particulate matter formation, and fossil resource scarcity, RER tends to have slightly lower environmental impacts. When combined with the contribution analysis (Figure 4), it becomes evident that energy consumption varies to varying degrees across all selected indicators. Additionally, there are significant differences in the environmental impacts of acidic chemicals in terrestrial ecotoxicity and mineral resource scarcity.

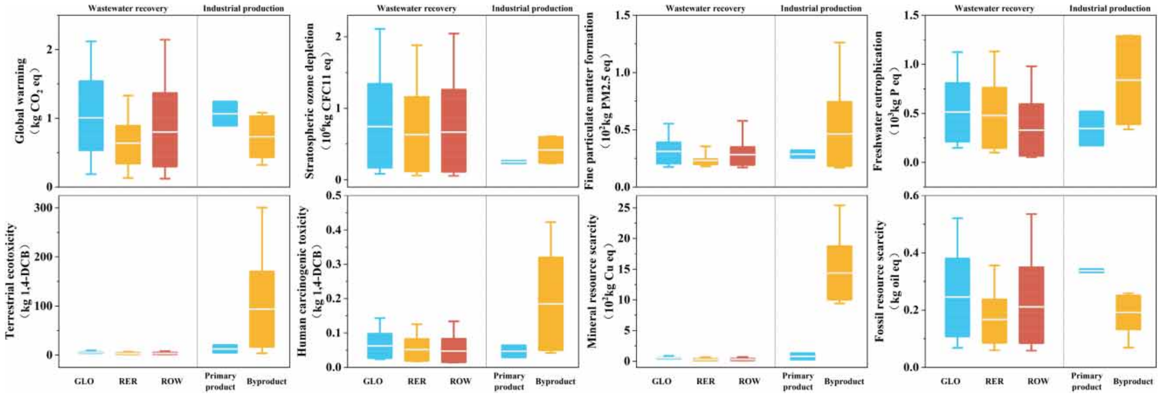


Figure 3 | Midpoint impact categories of wastewater-derived ammonium sulfate using three different region-specific background inventories (GLO, RER, and ROW).

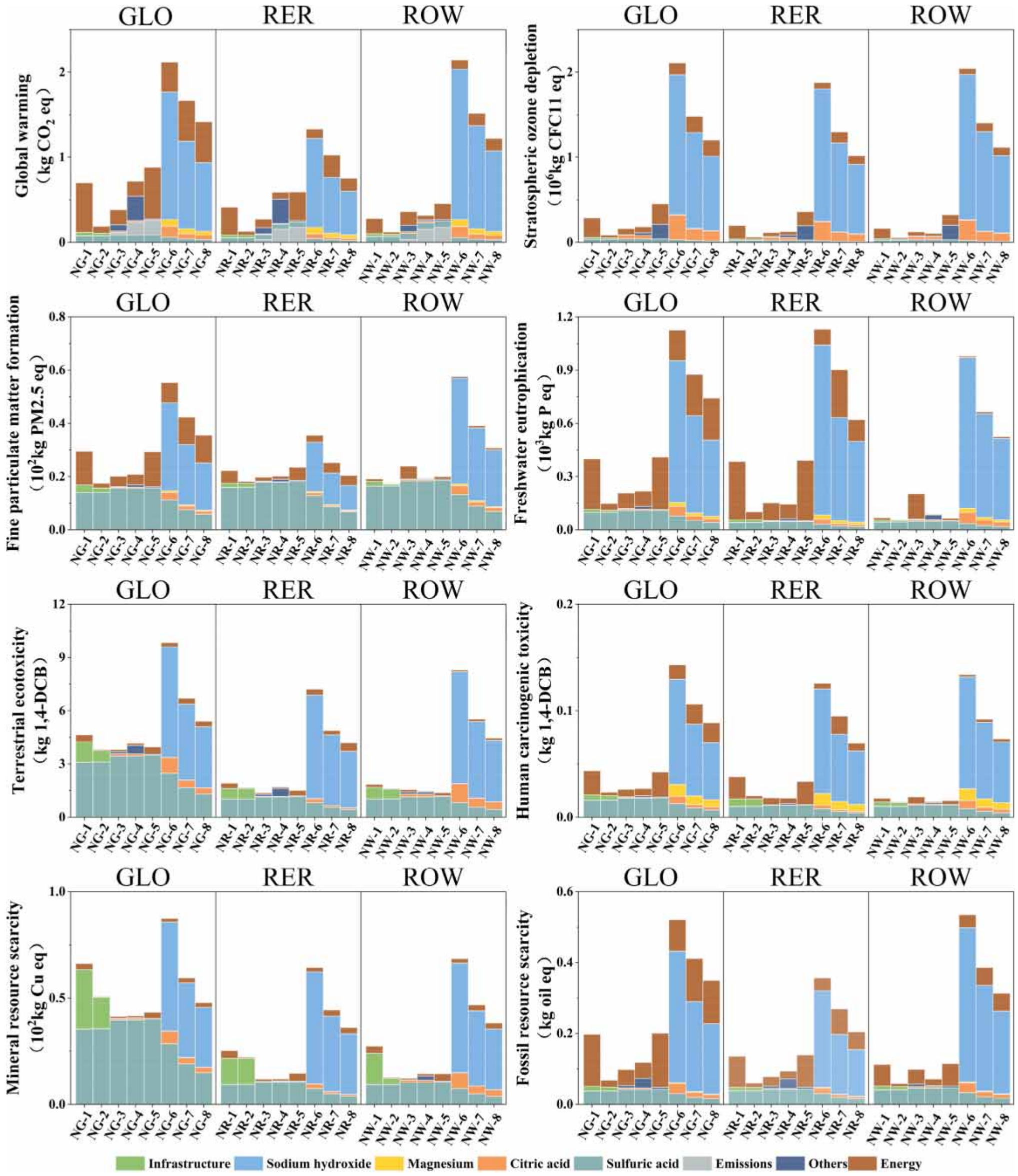


Figure 4 | Contribution analysis of using three different region-specific background inventories (GLO, RER, and ROW)

There are several reasons accounting for the differences among the three datasets. Firstly, variations in the energy structure play a significant role, with each region having distinct natural conditions and available resources, leading to different types and proportions of energy sources. Secondly, differences in industrial development, including diverse industrial layouts and processes across regions, contribute to disparities in the environmental impact of products. The third factor is environmental policy, as variations in environmental policies and practices among regions may influence the environmental factors and weights included in the dataset. Lastly, the data collection methodology can differ from one geographical region to another in terms of approach and availability, resulting in varying values for environmental indicators and factors in different contextual datasets.

As observed in this study, electricity consumption is a key contributor to the five indicators of global warming, stratospheric ozone depletion, freshwater eutrophication, human carcinogenic toxicity, and fossil resource scarcity in wastewater recovery and occupies an important place in WWTPs. Therefore, the carbon footprint of energy must be significantly reduced in order to minimize the impact on the environment. Renewable energy, recognized as a green and low-carbon form of energy, stands as a crucial pathway to mitigate the carbon footprint of energy supply. In addition to considering the contribution of common renewable energy sources such as solar, wind, and hydro in the energy mix, WWTPs can also recover thermal and chemical energy from wastewater. This adaptation reduces the reliance on traditional energy sources, reduces the environmental impact of nitrogen recycling, and facilitates the transition of WWTPs into resource recovery facilities.

3.3. Differences between recovery trains

The recovery of ammonium sulfate from wastewater varies significantly using different recovery trains. The eight recovery processes selected focus on recovering ammonium sulfate from source-separated urine or sidestream at WWTPs (Figure 5). Environmental impacts, assessed through eight indicators, show that source-separated urine centralized recovery train (N-2) has the lowest environmental impact, while membrane technologies such as ultrafiltration (N-6), reverse osmosis (N-7), and forward osmosis (N-8) have higher environmental impacts.

For the source-separated recovery processes (N-1, N-2), the inventory includes not only inputs of energy and chemicals but also the environmental impacts associated with source separation pipeline infrastructure and transportation of urine. Compared to centralized recovery (N-2), decentralized recovery (N-1) contributes significantly to environmental impact through energy consumption and infrastructure investments.

For the WWTP sidestream recovery processes (N-3, N-4, N-5, N-6, N-7, and N-8), the Nijhuis Ammonia Recovery system (N-3) and the ammonia air-stripping process (N-4) produce lesser environmental impacts. The Nijhuis Ammonia Recovery system is a new technology based on improvements to the ammonia air-stripping process. Both processes have similar workflows, but the Nijhuis Ammonia Recovery system has a lower demand for chemical input. In the membrane technology-based recovery train (N-5, N-6, N-7, and N-8), the recovery processes that use ultrafiltration, forward osmosis, and reverse osmosis for pretreatment (N-6, N-7, N-8) have a higher potential environmental impact than membrane contactor recovery (N-5). This is attributed to the use of sodium hydroxide, which is used not only for pH adjustment but also for cleaning the membranes.

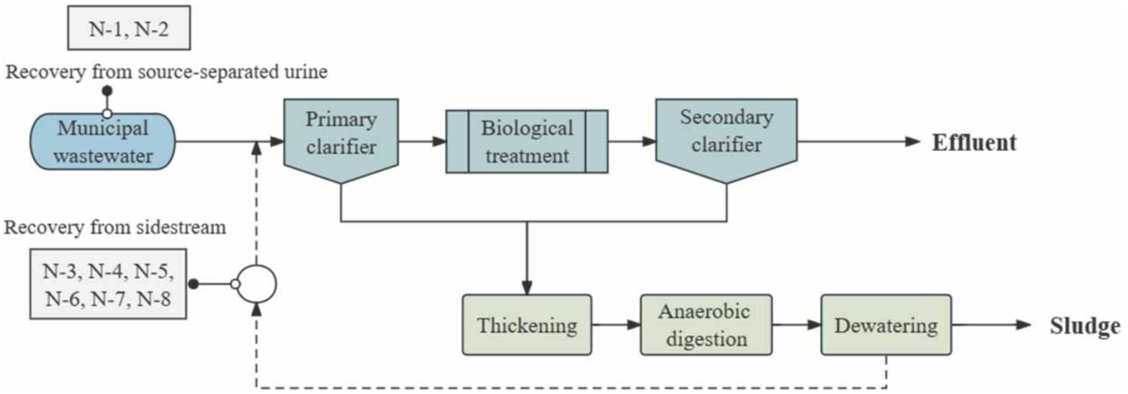


Figure 5 | Recovery points for wastewater-derived ammonium sulfate.

3.4. Uncertainties and limitations

Recycled ammonium sulfate from wastewater offers environmental advantages over industrially produced ammonium sulfate, although it still presents some limitations. Some of the emerging recycling technologies included in this study are currently at the pilot-scale stage, and the assessments conducted are prospective. Their inventory could carry more uncertainties compared to more mature technologies. The foreground data sources have limited information on uncertainties, which hinders the possibility of conducting a Monte-Carlo uncertainty analysis. In the future, a process model-based approach can potentially be used to simulate these recovery processes for generating foreground inventory and for exploring the uncertainty space of the environmental impacts of these systems.

The limitation of this study lies in the cut-off approach, which solely focuses on the environmental impacts of the ammonium sulfate recovery process. The impacts from the upstream wastewater treatment process and the construction of the wastewater treatment facility are not within the system boundary. Furthermore, our comparison is limited to assessing the environmental dimension of wastewater-derived ammonium sulfate and industrially produced ammonium sulfate.

In the future, with the development and optimization of new technologies for nitrogen recovery from wastewater, and the pressing need for sustainable development and decarbonization (Zheng & Lam 2024), the recovery of ammonium sulfate from wastewater is a promising alternative to industrial production of ammonium sulfate. Future professionals in this field could evaluate the differences between wastewater-derived ammonium sulfate and industrially produced ammonium sulfate from multiple dimensions such as economic aspects, social aspects, and uncertainties (van der Hoek *et al.* 2018). This would better inform decision-makers and end-users of ammonium sulfate about the product.

4. CONCLUSION

This study compared the potential environmental impacts between ammonium sulfate recovered from urban wastewater through eight different recovery trains and ammonium sulfate produced from six different industrial production processes. The key implications are as follows:

- In general, the overall results show that wastewater-derived ammonium sulfate has lower potential environmental impacts than industrially produced ammonium sulfate in most of the impact categories assessed.
- Of the eight different recovery trains, the impact of the source-separated urine centralized recovery train is the lowest.
- The contribution analysis shows that sulfuric acid, sodium hydroxide, and energy use are the primary contributing factors. Exploring alternatives that use less chemicals and energy is key to reducing environmental impacts.
- The background inventory analysis shows that the GLO dataset has the highest environmental impacts. The energy structure, industrial development, environmental policies, and data collection methods in different regions can potentially affect the results. Therefore, localized background inventory is needed to better assess the recovery technology applied in any given region.
- In the future, nitrogen recovery from urban wastewater is a promising pathway for the circular economy in cities. The choice of technology depends on the particular application scenario with the need to further assess the financial and social implications.

ACKNOWLEDGEMENT

The authors acknowledge funding from the Jiangsu Science and Technology Department (no. BK20220300), the Natural Science Research of Jiangsu Higher Education Institutions of China (no. 22KJB610013), and the Summer Research Scholars program at Duke Kunshan University.

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.

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First received 7 December 2023; accepted in revised form 5 March 2024. Available online 18 March 2024