Resources, Conservation & Recycling 198 (2023) 107167

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.elsevier.com/locate/resconrec

Developing product level indicators to advance the nitrogen circular economy

Chayse M. Lavallais[a], Jennifer B. Dunn[a,b,*]

[a] Chemical and Biological Engineering Department, Northwestern University, Evanston, IL, United States of America
[b] Center for Engineering Sustainability and Resilience, Northwestern University, Evanston, IL, United States of America

ARTICLE INFO

Keywords:
Circular economy
Nitrogen
Indicators

ABSTRACT

Increasingly, circularity indicators for material, energy, and water systems guide circular economy design. While indicators for products made from recycled carbon-based materials are somewhat common, peer indicators for waste nitrogen-derived products are limited. It is important, however, to develop such indicators to guide emerging technologies that transform waste nitrogen into products. In this study, we summarize the nitrogen circularity indicator literature, emphasizing the agricultural and wastewater sectors. Next, we use the Material Circularity Indicator (MCI) developed by the Ellen MacArthur Foundation, to quantify the circularity of products made from waste nitrogen in swine manure. We considered four test cases using different technologies to recover nitrogen from the manure. Our analysis indicates that technologies that seem to increase circularity on the surface may not yield a substantial increase in MCI results. Finally, we discuss the strengths and weaknesses of using the MCI for product-level analysis and further developments.

1. Introduction

The linear take-make-waste model is the dominant but unsustainable pathway for industrially developed products. The circular economy is a potentially sustainable alternative that can better steward limited energy, water, and material resources. It entails system-level innovations that design out waste, maximize resource value, minimize negative environmental impacts, and build economic, environmental, and social capital. To date, a consensus definition of a circular economy remains evasive. A literature review by Kirchherr et al. (2017) tallied 114 different circular economy definitions. Overall, the general topic of the circular economy is increasingly a focus of the academic community as evidenced by a growing body of literature. The ISI Web of Science database contains 2279 articles published on the topic of the circular economy since 2001 with 90% of those articles being published between 2015 and 2019 (Goyal et al., 2021).

To measure societal progress toward a circular economy, researchers and organizations have developed quantitative circularity indicators. These indicators quantify circularity at scales from the product to the national level. Corporations tend to favor product-level indicators that allow them to monitor and communicate their transition to circularity. Efforts to devise product-level circularity indicators have emphasized carbon-based systems such as plastics. Lonca et al. (2020) examined the environmental benefits and circularity effects of increasing the use of recycled polyethylene terephthalate (PET) on the U.S. plastic bottle market. Rossi et al. (2020) evaluated different indicators to assess the circularity of a company called CIMFLEX, which develops products in civil construction from recycled plastics. Given the immense challenge of eliminating plastic waste, much of the circular economy dialog has focused on the plastics industry and has led to the development of various policies and commitments to promote plastics circularity at an international (United Nations Environmental Program, 2022), national (US Plastic Pact, 2020), and regional level (European Commission, 2018).

Compared to carbon-based products, development of circularity indicators for nitrogen-based products is limited. Yet, it is important to develop these types of indicators to identify pathways and practices that improve the management of the nitrogen cycle. Currently, inefficient agricultural practices, wastewater treatment processes that don't recover nutrients, and high-emitting industrial processes (e.g., nitric acid production) produce nitrogen pollution in various forms. These forms include N_2O, NH_3, NO_x, and NO_3^-, each of which negatively affects the environment and human health. N_2O emissions have almost 300 times the global warming potential of CO_2. NH_3, and NO_3^- cause

* Corresponding author at: Chemical and Biological Engineering Department, Northwestern University, Evanston, IL, United States of America.
E-mail address: jennifer.dunn1@northwestern.edu (J.B. Dunn).

https://doi.org/10.1016/j.resconrec.2023.107167
Received 14 April 2023; Received in revised form 17 July 2023; Accepted 7 August 2023
Available online 18 August 2023

eutrophication, algal blooms, and dead zones in water systems. NO_x emissions increase ozone production which can lead to respiratory ailments among other negative health effects. Recovering N from waste will prevent emissions of these nitrogen compounds, and decrease the dependence on the Haber-Bosch process, which is responsible for 1% of global energy consumption and 1.4% of global CO_2 emissions annually (Kyriakou et al., 2020). Circularity indicators can play a role in rebuilding circularity into the nitrogen cycle.

Current and emerging pathways can transform waste nitrogen into usable precursors for multiple products. The realization of nitrogen recovery and incorporation into these products would enhance nitrogen circularity. One current example of such circularity is the application of animal manure or biosolids to crops as a nitrogen fertilizer. U.S. state nutrient management plans and best management practices that regulate the use of N-containing wastes on farm fields are growing increasingly stringent, however, over concerns related to the excessive nutrient loads, foul odors, and emissions these wastes contain (Centner, 2012). These guidelines and regulations therefore may limit increasing circular N use in this way. On the other hand, it is possible to use various technologies to extract nitrogen from waste streams so that it can re-enter a circular system in a chemical form that is identical to synthetic fertilizers. Examples of these technologies include gas-permeable membranes, struvite precipitation, air stripping, and ion exchange. Each of these technologies can produce synthetic fertilizers such as ammonium sulfate (Pandey and Chen, 2021). It is possible to create other products from waste N besides fertilizers. For example, the coupled aerobic-anoxic nitrous decomposition operation (CANDO) can convert ammonia in wastewater into N_2O, which, at a wastewater treatment plant, can be co-combusted with biogas to produce energy in a combined heat and power plant, for example (Scherson et al., 2013). In this case, the N_2O forms N_2 upon combustion. Ammonia can be "food" for microbes that produce proteins used in animal feed (Matassa et al., 2015). Other sources of waste nitrogen, such as NO_x, can also be transformed into ammonia to then used as an input for further chemical processing (Xue et al., 2021). These technologies could be used to recover nitrogen from animal manure, wastewater, food waste, and the chemical industry and displace much of the demanded nitrogen in the United States (Fig. 1).

Developing technologies that recover and convert waste nitrogen to products that are not immediately consumed (e.g., through combustion) improves nitrogen circularity, reduces reliance on nitrogen produced from the Haber-Bosch process, and has the potential to reduce the environmental burden of both the waste nitrogen and the product. However, to direct the development of these technologies it is important to quantify the circularity of nitrogen in the resulting products. A need therefore exists for an appropriate indicator to monitor the implementation of the circular economy to waste nitrogen-derived products.

Accordingly, we explore potential existing circularity indicators that could be used to quantify the circularity of products made from nitrogen. We then begin the exploration of circularity indicators applied to waste nitrogen-based products using the production of swine feed from waste nitrogen in manure as a case study. We conclude with a discussion of ongoing research and analysis needs to build robust waste nitrogen-based product circularity indicators.

2. Circularity indicators for nitrogen systems

Increasing the circularity of nitrogen systems could reduce their environmental and human health impacts and aid progress toward restoring the nitrogen cycle. Cataloging circularity indicators for quantifying nitrogen circularity in agriculture, wastewater treatment, and chemicals production is an important first step in broadening the use of these indicators to guide the development of a circular nitrogen economy.

Analysts have developed circularity indicators for agriculture systems for nitrogen, phosphorous, and other nutrients. Velasco-Muñoz et al. (2021) published a review analyzing existing agricultural circularity indicators. The authors selected indicators that were developed to communicate improved process efficiencies, extended material lifespans, and production from waste. Indicators were also separated into four groups from a sustainability standpoint: technical (i.e., energy or material efficiencies), social (i.e., human welfare), environmental (e.g., climate change), and economic (e.g., cost-effectiveness). Within these different categories, 41 different circularity indicators were identified, with 7 being directly applicable to nitrogen or other nutrient flows (Table 1). The remaining 34 indicators reviewed concern other topics such as water quality, greenhouse gas emissions, farm income levels, or changes in unpaid time individuals spend collecting biomass.

For wastewater treatment systems, many different indicators and frameworks have been developed to quantify the progress of the circular economy. While there are many indicators existing to quantify the circularity of water use in these systems, the number of indicators

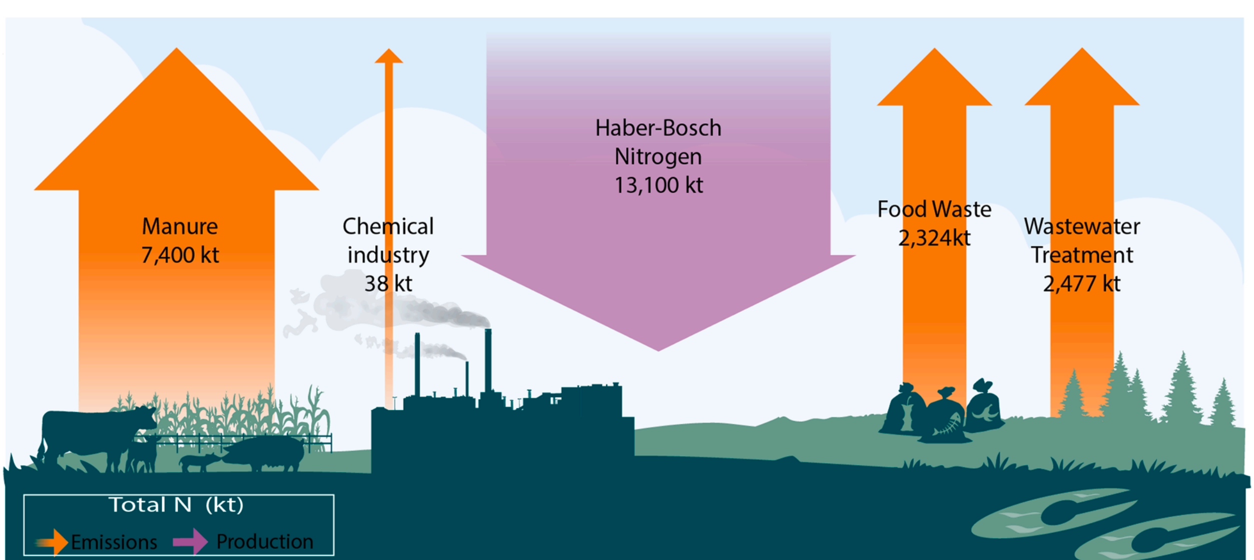

Fig. 1. Estimated U.S. total annual nitrogen produced from the Haber-Bosch process that enters the "nitrogen economy" (purple arrow) (Apodaca 2018) versus nitrogen generated from waste nitrogen sources (orange arrows) (Bian et al., 2020; EPA 2023; Liu et al., 2016).

Table 1
Existing nitrogen and phosphorus indicators for agricultural systems.

Nutrient (s)	Evaluated system	Name of indicator	Definition of indicator
N	Regional agro-food network in France (Fernandez-Mena et al., 2020)	Nitrogen balance	Difference between N content of fertilizer inputs and crop outputs
N&P	Organic farm waste management in Cantabria, Spain (Cobo et al., 2018)	Circularity indicator of components	Amount of N and P that is reused and taken up by crop with respect to total N and P present in organic farm waste
N	Crop-Livestock farms in Ethiopia (Tadesse et al., 2019)	Nitrogen recycling index	Proportion of input N that is recycled back into system
N		Nitrogen use efficiency	The ratio between harvested N output and managed N inputs
N		Crop–livestock ratio	The relative allocation of N to crop and livestock production and consumption
P	Food system in Brussels Capital Region (Papangelou et al., 2020)	Food Circularity	P potentially reused or reusable for agriculture, within and outside of a city boundary
N&P	Agro-food-waste systems (Harder et al., 2021)	Input Circularity	The fraction of total nutrient inputs that are supplied from waste streams
N&P		Output Circularity	The fraction of nutrients in waste streams that are recycled to agricultural production
N&P	Agriculture and food systems in Broadbalk, UK, Flanders, Belgium and Hengelo, Netherlands (Van Loon et al., 2023)	FinnCI	The proportion of flow (i.e. nitrogen) that cycles through components (i.e. crops, animal feed) of the system
N&P		FiggeCI	The number of times a resource or nutrient is used in a system, including its first time use
N&P		CyCt	The number of full cycles completed on average by a nutrient input cohort before the entire cohort has vanished through all exports and losses
N&P		CyCtR	The number of full cycles that would be completed on average by an input cohort if no products were exported

devoted to resource recovery is limited. Preisner et al. (2022) completed a literature review analyzing existing circularity indicators related to resource recovery in the wastewater treatment sector. 97 references were used to identify sixteen existing indicators, covering various aspects of resource recovery, such as nutrient removal, sludge treatment, and biogas production. However, only four of the indicators quantify the circularity of systems that recover nitrogen or other nutrients from wastewater systems (Table 2).

In the chemical industry, the development of circularity indicators to quantify circularity for a product's life cycle has been limited. To date, the literature does not address existing circularity indicators within the chemical industry relating to nitrogen. Wang and Hellweg (2021) completed a study to advance the idea of circularity within the chemical sector. The authors suggest that the absence of existing indicators is attributable to a lack of a clear definition of sustainable circularity within the chemicals industry.

Table 2
Existing nitrogen and phosphorus indicators for wastewater treatment systems.

Nutrient (s)	Evaluated system	Name of indicator	Definition of indicator
N&P	Wastewater Treatment Plant (European Commission, 1991)	Nutrient removal efficiency indicator	Percent of total N or P load reduced
N&P	Wastewater Treatment Plant (Shaddel et al., 2019)	Nutrient recovery indicator	N or P recovered annually
P	Wastewater Treatment Plant (Miksch and Sikora, 2012)	Biological dephosphatation potential indicator	P recovery potential from sludge dewatering liquors
N&P	Wastewater Treatment Plant (Li and Brett, 2012)	Effluent inorganic content indicator	Total mass of inorganic N or P in WWTP effluent

In each of these sectors, indicators exist to quantify the nitrogen circularity of a system. However, these circularity indicators do not take the perspective of evaluating the circularity of products made from recovered waste nitrogen. Yet, this perspective will be increasingly important as competing technologies and pathways evolve that make fuels, chemicals, and other products from waste that contains nitrogen. Therefore, there is a need to develop new indicators or develop methodological frameworks for the application of existing indicators for evaluation of the circularity of products made from waste nitrogen. Given its wide use for many applications in the circular economy, we began our exploration of these indicators with the Material Circularity Indicator (MCI).

3. Material circularity indicator

Developed by the Ellen MacArthur Foundation, the MCI is a prominent, product-centric circularity metric that uses material flow analysis as the basis of its calculations (Ellen MacArthur Foundation and Granta Design, 2015; de Oliveira et al., 2021). MCIs for a component or product range between zero and one. Zero indicates a fully linear product; one indicates a completely circular product. There are three main parameters needed to calculate a product's MCI: the amount of virgin material utilized, the amount of unrecoverable waste generated, and a utility factor that accounts for lifespan and use intensity of the product (Ellen MacArthur Foundation and Granta Design, 2015). The MCI is considered to be one of the more complete product-level circularity frameworks available (Garza-Reyes et al., 2019) and reflects many important aspects of the circular economy (Elia et al., 2017).

Eq. (1) defines the MCI.

$$MCI = 1 - LFI * F(x) \tag{1}$$

where LFI is the linear flow index and F(x) is the utility factor.

The LFI (Eq. (2)) reflects the linearity of a product.

$$LFI = \frac{V + W}{2M} \tag{2}$$

where V is the mass of virgin raw material consumed to make the product, M is the mass of the finished product, and W is the mass of unrecoverable waste generated at the end of the product's life.

Eq. (3) defines V.

$$V = M(1 - F_R - F_U - F_S) \tag{3}$$

Where F_R, F_U, and F_S are the fraction of material that is recycled, reused, or derives from sustainable biological sources respectively.

Eq. (4) defines unrecoverable waste.

$$W = M(1 - C_R - C_U - C_S - C_E) \quad (4)$$

Where C_R, C_U, C_S, and C_E are the mass fraction of the product at its end-of-life that is recycled, reused, composted (in the case of biological waste), or incinerated for energy recovery, respectively.

Overall, in Eq. (2), when V is large and W is small, LFI will approach one. On the other hand, products made from reused or recycled raw materials and are recycled or reused at end of life have an LFI of zero. When LFI is zero, the MCI is 1, which would indicate the product is fully circular.

The utility factor, F(x), (Eq. (5)) considers the length and intensity of the product's use.

$$F(x) = \frac{0.9}{\frac{L}{L_{ave}} \frac{U}{U_{ave}}} \quad (5)$$

Where L is the average lifetime of the product, L_{ave} is the industry average lifetime of the product, U is the number of functional units achieved during the use phase of the product, and U_{ave} is the average functional units achieved for an industry average product. The concept of functional unit arises from life cycle assessment frameworks, in which the functional unit captures the service performed by the system under study. It is the basis of comparison when multiple products or systems are evaluated.

Multiple studies have adapted the MCI to quantify circularity within a specific industry. For example, Verberne (2016) developed the building circularity indicator from the core concept of the MCI. This indicator introduces a weighting factor into the MCI that adjusts it to reflect the difficulty encountered in disassembling each component of a building. Additionally, Kakwani et al. (2022) adapted the MCI to create the Water Circularity Indicator, which removes the utility factor and introduces additional terms for reducing, restoring, and reclaiming the used water. Another example is Rocchi et al. (2021)'s modification of the MCI for application in the poultry industry to assess the circularity of broiler farming. The MCI, however, has yet to be used to quantify the circularity of products made from waste nitrogen.

Therefore, we assess its applicability to evaluate circular nitrogen products with a unique case study analyzing the production of swine feed from waste nitrogen. We illustrate how the different elements of the MCI affect perceptions of nitrogen product circularity. Finally, we present a framework for further development of the MCI to circular nitrogen products.

3.1. Applying the MCI to evaluate approaches to N circularity in swine feed production

Animal production gives rise to many negative environmental effects including greenhouse gas emissions. Raising and growing feed for increasing heads of pigs, cows, poultry, and other animals accounts for almost 60% of all food production-related greenhouse gas emissions, accounting for almost 10,000 $TgCO_2eq/yr$ (Xu et al., 2021). Additionally, animal feed production is also prone to heavy nitrogen losses. Only around 17% of nitrogen in fertilizer ends up in animal protein (Matassa et al., 2015). Increasing the nitrogen circularity of animal feed production would reduce greenhouse gas emissions and nitrogen runoff. Accordingly, we chose animal feed as our case study for applying the MCI.

Corn and meal from soybeans are major components of animal feed. Corn agriculture is more nitrogen-consuming than soybean farming. Of feed for cattle, swine, and poultry, swine feed has the greatest proportion of corn and therefore the greatest potential to benefit from increased nitrogen circularity in agriculture systems. Additionally, swine feed is a major agricultural product that at 61.6 M tons annual consumption constitutes about one third of all animal feed produced in the US (IFEEDER, 2020). For these two reasons, we selected swine feed as the specific feed type for analysis.

The two main sources of waste nitrogen associated with agriculture are farm field run off and waste manure. Because nitrogen from manure is more straightforward to capture and use than nitrogen in farm field runoff, we tap manure as the source of waste nitrogen in the case studies. In the United States, most manure goes unused and emits NH_3 and N_2O as it degrades in lagoons. It is a largely untapped resource for N circularity that can reduce demand for virgin nitrogen in fertilizers used to grow crops (corn, soy) used in feed and can be used to produce feed directly. We explore both of these options in four cases.

In the first case (baseline), swine feed (corn, soy) is produced conventionally with the use of traditional synthetic nitrogen fertilizer produced from the Haber-Bosch process. The second case introduces unprocessed animal manure as an additional source of nitrogen fertilizer for the production of corn and soybeans used as swine feed. Case three uses a gas-permeable membrane to increase nitrogen recovery from animal manure, producing ammonium sulfate, which can be used as a fertilizer. Finally, case four uses some of the nitrogen recovered with the membrane to produce microbial protein, which is used to substitute for the soybean meal in the swine feed formulation. The remaining recovered nitrogen from the gas permeable membrane is applied as a fertilizer for corn production. Fig. 2 shows the nitrogen flows in each of the four cases.

3.2. Adapting MCI for products made from waste nitrogen

Importantly, in our case studies, nitrogen is the material that we use the MCI to track. Accordingly, we begin by calculating the total mass of input nitrogen for a given product. For that product, which in our case studies is swine feed, any nitrogen that originates from the Haber-Bosch process is considered virgin (V in Eqs. (2) and 3). Unrecoverable waste (W in Eqs. (2) and 4) is any nitrogen that is lost to the environment at any point during the production of the product. Waste nitrogen can have different forms such as NH_3, N_2O, NO_3^-, NO_x, and N_2. In our analysis, we assess different technologies to recover waste nitrogen from sources like wastewater or manure. The recovery efficiency of these systems determines how much waste nitrogen they produce. The MCI also includes a utility factor (Eq. (5)) that considers how long and to what intensity (as reflected in the number of functional units achieved over the product's lifetime) the product is being used compared to industry standards. Currently, it is unknown how the use of waste nitrogen will impact the lifespan or intensity of the use of products developed from waste nitrogen. Therefore, we used the default utility factor of 0.9.

3.3. Parameter selection

To calculate the impact of introducing more circular nitrogen systems into animal feed, it is essential to understand swine feed formulations. While many formulations are possible, we used the standard swine feed contained in the Greenhouse gases, Regulated Emissions, and Energy Use in Technologies (GREET) model (Argonne National Laboratory 2021), which is about 88% corn and 12% soybean meal by mass. Producing one kilogram of corn requires 16 g of nitrogen and producing one kilogram of soybean meal requires two grams of nitrogen, resulting in about 14.3 g of nitrogen required for every kilogram of swine feed (Argonne National Laboratory, 2022). Although there are other components of swine feed, they are not included in our calculations as they do not contain nitrogen.

We used Intergovernmental Panel on Climate Change (IPCC) emission factors to estimate the amount of direct and indirect waste nitrogen generated from organic fertilizers (i.e., animal manure) and synthetic fertilizer (e.g., urea, ammonium sulfate) (IPCC, 2006). In the case of synthetic fertilizers, 1 wt% of applied nitrogen is emitted directly as N_2O

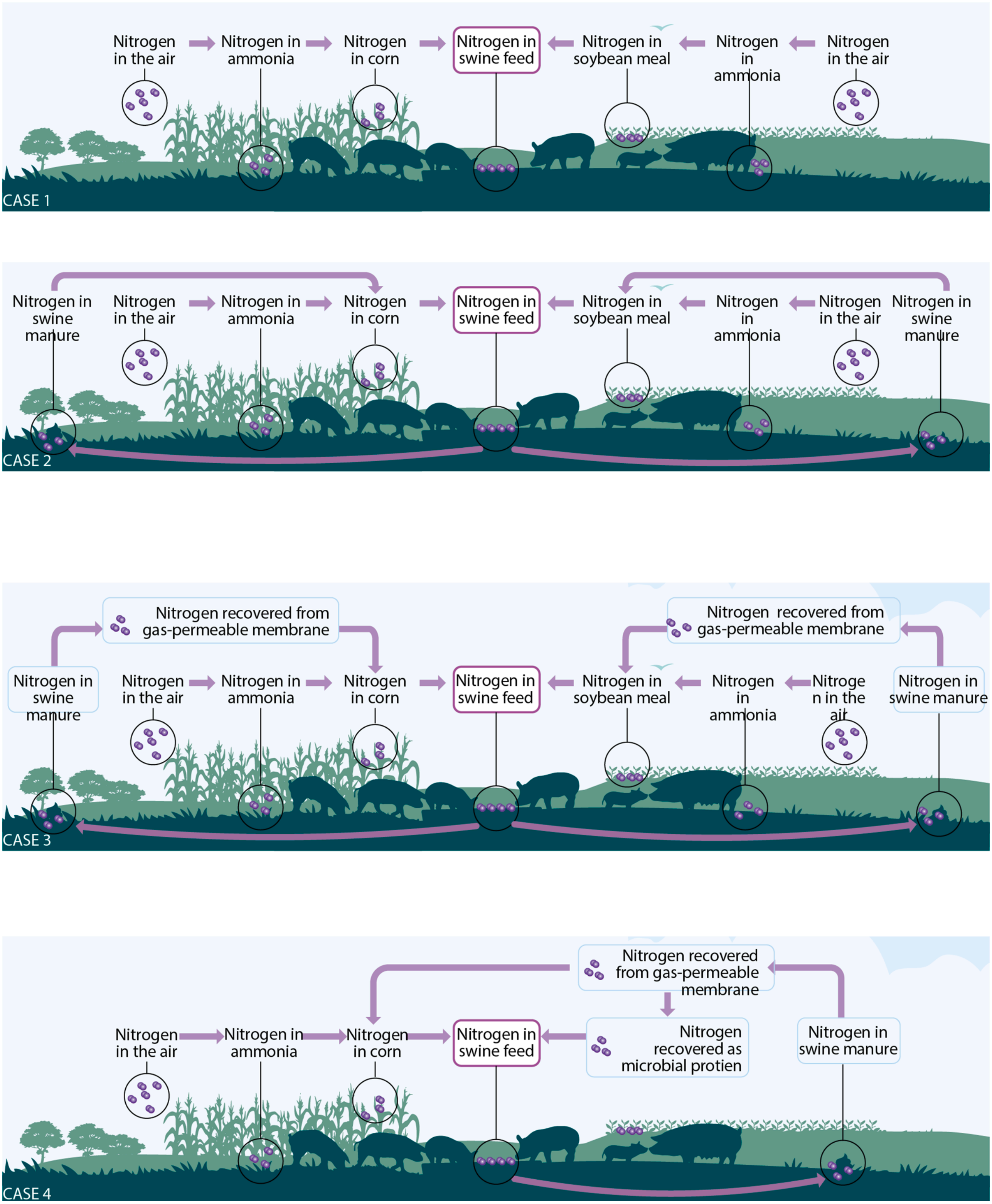

Fig. 2. Nitrogen flows in the four cases we considered: traditional use of synthetic nitrogen fertilizer to produce swine feed, use of swine manure fertilizer to produce swine feed, recovery of ammonium sulfate from manure as fertilizer for swine feed, and production of microbial protein from manure for animal feed.

via nitrification and denitrification. 40 wt% of applied nitrogen is lost indirectly through volatilization, leaching, and runoff. Comparatively, organic fertilizer has the same level of direct nitrogen emissions as synthetic fertilizer, but has higher (50 wt%) indirect nitrogen loss.

We conducted a resource analysis for manure in the U.S. to understand how much manure would be available as a source of recoverable nitrogen. Of manure generated in the U.S. (240 wet tons/yr), 28% is applied to crops as fertilizer. 83% of the applied manure is applied to corn, meeting 16% of its nitrogen demand, while 7.5% of the applied manure is applied to soybean farms, meeting about 3% of soybean nitrogen demand (Qin et al., 2015). Clearly, there is an opportunity to recover and use more of the nitrogen in manure in agricultural systems.

To explore a best-case circularity scenario, we assume that 100% of manure is collected and either used directly for land application or used as feedstock for N recovery technologies. Maintaining the same ratios for manure application for corn and soybean, this means the manure would account for 57% and 11% of the nitrogen needed for corn and soybean agriculture, respectively. We see this scenario play out in case two, in which manure is directly applied to farm fields and the remaining nitrogen demand is met with traditional synthetic fertilizer.

Literature-based data informed the recovery rates of the nitrogen in cases three and four in which nitrogen is first recovered from manure, then converted into fertilizer or animal feed. In case three, a gas-permeable membrane removes 82% of nitrogen from manure and transforms 96% of it into ammonium sulfate fertilizer (Filho et al., 2018). Using the same nitrogen application ratios from case two, the recovered nitrogen can meet 44% and 9% of nitrogen fertilizer requirements for corn and soybean farming, respectively. We assume that the 18% of nitrogen the membrane does not recover is recycled. Finally, in case four, we assume that 99% of the nitrogen that the membrane recovers is taken up into cell biomass to form animal-digestible microbial protein. (Matassa et al., 2015) This microbial protein, which has a similar nutritional profile to soybean meal, can completely replace it in a standard swine feed formulation (Matassa et al., 2016). There is enough additional nitrogen recovered to meet 44% of the demand for corn agriculture as in case three.

4. Results and discussion

We report MCI results per kg swine feed for all four cases in Table 3. For each case, the required mass of nitrogen used to fertilize corn and soy is constant (M, 14 kg). However, the amounts of virgin nitrogen (V) and unrecovered nitrogen waste generated (W) vary. V is higher in cases three and four than in case 2 because the nitrogen recovery technologies used in these cases incur some nitrogen losses. Different rates of nitrogen volatilization for organic and synthetic fertilizers influence W values. Organic fertilizers have a higher volatilization rate. W is highest in case two in which the amount of organic fertilizer used is much higher than in cases three and four. We assumed a constant F(x) of 0.9. As a result, it is the interplay between V and M that most influences the MCI in Table 3. Case one is the least circular of the options and therefore has the lowest MCI. Despite a slight increase in generated waste nitrogen, case two's MCI is 62% higher than case one's because of the large decrease in consumed virgin nitrogen. If low-emission storage and application manure technologies (Bittman et al., 2014) were used, case two's MCI score could increase. Going from case two to case three, the MCI drops 5% due to the increased amounts of virgin nitrogen required to produce the animal feed. As nitrogen recovery technologies continue to become more efficient and recover higher amounts of nitrogen, MCIs for fertilizer products that use this technology could become greater than MCIs for manure-based systems.

The MCI score can also be used as a basis to compare the nitrogen circularity potential of different nitrogen recovery technologies. The MCI for case four is only 2% greater than case three's MCI. Although the introduction of microbial protein removes both the need for fertilizer for soybean agriculture and the nitrogen emissions associated with fertilizer use, because soybean meal makes up a small fraction of the swine feed formulation, replacing meal has a small effect on the MCI. As in case three, increasing the efficiency of nitrogen recovery technologies would increase the MCI. In all cases, further reduction of unrecoverable nitrogen from the use of fertilizers in corn agriculture could improve the MCI score. The application of controlled-release fertilizers, which emit less nitrogen compared to traditional synthetic fertilizers, is one possible solution (Shoji et al., 2001). Although the approaches to circularity in cases 2–4 are fairly different, their MCI scores for these cases are similar. From a circularity perspective, not much advantage is gained by extracting N from manure before using it. Claims that new or better technology enhances circularity should therefore be evaluated with indicators like the MCI.

Table 3
MCI per kg swine feed results for the four case studies.

	Case 1	Case 2	Case 3	Case 4
Mass of nitrogen required for corn and soy agriculture (M, g)	14	14	14	14
Virgin nitrogen used (V, g)	14	6.3	8.1	7.8
Waste nitrogen generated (W, g)	5.7	6.5	5.7	5.6
LFI	0.73	0.36	0.48	0.47
MCI	0.37	0.60	0.57	0.58

4.1. Developing nitrogen recovery technologies to improve nitrogen circularity

As nitrogen recovery technologies evolve and improve, it will be essential to quantify their impact on nitrogen circularity to help prioritize research and development efforts. As an example of these technological advances, Kogler et al. (2021) reviewed 133 different biological and physiochemical technologies at various technology readiness levels capable of removing or recovering nitrogen from wastewater treatment systems. Some technologies like ion exchange columns have already been proven to work at scale, while others like iron-based autotrophic denitrification are at the experimental proof-of-concept stage. Scientists and engineers continue to develop many of these technologies, a subset of which will enter the market and increase opportunities to make products from waste nitrogen. Eventually, many of these technologies may be able to recover nitrogen animal manure or food waste in addition to wastewater. Other potential waste streams from which nitrogen could be recovered are N_2O and NO_x emissions.

Improved nitrogen recovery technologies also present an opportunity to expand the portfolio of products made from recovered waste nitrogen beyond fertilizer. Recovered nitrogen in the form of ammonia can be inputs for the production of other nitrogen containing chemicals such as nitric acid. Moreover, recovered nitrogen can be used to produce cell biomass that contains microbial protein suitable for animal feed (as in case four), or into N_2O which can be used as an oxidant for increased energy production (Scherson et al., 2014). It could also be used as a feedstock for chemical processes that convert benzene to phenol (Uriarte et al., 1997). As technologies evolve to capture the valuable nitrogen in a variety of waste streams, many combinations of source nitrogen, recovery technology, and target product are possible. Applying a circularity indicator like MCI to these combinations across industries would highlight which combinations best advance the circular nitrogen economy.

4.2. Combining MCI with life cycle assessment (LCA) and techno-economic analyses (TEA)

One of the shortcomings of the MCI is that it struggles to provide a complete view of a product's sustainability. It centers on mass flows of the material under study and omits economic viability of circular nitrogen products and their environmental effects beyond waste reduction. For example, waste nitrogen-based pathways may be less energy-intensive than conventional pathways, which is an important benefit to quantify and use in decision-making. For instance, Kar et al., (2023) concluded that ammonium sulfate that is produced from ammonia air stripping of wastewater produced six times less CO_2 intensive compared to the Haber-Bosch pathway. An isolated indicator like MCI doesn't capture this benefit.

However, pairing MCI with life cycle assessment (LCA) can lend insight into how changes in circularity influence key environmental metrics such as greenhouse gas emissions or water and energy intensity. Glogic et al. (2021) completed a case study on alkaline batteries using LCA and MCI, suggesting that improving circularity generally reduces the environmental burden of the product. However, Lonca et al. (2018) completed a case study on tires using LCA and MCI and determine that increasing the MCI through increasing the use of recycled material did not always reduce environmental burdens. While indicators to advance the circular economy are necessary, it is important to account for increasing demand for products, which may drive increased production and influence land use among other types of resource consumption. Clearly, circular is not always better. For waste nitrogen recovery pathways, pairing LCA and MCI could be particularly important so that differences in energy and overall efficiency are evaluated alongside circularity. In the event that similar MCI scores arise (e.g., cases three and four in our analysis), LCA can serve as an important, nuanced tie-breaker.

Different approaches to achieving nitrogen circularity will also vary in cost. MCI on its own does not capture the economic impacts of improving circularity. Completing a techno-economic analysis (TEA) alongside MCI calculations can guide the development of technologies that use wastes, including waste nitrogen, as feedstocks toward economic competitiveness. Braakman et al. (2021) combined circularity and life cycle cost to show that the circularity of a one-family home could be doubled without increasing its cost. Specific circularity improvements included replacing virgin material with recycled materials and using parts that could be easily disassembled. However, any further increase in circularity resulted in a large increase in product cost. Overall, understanding the changes in capital cost, operation cost, and revenue for implementing technologies to improve nitrogen circularity in a product can help provide clarity on what the overall economic impact is per unit of nitrogen that is recovered and used. Using life cycle assessment, techno-economic analysis and material circularity indicators, a robust framework can be developed to guide technology development to improve nitrogen circularity (Fig. 3).

As the number of nitrogen recovery technologies grows, many factors will influence which is the most appropriate to employ in a specific scenario. Completing an LCA, TEA, and calculating an MCI might increase the time and complexity analyses to guide decision making. Yet these steps help provide a complete view of which combinations of waste nitrogen sources, conversion technologies, and product options have the largest potential to improve nitrogen circularity while reducing environmental impact in the most cost-effective manner.

4.3. Limitations and further improvements

Adopting the MCI to evaluate products derived from waste nitrogen can allow for a more detailed understanding of how different technologies and pathways can improve the nitrogen circular economy. Yet, the MCI faces several limitations that curtail its utility.

First, the MCI could be expanded to address indirect sources of waste nitrogen within a product's life cycle. In cases three and four, for example, electricity would be consumed to operate pumps that push the manure through the gas-permeable membrane. If this electricity comes from power plants that combust fossil fuels or even biomass, it may be desirable to account for N_2O emitted during electricity generation as a component of unrecoverable waste.

Second, different forms of unrecoverable nitrogen pollution (included in the term W) are treated equally despite differences in the extent of environmental damage they cause. For example, unrecoverable, benign N_2 is treated in the same manner as nitrogen pollution, including N_2O, a potent greenhouse gas. Furthermore, as currently applied, the MCI does not capture that N_2 is ultimately recoverable whereas reactive nitrogen is not. As a result, the MCI does not guide engineers toward an emphasis on reducing specific types of unrecoverable waste that pose the greatest environmental risks. Further development of the MCI might include a framework that provides unique weights for different forms of nitrogen pollution that reflect their degree of recovery and their environmental effects. The latter factor could potentially be based on the economic impacts of their emissions such as

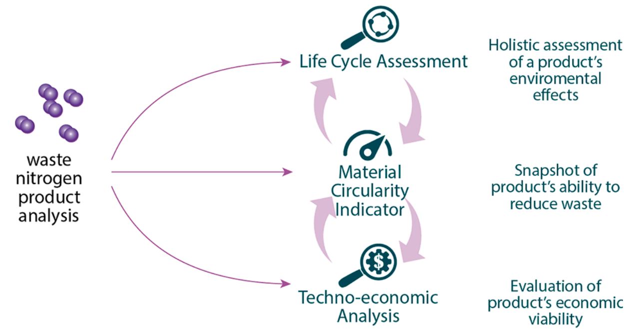

Fig. 3. Relationship between MCI, LCA, and TEA in the guidance of technology development for improved nitrogen circularity.

the recently-proposed social cost of N_2O (EPA 2022).

Next, the utility factor's role in circular nitrogen systems needs to be explored. It can capture differences in product lifetime, which is not necessarily relevant in the case studies we considered. However, if there were nutritional differences in animal feed from microbial sources in case four as compared to animal feed, the utility factor could potentially be used to capture that effect. If polymers or other materials that could exhibit differences in key properties are made from waste nitrogen, the utility factor would play a more important role.

Finally, we note that because we are emphasizing the assessment of circular nitrogen systems, this manuscript does not address the co-benefits of using manure as a soil amendment including, for example, the potential to boost soil carbon levels (Qin et al., 2018).

5. Conclusion

Using MCI to quantify circularity of waste nitrogen utilization pathways can allow for improved nitrogen management. Policymakers can use the MCI to guide policies that target increased nitrogen circularity. Different sectors that produce large quantities of reactive nitrogen can monitor their progress towards nitrogen circularity with the MCI. Developing case studies combining MCI, LCA, and TEA for these different nitrogen pathways should be completed to understand the interrelationship between nitrogen circularity and other sustainability metrics like life-cycle greenhouse gas emissions and water consumption. Case studies should also be completed comparing the circularity of industrial nitrogen-containing products from using virgin nitrogen from the Haber-Bosch process and recovered nitrogen from waste streams to understand the potential impact of nitrogen circularity in these sectors.

CRediT authorship contribution statement

Chayse M. Lavallais: Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Jennifer B. Dunn:** Conceptualization, Methodology, Formal analysis, Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jennifer B. Dunn reports financial support was provided by National Science Foundation. Chayse Lavallais reports financial support was provided by National Science Foundation.

Data availability

We cite all publicly available data sources used in our analysis.

Acknowledgments

Chayse Lavallais and Jennifer Dunn acknowledge support from the National Science Foundation Division of Chemical, Biological, Environmental, and Transport Systems Division (NSF CBET-2033793). We thank Anita Magriplis for assistance with figures.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2023.107167.

References

Apodaca, L., 2018. Minerals Yearbook: Nitrogen. United States Geological Survey, Washington D.C., USA.
Argonne National Laboratory, 2022. GREET® Model: The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) Model. Energy Systems Argonne National Laboratory, Lemont, IL, USA.
Bian, Z., Tian, H., Yang, Q., Xu, R., Pan, S., Zhang, B., 2020. Production and application of manure nitrogen and phosphorus in the United States since 1860. Biosph. Biogeosci. https://doi.org/10.5194/essd-2020-185.
Bittman, S., Dedina, M., Howard, C.M., Oenema, O., Sutton, M.A., 2014. Options For Ammonia Mitigation: Guidance from the UNECE Task Force On Reactive Nitrogen. Centre for Ecology and Hydrology, Edinburgh, UK.
Braakman, L., Bhochhibhoya, S., de Graaf, R., 2021. Exploring the relationship between the level of circularity and the life cycle costs of a one-family house. Resour. Conserv. Recycl. 164, 105149 https://doi.org/10.1016/j.resconrec.2020.105149.
Centner, T.J., 2012. Regulating the land application of manure from animal production facilities in the USA. Water Policy 14 (2), 319–335. https://doi.org/10.2166/wp.2011.086.
Cobo, S., Dominguez-Ramos, A., Irabien, A., 2018. Trade-offs between nutrient circularity and environmental impacts in the management of organic waste. Environ. Sci. Technol. 52 (19), 10923–10933. https://doi.org/10.1021/acs.est.8b01590.
de Oliveira, C.T., Dantas, T.E.T., Soares, S.R., 2021. Nano and micro level circular economy indicators: assisting decision-makers in circularity assessments. Sustain. Prod. Consump. 26, 455–468. https://doi.org/10.1016/j.spc.2020.11.024.
Elia, V., Gnoni, M.G., Tornese, F., 2017. Measuring circular economy strategies through index methods: a critical analysis. J. Clean. Prod. 142, 2741–2751. https://doi.org/10.1016/j.jclepro.2016.10.196.
Ellen MacArthur Foundation and Granta Design, 2015. Ellen MacArthur foundation and Granta design. Circularity indicators - an approach to measuring circularity – methodology.
EPA. 2023. Inventory of U.S. greenhouse gas emissions and sinks: 1990-2021. U.S. Environmental Protection Agency, EPA 430-D-23-001. https://www.epa.gov/ghgemissions/draft-inventory-us-greenhouse-gas-emissions-10 and-sinks-1990-2021.
EPA. 2022. Report on the social cost of greenhouse gases: estimates incorporating recent scientific advances. Environmental Protection Agency, EPA-HQ-OAR-2021-0317. https://www.epa.gov/system/files/documents/2022-11/epa_scghg_report_draft_0.pdf.
European Commission. 1991. Council directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment.
European Commission, 2018. A European Strategy for Plastics in a Circular Economy. European Commission, Brussels, Belgium.
Fernandez-Mena, H., Gaudou, B., Pellerin, S., MacDonald, G.K., Nesme, T., 2020. Flows in Agro-food Networks (FAN): an agent-based model to simulate local agricultural material flows. Agric. Syst. 180, 102718 https://doi.org/10.1016/j.agsy.2019.102718.
Garza-Reyes, J.A., Salomé Valls, A., Peter Nadeem, S., Anosike, A., Kumar, V., 2019. A circularity measurement toolkit for manufacturing SMEs. Int. J. Prod. Res. 57 (23), 7319–7343. https://doi.org/10.1080/00207543.2018.1559961.
Glogic, E., Sonnemann, G., Young, S.B., 2021. Environmental trade-offs of downcycling in circular economy: combining life cycle assessment and material circularity indicator to inform circularity strategies for alkaline batteries. Sustainability 13 (3), 1040. https://doi.org/10.3390/su13031040.
Goyal, S., Chauhan, S., Mishra, P., 2021. Circular economy research: a bibliometric analysis (2000–2019) and future research insights. J. Clean. Prod. 287, 125011 https://doi.org/10.1016/j.jclepro.2020.125011.
Harder, Robin, Giampietro, Mario, Smukler, Sean, 2021. Towards a circular nutrient economy. A novel way to analyze the circularity of nutrient flows in food systems. Resour. Conserv. Recycl. 172 https://doi.org/10.1016/j.resconrec.2021.105693.
IFEEDER, 2020. Animal Feed/Food Consumption and COVID-19 Impact Analysis. Institute for Feed Education and Research.
IPCC. 2006. Chapter 11: N2O emissions from managed soils, and CO2 emissions from lime and urea application.
Kakwani, Nikita, S., Kalbar, Pradip P., 2022. Measuring urban water circularity: development and implementation of a water circularity indicator. Sustain. Prod. Consump. 31 (May), 723–735. https://doi.org/10.1016/j.spc.2022.03.029.
Kar, S., Singh, R., Gurian, Patrick, L., Hendricks, A., Kohl, P., McKelvey, S., Spatari, S., 2023. Life cycle assessment and techno-economic analysis of nitrogen recovery by ammonia air-stripping from wastewater treatment. Sci. Total Environ. 857 https://doi.org/10.1016/j.scitotenv.2022.159499.
Kirchherr, J., Reike, D., Hekkert, M., 2017. Conceptualizing the circular economy: an analysis of 114 definitions. Resour. Conserv. Recycl. 127, 221–232. https://doi.org/10.1016/j.resconrec.2017.09.005.
Kogler, A., Farmer, M., Simon, J.A., Tilmans, S., Wells, G.F., Tarpeh, W.A., 2021. Systematic evaluation of emerging wastewater nutrient removal and recovery technologies to inform practice and advance resource efficiency. ACS ES&T Eng. 1 (4), 662–684. https://doi.org/10.1021/acsestengg.0c00253.
Kyriakou, V., Garagounis, I., Vourros, A., Vasileiou, E., Stoukides, M., 2020. An electrochemical Haber-Bosch process. Joule 4 (1), 142–158. https://doi.org/10.1016/j.joule.2019.10.006.
Li, B., Brett, M.T., 2012. The impact of alum based advanced nutrient removal processes on phosphorus bioavailability. Water Res. 46 (3), 837–844. https://doi.org/10.1016/j.watres.2011.11.055.
Liu, J., Ma, K., Ciais, P., Polasky, S., 2016. Reducing human nitrogen use for food production. Sci. Rep. 6 (1), 30104. https://doi.org/10.1038/srep30104.
Lonca, G., Lesage, P., Majeau-Bettez, G., Bernard, S., Margni, M., 2020. Assessing scaling effects of circular economy strategies: a case study on plastic bottle closed-loop recycling in the USA PET market. Resour. Conserv. Recycl. 162, 105013 https://doi.org/10.1016/j.resconrec.2020.105013.
Lonca, G., Muggéo, R., Imbeault-Tétreault, H., Bernard, S., Margni, M., 2018. Does material circularity rhyme with environmental efficiency? Case studies on used tires. J. Clean. Prod. 183, 424–435. https://doi.org/10.1016/j.jclepro.2018.02.108.
Matassa, S., Boon, N., Pikaar, I., Verstraete, W., 2016. Microbial protein: future sustainable food supply route with low environmental footprint. Microb. Biotechnol. 9 (5), 568–575. https://doi.org/10.1111/1751-7915.12369.
Matassa, S., Batstone, D.J., Hülsen, T., Schnoor, J., Verstraete, W., 2015. Can direct conversion of used nitrogen to new feed and protein help feed the world? Environ. Sci. Technol. 49 (9), 5247–5254. https://doi.org/10.1021/es505432w.
Miksch, K., Sikora, J., 2012. Wastewater Biotechnology (In Polish). Polish Scientific Publishers PWN.
J. de S. Filho, Oliveira, Daguerre-Martini, S., Vanotti, M.B., Saez-Tovar, J., Rosal, A., Perez-Murcia, M.D., Bustamante, M.A., Moral, R, 2018. Recovery of ammonia in raw and co-digested swine manure using gas-permeable membrane technology Front. Sustain. Food Syst. 2, 30. https://doi.org/10.3389/fsufs.2018.00030.
Pandey, B., Chen, L., 2021. Technologies to recover nitrogen from livestock manure—a review. Sci. Total Environ. 784, 147098 https://doi.org/10.1016/j.scitotenv.2021.147098.
Papangelou, A., Achten, W.M.J., Mathijs, E., 2020. Phosphorus and energy flows through the food system of Brussels Capital Region. Resour. Conserv. Recycl. 156, 104687 https://doi.org/10.1016/j.resconrec.2020.104687.
Preisner, M., Smol, M., Horttanainen, M., Deviatkin, I., Havukainen, J., Klavins, M., Ozola-Davidane, R., Kruopienė, J., Szatkowska, B., Appels, L., Houtmeyers, S., Roosalu, K., 2022. Indicators for resource recovery monitoring within the circular economy model implementation in the wastewater sector. J. Environ. Manage. 304, 114261 https://doi.org/10.1016/j.jenvman.2021.114261.
Qin, Z., Canter, C.E., Dunn, J.B., Mueller, S., Kwon, H., Han, J., Wander, M.M., & Wang, M. 2015. "Incorporating agricultural management practices into the assessment of soil carbon change and life-cycle greenhouse gas emissions of corn stover ethanol production." ANL/ESD–15/26, 1221938. https://doi.org/10.2172/1221938.
Qin, Z., Canter, C.E., Dunn, J.B., Mueller, S., Kwon, H., Han, J., Wander, M.M., Wang, M., 2018. Land management change greatly impacts biofuels' greenhouse gas emissions. GCB Bioenergy 10, 370–381. https://doi.org/10.1111/gcbb.12500.
Rocchi, L., Paolotti, L., Cortina, C., Fagioli, F.F., Boggia, A., 2021. Measuring circularity: an application of modified material circularity indicator to agricultural systems. Agric. Food Econ. 9 (1), 9. https://doi.org/10.1186/s40100-021-00182-8.
Rossi, E., Bertassini, A.C., Ferreira, C.dos S., Neves do Amaral, W.A., Ometto, A.R, 2020. Circular economy indicators for organizations considering sustainability and business models: plastic, textile, and electro-electronic cases. J. Clean. Prod. 247, 119137 https://doi.org/10.1016/j.jclepro.2019.119137.
Shoji, S., Delgado, J., Mosier, A., Miura, Y., 2001. Use of controlled release fertilizers and nitrification inhibitors to increase nitrogen use efficiency and to conserve air and water quality. Commun. Soil Sci. Plant Anal. 32 (7–8), 1051–1070. https://doi.org/10.1081/CSS-100104103.
Scherson, Y.D., Woo, S.G., Criddle, C.S., 2014. Production of nitrous oxide from anaerobic digester centrate and its use as a co-oxidant of biogas to enhance energy recovery. Environ. Sci. Technol. 48 (10), 5612–5619. https://doi.org/10.1021/es501009j.
Scherson, Y.D., Wells, G.F., Woo, S.G., Lee, J., Park, J., Cantwell, B.J., Criddle, C.S., 2013. Nitrogen removal with energy recovery through N_2O decomposition. Energy Environ. Sci. 6 (1), 241–248. https://doi.org/10.1039/C2EE22487A.
Shaddel, S., Ucar, S., Andreassen, J.P., Østerhus, S.W., 2019. Enhancing efficiency and economics of phosphorus recovery process by customizing the product based on sidestream characteristics – an alternative phosphorus recovery strategy. Water Sci. Technol. 79 (9), 1777–1789. https://doi.org/10.2166/wst.2019.178.
Tadesse, S.T., Oenema, O., van Beek, C., Ocho, F.L., 2019. Nitrogen allocation and recycling in peri-urban mixed crop–livestock farms in Ethiopia. Nutrient Cycl. Agroecosyst. 115 (2), 281–294. https://doi.org/10.1007/s10705-018-9957-z.
United Nations Environmental Program. 2022. Historic day in the campaign to beat plastic pollution: nations commit to develop a legally binding agreement. United Nations Environmental Program. https://www.unep.org/news-and-stories/press-release/historic-day-campaign-beat-plastic-pollution-nations-commit-develop.
Uriarte, A.K., Rodkin, M.A., Gross, M.J., Kharitonov, A.S., Panov, G.I., 1997. Direct hydroxylation of benzene to phenol by nitrous oxide. In: Studies in Surface Science and Catalysis, 110. Elsevier, pp. 857–864. https://doi.org/10.1016/S0167-2991(97)81048-8.

US Plastic Pact, 2020. U.S Plastics Report 2020 Baseline Report. The Recycling Partnership and World Wildlife Fund.

Van Loon, Marloes P., Wytse, J.Vonk, Renske Hijbeek, Van Ittersum, Martin K., Ten Berge, Hein F.M., 2023. Circularity indicators and their relation with nutrient use efficiency in agriculture and food systems. Agric. Syst. 207 https://doi.org/10.1016/j.agsy.2023.103610.

Velasco-Muñoz, J.F., Mendoza, J.M.F., Aznar-Sánchez, J.A., Gallego-Schmid, A., 2021. Circular economy implementation in the agricultural sector: definition, strategies and indicators. Resour. Conserv. Recycl. 170, 105618 https://doi.org/10.1016/j.resconrec.2021.105618.

Verberne, J.J., 2016. Building Circularity Indicators: an Approach for Measuring Circularity of a Building. Technische Universiteit Eindhoven.

Wang, Z., Hellweg, S., 2021. First steps toward sustainable circular uses of chemicals: advancing the assessment and management paradigm. ACS Sustain. Chem. Eng. 9 (20), 6939–6951. https://doi.org/10.1021/acssuschemeng.1c00243.

Xu, X., Sharma, P., Shu, S., Lin, T.S., Ciais, P., Tubiello, F.N., Smith, P., Campbell, N., Jain, A.K., 2021. Global greenhouse gas emissions from animal-based foods are twice those of plant-based foods. Nature Food 2 (9), 724–732. https://doi.org/10.1038/s43016-021-00358-x.

Xue, M., Lin, B., Tsunemi, K., Minami, K., Nanba, T., Kawamoto, T., 2021. Life cycle assessment of nitrogen circular economy-based NO_x treatment technology. Sustainability 13 (14), 7826. https://doi.org/10.3390/su13147826.