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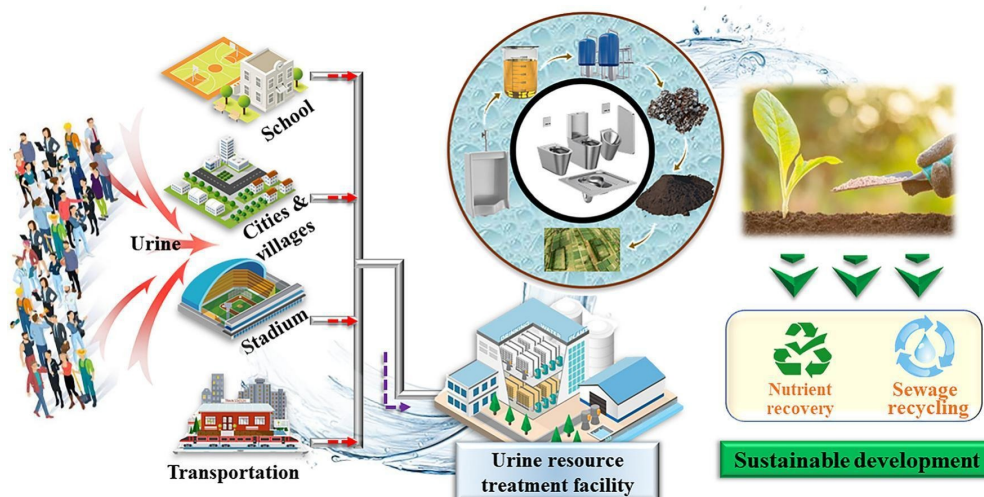
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Integrated processes for simultaneous nitrogen, phosphorus, and potassium recovery from urine: A review

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A B S T R A C T Urine, traditionally regarded as liquid waste, contains valuable elements – nitrogen (N), phosphorus (P), and potassium (K) which can be repurposed as crop fertilizers. Under the context of dwindling resources, recent researches have concentrated on the development of advanced methods recovering valuable compounds from urine. This review explores urine concentration technologies for recovering N, P, and K fertilizers from urine, including alkaline dehydration, forward osmosis, membrane distillation, and freeze concentration. Furthermore, the paper focuses on advanced technologies, combined systems, and simultaneous recovery sequences tailored to different application scenarios for recovering specific nutrient elements (N, P, and K) simultaneously. In contrast to other relevant articles on urine nutrient recovery, this study provides a thorough overview of key technologies for nitrogen, phosphorus, and potassium recovery from urine, offering diverse recovery process options based on specific recycling needs. The central focus of this paper lies in the systematic integration of diverse technologies to achieve the simultaneous recovery of multiple resources from urine. It delves into optimal recovery sequences and process coupling strategies for individual recoveries. In summary, this paper presents innovative strategies for the simultaneous recovery of N, P, and K in urine, addressing the pressing need for sustainable resource management.

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Urine
Multi-resource
Simultaneous recovery
Fertilizer
Combined processes

1. Introduction

The management of source-separated urine has gained significant attentions [1]. This surge in interest arises from the recognition of urine's dual role—as a liquid waste leading to pollution control and a valuable resource for energy and nutrient recovery [2]. A landmark publication in Nature in 2022 underscored that the escalating global demand for fertilizers, spurred by population growth, necessitates a sustainable approach to recycle and reuse crucial fertilizer components, including Nitrogen (N), Phosphorus (P), and Potassium (K) [3]. Recycling urine emerges as an optimal solution, not only curbing waterway pollution by these essential nutrients but also providing a sustainable source of fertilizer.

Human urine is regarded as a typical liquid waste [4]. According to reports, 80 % of N, 50 % of P, and 60 % of K found in domestic sewage are produced by human urine, although urine only accounts for 1 % in volume of the total domestic sewage [5,6]. N, P, and K are essential

nutrients for plant growth and development [7]. They are components of many essential organic compounds in plants and are involved in plant metabolism in various forms. Currently, the production of commercial nitrogen fertilizer mostly uses the Haber-Bosch method. This process uses hydrogen to fix nitrogen in liquefied air under high temperatures and high pressure for ammonia synthesis, which consumed about 1 % of the world's energy [8,9]. With a current world population of 7.5 billion, the resources loss to wastewater discharges are roughly 25 metric tons of N, 1.8 metric tons of P, and 5.5 metric tons of K per year [10]. Since most of these energy sources come from fossil fuels, greenhouse gas emissions have greatly increased [11]. The current global industrial production of reactive N is approximately 120 tons per year. However, the amount of N excreted in urine worldwide is as high as 30 tons per year, so recycling N from urine can significantly reduce the demand for industrially produced reactive N [12].

Most of the P used in fertilizers comes from phosphate reserves being depleted rapidly [13,14]. At the current consumption rate of P, global

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phosphorite reserves are depleting rapidly [15,16]. In addition, the mining of phosphate rock also generates waste. These wastes cause pollution, and the generated sludge will usually be accumulated in the landfill, requiring a larger space for storage [17]. Approximately 3 million tons out of the 14 million tons of mineral P used in global agriculture enters the wastewater system through human excretion [18]. Thus, recovery of P from human excretion is also imperative to promote sustainable use of P. Another crucial agricultural fertilizer K, predominantly originates from potash mines. Some countries, including China, the United States, India, and Brazil, face a significant disparity between the production of industrial potash fertilizer and the demand for agricultural development [19]. Especially in southern China, the K content of the soil is rapidly decreasing due to rainfall leaching. The lack of K in the soil has been a major constraint for crop production in several regions. The uneven distribution of global K ore poses a major risk for agricultural production in countries where K fertilizers are deficient [20]. Therefore, the recovery of K from urine carries profound economic and agricultural significance. In summary, the extraction and recovery of N, P, and K from urine offer multifaceted benefits. This approach not only reduces the cost of domestic sewage treatment but also facilitates wastewater recycling, diminishes the reliance on industrial fertilizers in agriculture, and effectively mitigates water body eutrophication while addressing resource scarcity challenges.

Currently, most articles on the recovery of N, P, and K from urine primarily focus on the recovery of individual elements [18,21], or the combined recovery of N P [22], or the recovery using specific technologies [23,24]. Few comprehensively address the recovery of N, P, and K elements together, and even fewer delve into the synchronized recovery aspect. Given the resource-rich nature of urine, singular recovery of one or two elements leads to resource inefficiency and wastage. This paper distinguishes itself from the previous reviews by prioritizing synchronous recovery of resources, emphasizing the compatibility of diverse technologies for mutual enhancement, and evaluating the outcomes of synchronized resource recovery following process coupling.

1.1. Systematic literature review methodology

Relevant articles were gathered from the Web of Science and Google Scholar databases using search terms such as “urine nitrogen and phosphorus recovery”, “urine nutrients recovery”, “urine potassium recovery”, and “urine fertilizer concentrate”, separated by the Boolean operator “OR.” Retrieve relevant research papers from the years 2010 to 2023, excluding patents, unpublished manuscripts, abstracts, and theses. The selected literature must focus on the treatment of human urine and emphasize the recovery of N, P, and K elements, as well as processes for simultaneous recovery. A multi-layered filtration process was implemented, and a systematic review established strict inclusion and exclusion criteria to encompass the most relevant articles, with the verification process shown in Fig. S1.

2. Composition of urine

Human urine contains low-risk compounds and heavy metals [25] and is rich in essential nutrients for plant life, making it a fertilizer with significant application potential [26]. The high-concentration selective nutrient recovery of urine has recently attracted increasing attention from the scientific community [27]. To maximize the utilization of valuable nutrients in urine, technologies that enable the collection and separation of urine at the source are gaining recognition and adoption [28].

The chemical composition of urine is intricate [29]. Urine is a fluid product of the body secreted by kidneys. It is 93–96 % composed of water and the rest consists of soluble waste and excess substances of the body such as urea, dissolved ions (e.g. chloride, sodium, and K), creatinine, inorganic and organic compounds, or salts [30]. It encompasses a majority of the nutrients excreted by the human body, including 87 % of

N (with 75–90 % from urea), 50 % of P, and 12 % of organic matter [31,32]. The volume and composition of urine vary among individuals due to their physical exertion, survival environment, and intake of water, salt, and high protein [33]. Therefore, it is difficult for synthetic urine to completely mimic the composition of real urine. Although using synthetic urine for research is beneficial for the development of new technologies, more emphasis should be placed on studying real urine or using real urine to verify experimental results. Besides, the characteristics of urine are affected by many factors: gender, elapsed time after collection, temperature, etc.

The characteristics of fresh urine are also influenced by individual differences [34]. The fresh urine used by researchers generally refers to the urine just collected, or immediately frozen after collection with treatment of lower pH. Hydrolyzed urine refers to urine with urea been completely broken down after being stored [35]. Table 1 provides a list of common organic and inorganic chemicals in fresh urine and hydrolyzed urine. All the data quoted in the table were taken from real urine, which varies in composition and structure in different regions due to their dietary structure and climate, among other factors. During hydrolysis of urine, Ca^{2+} and Mg^{2+} spontaneously precipitate in combination with PO_4^{3-} , Ca^{2+} and Mg^{2+} and the concentration of PO_4^{3-} is reduced.

Approximately 85 % of the N in urine initially exists in the form of urea, and 5 % exists as ammonium (NH_4^+) or ammonia (NH_3) [44]. During the hydrolysis of urine, urea can be hydrolyzed into NH_3 , the conjugate base of NH_4^+ . The pH of fresh urine typically ranges from 6.0 to 7.0. After hydrolysis of urine, the pH of the urine rises to about 9.2 [44]. P exists in the form of phosphate ions, showing an acid-base relationship with phosphoric acid. At a pH of 2.0–7.0, P primarily exists in the form of H_2PO_4^- . When the pH is 7.0–11.0, P mainly exists as HPO_4^{2-} . At a pH >12.0, P primarily exists as PO_4^{3-} [45]. K is a reactive alkali metal that exists in urine in the form of K^+ .

K is a monovalent cation that is independent of pH, P is a mono/di/trivalent anion depending on pH, and N can be dissolved gas or monovalent cation depending on pH. Thus, pH is critical for its impact on the separation/recovery processes. When urine is excreted from the body, it contains a substantial amount of nitrogen in the form of urea [46]. Urease-positive bacteria hydrolyze urea into ammonium and bicarbonate during urine storage [11,47], as shown in Eq. (1). The concentration of NH_4^+ in hydrolyzed urine, as indicated in Table 1, was significantly higher than that in fresh urine from samples collected by various researchers. Therefore, pre-recovery of NH_4^+ is recommended when extracting nutrients from water.



In the reaction equation, ΔG represents the Gibbs free energy, and the Gibbs free energy of each substance is shown in Table S1.

During the hydrolysis of urea, the pH of the urine increases from 6.0 to about 9.0, which is more suitable for resource recovery processes [48]. As the pH rises, some pathogens contaminated with feces are killed, spontaneously with calcium and magnesium (Mg) in the urine precipitated [49,50]. Through the hydrolysis process, volatile ammonia is fixed, making it more amenable to reaction and separation from other substances. This facilitates the recovery and utilization of elemental N. Due to the difference in the chemical composition of fresh urine and stored urine, hydrolyzed urine should be used when recovering N, P, and K in the urine [44].

In addition to hydrolyzing urine, many researchers, in order to inhibit the hydrolysis of urea and the volatilization of NH_3 [51], employ various pre-treatment technologies to stabilize urine and maximize the recovery of nutrients. There are numerous stabilization methods treating urine, such as acid addition [52], calcium hydroxide addition [53], nitrification [54], electrochemical treatment [55], heat-activated peroxydisulfate addition [56], and air and CO_2 bubbling [57]. Compared to hydrolyzed urine, stabilized urine is more conducive to the recovery of

Table 1

Comparison of the composition of fresh and hydrolyzed urine.

Fresh urine							Hydrolyzed urine						
Parameter	Unit	A	B	C	D	E	Mean \pm SD*	F	D	E	H	J	Mean \pm SD*
pH	–	5.6	6.3	5.6	6.6	4.2	5.66 \pm 0.93	9.1	9.1	8.9	9.3	9.3	9.14 \pm 0.168
NH ₄ ⁺ -N	mg L ⁻¹	382	436	438	341	443	408.00 \pm 44.93	3400	4200	5615	3700	2393	3861.6 \pm 1181.62
PO ₄ ³⁻ -P	mg L ⁻¹	1300	476	388	332	437	586.60 \pm 402.43	140	174	384	223	121	208.4 \pm 105.53
K ⁺	mg L ⁻¹	1810	1240	1870	529	1181	1326.00 \pm 546.31	1500	1611	1166	1900	446	1324.6 \pm 556.99
Na ⁺	mg L ⁻¹	2440	1856	3240	2533	2300	2473.80 \pm 500.90	1850	2211	2300	2400	1185	1989.2 \pm 495.14
Mg ²⁺	mg L ⁻¹	32	48	45.4	50	49	44.88 \pm 7.40	–	–	28	0.9	6.4	11.77 \pm 14.32
Ca ²⁺	mg L ⁻¹	58	82	89.2	47	293	113.84 \pm 101.62	–	–	267	17	12.2	98.73 \pm 145.74
SO ₄ ²⁻	mg L ⁻¹	590	1709	878	693	1008	975.60 \pm 440.74	1090	693	1181	800	–	990.5 \pm 231.79
Cl ⁻	mg L ⁻¹	2990	4430	6620	1320	4450	3962.00 \pm 1965.14	3770	1300	4400	3400	–	3900.0 \pm 1343.34
Urea	mg L ⁻¹	2680	4500	4450	9016	6032	5335.60 \pm 2375.13	–	–	–	–	–	–

Fresh urine references:

A fresh urine in Taiwan [36];

B fresh urine from 20 persons, age 25–65 in Sweden [37];

C fresh urine from rural village in Nepa [38];

D fresh urine in Korea [39];

E fresh urine from pregnant women in the Netherlands [40].

Hydrolyzed urine references:

F real urine from volunteers age 18–40 in the Arizona State University [41];

H real urine from male employees in the Netherlands [42];

J real urine from two male toilet in the Beijing, China. [43].

nutrient elements in urine.

However, pre-treatment may affect the combination of recovery processes, increase the overall cost of the recovery process, and the chemicals added in certain stabilization methods may impact the effectiveness of subsequent recovery processes. Researchers need to evaluate whether to pre-treat urine based on aspects such as recovery efficiency, operational stability, and operating costs.

3. Recovering N, P, and K from concentrated urine

Although urine is a rich source of nutrients and can be a valuable fertilizer, there are still some challenges in converting urine into fertilizer. Firstly, urine requires stabilization because urease secreted by microorganisms can rapidly hydrolyze urea into ammonia, and the volatilization of ammonia can lead to a loss of total nitrogen (TN) in the collected urine [58]. Although urine is directly used as fertilizer in various regions, its nutrient imbalance and potential contamination with harmful pollutants, such as hormones and drugs, pose significant challenges for large-scale applications [59]. When considering the need to transport urine to distant farmlands, the associated transportation and storage costs alone can reduce the market value of urine as a fertilizer [60]. Besides, urine consists of 97 % water [61], it is an aqueous, diluted fertilizer compared to chemical fertilizers. This necessitates reducing the volume of urine and maximizing nutrient recovery to achieve efficient storage, transport, and field application [62]. Therefore, researchers have explored methods to concentrate urine, utilizing nutrient extraction techniques to separate nutrients from the influent and produce a concentrated fertilizer [63]. These methods include Alkaline dehydration [64], evaporation [60], freeze concentration [65,66], electro dialysis [67], membrane distillation [68], and forward osmosis [27].

3.1. Recover N, P, and K fertilizers from urine through freeze concentration

Alkaline dehydration can be used to treat human urine and produce a fertilizer that is dry and rich in nutrients. Alkalization is the process by which the addition of base or alkaline media elevates the pH of fresh urine, preventing the enzymatic hydrolysis of urea [53]. The hydrolysis of urea into ammonia occurs due to the catalytic action of the specific enzyme urease [69]. The activity of urease and the enzymatic breakdown of urea can be suppressed through alkalization [70]. It is important to note that base should be added to fresh urine; if added to

hydrolyzed urine, the increase in pH will promote the volatilization of NH₃ [71]. Simha et al. conducted a study on dehydrating fresh urine by adding five different alkaline agents (Wheat bran, biochar, desert soil, calcium hydroxide, and wood ash) at high temperatures (50 and 60 °C). They successfully concentrated the urine by 48 times, yielding a dried end product with high fertilizer value: approximately 10 % N, 1 % P, and 4 % K [72]. Vasiljev et al. added urine to various magnesium-based alkaline substrates (MgO, Mg(OH)₂, MgCl₂ + Mg(OH)₂) and dried them at 38 °C. The resulting product was a solid fertilizer containing approximately 10–11 % N, 1–2 % P, and 2–3 % K. Urine can also be alkalized using an anion exchange resin. Another study by Simha et al. in 2018 developed an ion exchange resin and alkaline substrate coupled device, achieving >90 % reduction in urine volume, >70 % N retention, and 100 % P and K recovery [72]. Simultaneously, Simha et al. conducted on-site tests of an alkaline dehydration pilot-scale system at a military training camp in Finland, marking a significant step towards the widespread application of alkaline dehydration [64]. In addition, Senecal et al. conducted a sanitation assessment which demonstrated that the use of an alkaline dehydration system can concentrate nutrients in urine, eliminate liquids, and produce a dry end product that can be safely used as fertilizer after four days of storage [73].

3.2. Recover N, P, and K fertilizers from urine through forward osmosis and reverse osmosis

Forward osmosis (FO) is driven by the osmotic pressure differential across a selectively permeable membrane which facilitates the transfer of water [74]. As a result of the pressure difference, water molecules from the low-solute concentration solution will diffuse through the semi-permeable membrane into higher solute concentration solution, while the solute remains on the original solution side [75]. FO allows for the transfer of urea and ammonia while inorganic ions, drugs, and organic compounds are effectively rejected, as seen in Fig. 1.

Volpin et al. used Mg²⁺ salts (such as MgSO₄ and Mg(NO₃)₂) as the extraction solution (DS) for dehydrating and combining non-hydrolyzed urine [27]. The process relied on Mg²⁺ reverse salt flux (RSF) to reclaim P as hard gypsum. Simultaneously, urea was retrieved in the DS. The findings indicated that concentrating the urine by 60 % resulted in the retrieval of approximately 40 % of P and 50 % of N. Furthermore, the combination of FO and electrochemical techniques allows for the simultaneous recovery of elemental N and P from urine. Jiang et al. developed the FO integrated bioelectrical concentration recovery

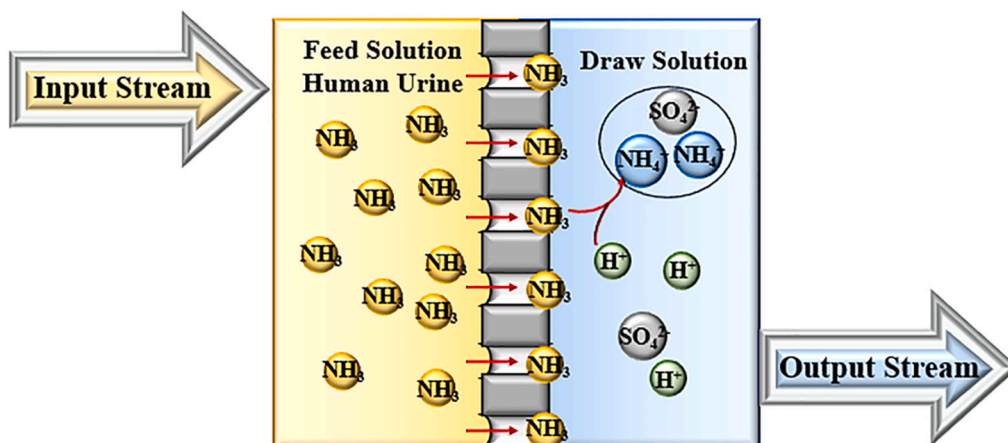


Fig. 1. The diagram of the transfer principle of low molecular weight neutral compounds such as ammonia in the FO system.

system (OsBCRS) by combining FO and bioelectrical concentration [76]. Ultimately, the recovery rate of nitrogen in synthetic urine exceeds 90 %, and the recovery rate of phosphorus exceeds 80 %. The study used synthetic urine, and should further utilize real urine in experiments for better applicability and promotion.

Studies by several other researchers have shown that the FO process can also be used to recover N, P, and K from urine simultaneously. Zhang et al. applied the FO dehydration process to recover nutrients from source-separated urine under various conditions, using seawater or desalinated brine as a low-cost extraction solution [77]. The process showed relatively low removal of neutral organic N (urea nitrogen) from fresh urine, but improved removal of ammonium (50–80 %) from hydrolyzed urine, and high removal of phosphate and K (>90 %) in most cases. Pocock et al. demonstrated that positive osmosis, employing ammonium bicarbonate solution, serves as a closed-loop nutrient recovery technique for source-separated urine [78]. The recovery unit achieved removal rates of up to 95 % of P, 85 % of N and chloride, and 75 % of K and sodium over an eight-hour operating period. A cost-benefit analysis and life cycle assessment conducted by Shi illustrates the economic and environmental advantages of resource-oriented toilets that utilize ortho-osmosis technology for the retrieval of N, P, and K from urine [79].

Apart from FO, reverse osmosis (RO) technology is also employed to recover nutrients from urine [60]. In the RO process, water from urine permeates through a semi-permeable membrane due to the application of pressure higher than the osmotic pressure, thereby achieving the reduction of water content in the urine. Ray et al. employed reverse osmosis and nanofiltration (NF) for the selective separation of free ammonia from hydrolyzed urine. NF enabled the recovery of 90 % of free ammonia, while RO allowed for the recovery of 64 % of free ammonia [80]. Courtney and Randall utilized RO to concentrate and stabilize urine. Ultimately, at a water recovery rate of 60.0 %, 85.5 % of urea and 99.2 % of K were recovered in the saline stream. Additionally, over 99.9 % of P was recovered as a separate solid calcium phosphate fertilizer [62].

3.3. Recover N, P, and K from urine through membrane distillation and nitrification distillation

Membrane distillation is a process driven by temperature gradient in vapor pressure [81]. Low temperature promotes the passage of volatile compounds through the hydrophobic microporous membrane, leading to the concentration of urine and recovery of ammonia due to its high volatility (Fig. 2). To overcome the limitations of conventional molecular distillation for the separation of volatile compounds from aqueous solutions, especially for the recovery of ammonia from hydrolyzed urine, isothermal membrane distillation (IMD) with an acid collector

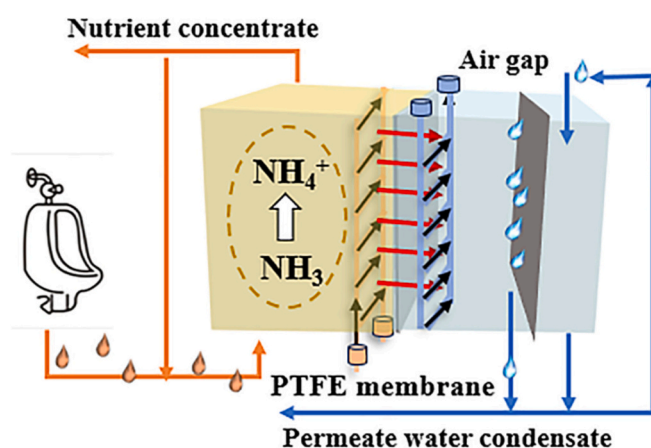


Fig. 2. Schematic diagram of nutrient transport pathways in a membrane distillation system.

was introduced by McCartney et al. [82]. After 6 h of reactor operation, 60 % of the TAN (total ammonia nitrogen) was removed and captured in the collector stream. Furthermore, Tun et al. used a direct contact membrane distillation (DCMD) process to treat acidified and filtered urine, and the TAN was successfully concentrated to $<2.06 \times 10^{-3}$ g-N/g-H₂O [83]. With this treatment, the ammonia transfer rate can be reduced to <5 %, achieving removal efficiency up to 90 %.

Utilizing membrane distillation to obtain liquid fertilizer from concentrated urine can recover N, P, and K simultaneously. Yaping et al. developed a novel system combining thermally activated bisulfate with membrane distillation (TAP-MD) for the simultaneous recovery of nutrients from human urine [84]. The results showed that nearly 92.9 % of urea, 97.4 % of P, and 99.8 % of K were recovered as concentrated liquid fertilizer. Xu et al. increased the concentration of hydrolyzed urine by 17.8-fold through direct contact membrane distillation (DCMD), achieving high phosphate and K removal rates (>97.0 %) [85]. They propose that preliminary biostabilization of the hydrolyzed urine prior to MD can help reduce ammonia loss, thus improving overall ammonia removal (94.0 %).

Liu et al. combined forward osmosis and membrane distillation (FO-MD) to concentrate and treat urine [86]. The FO-MD system significantly reduced the TN and NH₄⁺-N concentrations in the produced water by two orders of magnitude compared to the pure MD system. Gau et al. investigated the recovery of nutrients from human urine using a seawater-driven FO-MD mixing system [87]. The recovery rate of the nutrients in the feed solution was around 85 %, and the effectiveness of rejection and recovery remained unaffected by the draw solution

temperature. These findings suggest that FO-MD based human urine treatment is capable of sustainable utilization. This also implies that co-processing may be more effective for resource recovery than mono-processes.

Udert and Wächter combined biological nitrification and distillation to recover a dry solid containing all nutrients from urine. The solid residue contained a substantial amount of nutrients such as ammonium nitrate, K, sulfate, and phosphate [60]. This also demonstrates that biological nitrification and distillation can be a stable and efficient process to concentrate and recover nutrients from urine. Fumasoli et al. constructed a nitrification/distillation apparatus and tested this combination on a pilot-scale [88]. The resulting concentrate exhibited low heavy metal content but high nutrient content, indicating its suitability as an integrated fertilizer.

3.4. Recover N, P, and K fertilizers from urine through freeze concentration

The freeze concentration technology harnesses the solidification property of water by cooling urine below its freezing point, prompting the formation of ice crystals in the urine solution [89]. By separating the ice crystals from the urine, clean ice and concentrated nutrients are obtained. While evaporation could be used for further concentration, freezing is advantageous as the latent heat of fusion is approximately six times less than the heat of vaporization [90]. According to existing literature and research findings, cold concentration technologies mainly include freeze-thaw [65], block freeze concentration [91], progressive freeze concentration [92], eutectic freeze crystallization [90], and suspension freeze concentration [93]. Lind et al. froze the urine at a temperature of $-14\text{ }^{\circ}\text{C}$, concentrating approximately 80 % of nutrients through freeze-thaw methods within 25 % of the original volume [65]. Noe-Hays et al. assessed the performance of a scalable block freeze device for concentrating source-separated urine. Specifically, the relative mass of N, P, and K remained constant during the processing, and the evaluation indicated that the two-stage system recovered 92 % of the nutrients, achieving a concentration factor of 5.8 [91].

Singular freeze concentration technique could combine with other urine recovery technologies to achieve more efficient and energy-saving reclamation of nutrients. Courtney and Randall studied a novel hybrid system that combines eutectic freeze crystallization and RO technologies to concentrate human urine [90]. Zsófia and Nilsson combined freeze-thaw, struvite precipitation, and adsorption with zeolite or activated carbon technologies to recover N and P resources from human urine [94]. The freeze-thaw method concentrated 60 % of the nutrients within 40 % of the initial volume, with a P recovery rate of 95–100 %. Ganrot et al. utilized pretreated zeolite and struvite precipitation techniques in conjunction with freeze-thaw cycles to recover nutrient from human urine [95]. They conducted plant availability tests on common wheat, achieving a high phosphorus recovery rate (>97 %) and a nitrogen recovery rate ranging from 50 % to 60 %. The treated solution exhibited significantly lower toxicity than the original urine, demonstrating the potential of these minerals to serve as controlled-release fertilizers in a spring wheat greenhouse.

4. Processes recovering multi-nutrients from urine without urine concentration

4.1. Simultaneous recovery of N and P from urine by struvite precipitation

Struvite precipitation has attracted interest as a route for P recovery and has been widely used for resource recovery from urine [96]. Compared to other crystalline forms such as hydroxyapatite ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$) and calcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$), struvite exhibits obvious advantages [97]. Recycled struvite has two to three orders of magnitude lower impurities than commercial phosphate fertilizers and releases nutrients at a slower rate. In addition to this, struvite can bind

essential nutrients (P, N (ammonia), and Mg (Mg^{2+})) on the same crystal [98]. In practice, struvite crystals can be recovered due to the relatively high recovery value of the product. The reaction of struvite formation can be expressed as Eq. (2):



In the reaction equation, ΔG represents the Gibbs free energy, and the Gibbs free energy of each substance is shown in Table S1.

Urea hydrolysis increased the pH from 5.9 to 8.6, resulting in a decrease in PO_4^{3-} concentration due to the reaction with NH_4^+ and Mg^{2+} ions. Liu et al. reported that the pH increased and stabilized at about 9.0 during urea hydrolysis. As a result of the elevated pH, it was observed that after 6 days of urine storage, about 31 % of the P was recovered by spontaneous quantity precipitation [33]. Similar results were reported by Etter et al., where about 30 % of the P was removed by spontaneous precipitation as calcium phosphate and granite in the undiluted urine [99]. Liu et al. demonstrated that after 5 days of urine storage, almost 38 % of the P was recovered [100]. Struvite recovered from urine is a sanitary and safe fertilizer. Compared with other recycled products or commercial minerals and organic fertilizers, it contributes less to heavy metals in agricultural land. When used in combination with other soil amendments, struvite is an effective fertilizer that can meet more than half of the Mg and P required by crops [101].

Mg concentration in urine is generally considered to be the limiting element for struvite crystallization, as its concentration is much lower than that of N and P [102]. Therefore, to better recover struvite from urine, chemicals need to be added, including alkali sources and Mg sources to adjust the pH and to achieve the best conditions for struvite recovery [103]. Due to the lack of sufficient Mg in the urine, Mg supplementation is essential. The most common sources of Mg are magnesium oxide (MgO), magnesium chloride (MgCl_2), and magnesium hydroxide ($\text{Mg}(\text{OH})_2$) [104]. Due to their basic character, MgO and $\text{Mg}(\text{OH})_2$ can serve the dual purpose of adding Mg^{2+} and increasing pH simultaneously [105].

However, the Mg sources tend to be more expensive [106]. Some researchers have attempted to recover P through less commonly used sources of Mg [107]. As a result, many cheap alternative Mg sources have emerged. Etter used Mg oxide produced by magnesite and waste bittern from salt making [99]. Pinatha et al. used a mixture of solid waste (SW) incineration ash and sea salt as Mg sources and compared the efficiency of P recovery from urine with MgCl_2 [108]. Another research by Rubio-Rincón et al. tested flushing toilets with seawater to precipitate struvite when mixed with urine. The experiments showed that up to 99.0 % of P was recovered while maintaining a seawater-to-urine ratio of <3.3:1 at a pH of 9.0 [109]. Guadie et al. tested the efficiency of Lake Afrera in the Rift Valley as a potential source of Mg for the recovery of P in the form of struvite from real and synthetic urine, with P recoveries of up to 98.0 % [110].

The main limiting factors for recovering struvite from urine are Mg sources and pH. Finding cheaper Mg sources and ways to adjust pH are still the challenges of recovering struvite in the future. These cheap Mg sources help reduce the cost of struvite recovery from urine.

4.2. The electrochemical coupled struvite precipitation for simultaneous recovery of N and P from urine

Electrochemically driven precipitation of struvite has attracted considerable interest. Perera and Englehardt used electrochemical pH adjustment, traditional precipitation, and ammonia stripping to recover 89 % of the TN and 97 % of the total phosphorus from the precipitated waste in a continuously flowing reactor [111] (Fig. 3a). Similarly, Gao et al. used membrane capacitor deionization after bipolar membrane electrodialysis and struvite precipitation to capture and recover 72 % of the remaining ammonia nitrogen as a concentrated stream [112] (Fig. 3b).

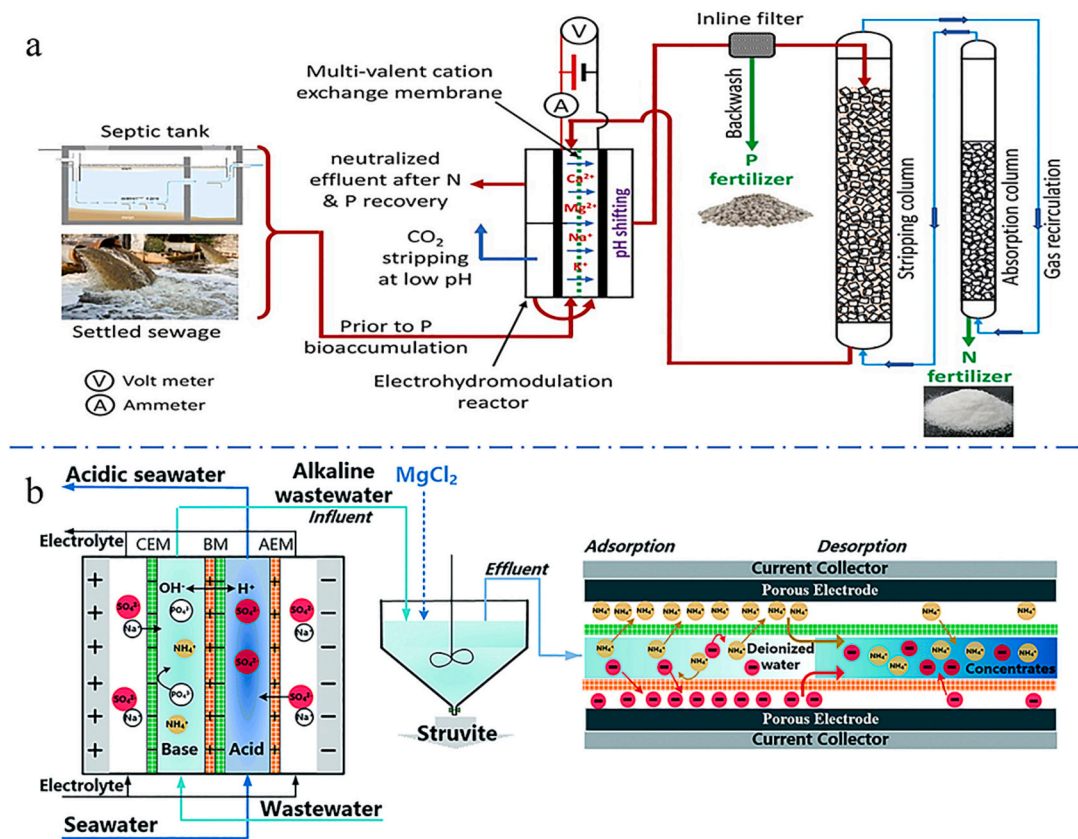


Fig. 3. Mechanistic diagrams of two typical ammonium vapor stripping-coupled electrochemical techniques: (a) combined electrohydroconditioning reactor, conventional precipitation, and ammonia vapor stripping are used to recover both nitrogen and phosphorus from precipitated effluent in a continuous flow mode [111] (reproduction with permission, Copyright 2022 Elsevier). (b) Schematic diagram of the bipolar membrane electrodialysis-struvite reactor-membrane capacitance deionization method is utilized for the complete recovery of both nitrogen and phosphorus [112]. (Reproduction with permission, Copyright 2020 Royal Society of Chemistry).

The use of Mg sacrificial anodes in the electrochemical precipitation of struvite is a new potential source of Mg. It reduces the amount of chemical addition required for precipitation or pH adjustment of struvite. Mg has the lowest standard reduction potential compared to other engineering metals [113]. At 25 °C, the standard potential of Mg is $-2.37 V_{\text{NHE}}$ [114] and the typical corrosion potential (E_{corr}) in aqueous solution is $-1.7 V_{\text{NHE}}$ [115]. Therefore, Mg becomes an excellent candidate for sacrificial anode materials [116]. Mg alloys AZ31 and AZ91 are commonly used by some researchers in electrochemical experiments for struvite recovery [117]. Pure Mg has a Mg content of 99.8 %, while other alloys (AZ31 and AZ91) have a Mg content of 90–96 %. The addition of alloying components can increase the conversion of Mg [118]. Kékedy-Nagy et al. reported that pure Mg and Mg alloy (AZ31) were used as anodes, and struvite crystals with slender needle-like shapes were obtained at the anodes. In a 6 h batch experiment, the pure Mg anode produced 4.5 times higher mass of struvite than AZ31 alloy and 2.8 times higher steady-state current density than AZ31 alloy [119].

Kruk and Oleszkiewicz reported the use of Mg alloy (AZ91HP) to recover struvite, with a high P recovery of $4.0 \text{ mg PO}_4\text{-P cm}^{-2} \text{ h}^{-1}$ at a current density of 45 A m^{-2} [120]. Hug et al. used a Mg alloy as the anode and a steel plate as the cathode and achieved an average phosphate removal rate of $3.7 \text{ mg P cm}^{-2} \text{ h}^{-1}$ at an anode potential above -0.9 V . The cost of recovering struvite is lower than adding MgCl_2 and MgSO_4 directly [102]. Liao et al. reported that single-chamber Mg air fuel cells simultaneously recover struvite and electrical energy. Mg^{2+} produced by anodizing Mg alloys provides a source of Mg for struvite precipitation. Meanwhile, OH^- is generated by a four-layer air cathode reduction reaction to maintain a proper pH value for struvite

precipitation. After 25 min of reaction, the P in the urine was removed by 99.48 %, and a precipitate of 98.26 % purity equivalent to that of commercial struvite was obtained. It was also found that the presence of struvite precipitation can enhance the electrical properties of MAFC. The discharge voltage of MAFC with urine as the electrolyte is greater than the discharge voltage of MAFC with P-free urine as the electrolyte [121].

In summary, the electrochemically induced precipitation of struvite was successful in both real and synthetic urine, but most studies have been conducted using synthetic urine in laboratory environments. Although this process has shown potential for larger-scale application, using synthetic urine is insufficient to demonstrate its effectiveness in actual applications for recovering struvite. Therefore, in addition to adjustments in operational factors such as anode selection, pH value, flow rate, current density, and external current, the use of real urine and scaled-up experiments are crucial for the widespread implementation of the process density, and applied current are crucial to operate an efficient and productive process.

4.3. The air stripping coupled struvite precipitation for simultaneous recovery of N and P from urine

Nitrogen stripping is primarily achieved by adding a base such as sodium hydroxide to adjust the pH of urine to 9.0–13.0. The urine is then heated to 40–60 °C and fed into the stripper [122]. As the bubbles rise, ammonia is transferred from the liquid phase to the gas phase. The ammonia-rich gas enters the absorption tower and comes into contact with the sulfuric acid solution. H_2SO_4 is the most common acid used to absorb ammonia and one of its end products is ammonium sulfate [123]. Hydrolyzed urine has a high alkalinity ($\sim 15 \text{ g CaCO}_3/\text{L}$) and is better

suited for stripping and recovery [47]. Ammonia stripping can be used in conjunction with struvite precipitation to facilitate the simultaneous recovery of N and P from urine [122]. Although the struvite precipitation method is effective in recovering equal molar ratios of $\text{NH}_4^+\text{-N}$ and $\text{PO}_4^{3-}\text{-P}$, the concentration of NH_4^+ in the hydrolyzed urine is much higher than that of PO_4^{3-} [124], and this discrepancy results in a lower recovery efficiency of N than P. The results of numerous experiments have shown that ammonia stripping can be effective in the recovery of N from urine. For instance, TAO et al. used a pilot-scale stripping device that recovers 72–77 % ammonia from urine and produces 37.6–39.7 g/L ammonium sulfate [125].

To enhance $\text{NH}_4^+\text{-N}$ recovery efficiency, some researchers have combined ammonia stripping and struvite precipitation to recover the ammonium nitrogen fraction that cannot be recovered by struvite precipitation alone. Wei et al. accomplished the retrieval of struvite and ammonium sulfate from urine by combining struvite precipitation and air stripping in a pilot-scale treatment [40]. The system achieved 94 % struvite precipitation efficiency. The results indicate that combining struvite precipitation with air stripping can achieve continuous recovery of N and P. Although the order of stripping and precipitation does not significantly affect the recovery efficiency of N and P, a high concentration of $\text{NH}_4^+\text{-N}$ is beneficial for the formation of struvite. Therefore, the sequence of ‘struvite precipitation → air stripping’ is optimal. However, when simultaneously recovering N, P, and K elements, $\text{NH}_4^+\text{-N}$ must be recovered first and then K, as the presence of $\text{NH}_4^+\text{-N}$ interferes with the precipitation of potassium in hard rock. In addition to the aforementioned process combination, the air stripping method can be combined with electrochemistry and other technologies to simultaneously recover nutrients from urine.

4.4. Bioelectrochemical systems for simultaneous recovery of N and P from urine

Bioelectrochemical systems (BESs) that utilize urine as feedwater have emerged as a potential alternative source for extracting valuable nutrients and as a cutting-edge technology for wastewater treatment [126]. BESs consist of two electrodes: an anode and a cathode, usually separated by an ion-selective membrane. At the anode, bacteria catalyze the oxidation of organic matter into electrons, carbon dioxide, and protons [127]. In the absence of an electron acceptor (oxygen) at the anode, electrons are transferred from bacteria to the electrodes and transported through a circuit to the cathode, where they undergo a reduction reaction.

Microbial fuel cell (MFC) and microbial electrolysis cell (MEC) are the two most widely used bioelectrochemical system [128]. MFCs utilize microorganisms that draw electrons from organic compounds to generate energy. MFCs have advantages over other types of bioenergy conversions as they produce considerably less sludge and can operate in ambient conditions, thereby reducing costs [129]. MFCs are a potential solution for energy-poor regions because they can produce energy and treat different kinds of feedstocks simultaneously. Urine is not only an efficient feedstock for MFCs but also provides an opportunity for N, P, and K recovery [130].

The principle of an MEC is opposite to that of an MFC. On the MFC cathode, oxygen is reduced to water, and hydroxyl ions are generated. However, at the cathode of an MEC, water is reduced to hydrogen (H_2) and hydroxide ions [131]. Therefore, MECs can generate H_2 while recovering nutrients from urine, reducing system costs, and enhancing overall system resource recovery efficiency. Different BESs can be selected to treat urine for various application scenarios and purposes. Table 2 presents a summary of the main processes and data on the

Table 2
Bioelectrochemical systems for nitrogen and phosphorus recovery from human urine.

Type of BES used	Wastewater	Operational condition	Anode	Cathode	Nitrogen recovery efficiency	Phosphorus recovery efficiency	Reference
Single-chamber MFC	Hydrolyzed urine	Temp: 25 °C	Carbon felt	Pt-coated Carbon paper	$\text{NH}_4^+\text{-N}$ recovery: 28.6 %	PO_4^{3-} recovery: 94.6 %	[129]
Single-chamber air-cathode MFC	Five times diluted urine	Temp: 25 °C	Carbon felt	Pt-coated carbon paper	$\text{NH}_4^+\text{-N}$ removal: 40 % $\text{NH}_4^+\text{-N}$ recovery: 27.9 %	PO_4^{3-} recovery: 42.6 %–62.4 %	[129]
Stacked MFC	Neat human urine	pH: 6.4	Carbon fiber veil	AC-carbon fiber veil	–	PO_4^{3-} recovery: 82 %	[133]
Air-cathode MFC	Fresh urine	Temp: 30 °C	Graphite fiber	Carbon cloth	TN removal: 84.9 % $\text{NH}_4^+\text{-N}$ recovery: 52.8 %	–	[134]
Three-chamber resource recovery MFC	Synthetic urine-containing wastewater with various organic pollutants	Anode chamber pH: 6.00–7.90 Middle chamber pH: 7.20 Cathode chamber pH: 6.00–7.90	Graphite fiber brush	Circular activated carbon air-cathode	Urea removal: 99 % TN recovery: 84.9 %	PO_4^{3-} removal: 99 % (in the anode chamber and cathode chamber) PO_4^{3-} recovery: 82 % (middle chamber)	[135]
Air-exposed biocathode MFC	Synthetic urine	Temp: 20–25 °C	Carbon felts (fixed vertically with an angle of 60°)	Carbon felts (fixed vertically with an angle of 60°)	$\text{NH}_4^+\text{-N}$ recovery: 62.7 % ± 2.3 % TN recovery: 86.8 % ± 1.5 % $\text{NH}_4^+\text{-N}$ recovery: 46 ± 2.16 %	–	[136]
Field-scale MFC	Fresh urine	Hydraulic retention time: 2.5–4.5 L/day	Stainless-Steel mesh	Stainless-Steel mesh	–	PO_4^{3-} recovery: 90 ± 1.5 %	[137]
Terracotta clay membrane material MFC	Fresh human urine (mixed with artificial seawater)	pH:9–9.28	Carbon veil	Activated carbon	Struvite recovery: 94 %	Struvite recovery: 94 %	[138]
Scaled-up MEC	Complete hydrolysis urea	Conductivity: 27.4 ± 1.9 mS cm^{-1} , pH:9.1 ± 0.2	Titanium plates with a metal oxide coating	Titanium plates with a mixed metal oxide coating	TAN removal: 31 ± 13 % TAN recovery: 31 ± 59 %	Struvite recovery: 96 %	[139]
MEC	Five times diluted urine	Applied voltage: 1.0 V, pH:9.1	Graphite felt	Platinized flow field.	Ammonium removal rate: 173.4 ± 18.1 g $\text{N m}^{-2} \text{d}^{-1}$	–	[127]

recovery of N and P from urine using BES by different researchers. The main types of MFCs used for urine processing include single-chamber MFC (SCMFC), dual-chamber MFC (DCMFC), tubular stack MFC, origami star-inspired, and up-flow mode MFC [132].

4.5. BES coupled struvite precipitation to recover N and P from urine

The microbial electrochemical method for struvite recovery is an emerging approach in the field of struvite reclamation. In the MFC process, the oxygen obtained at the cathode is reduced and combined with protons to form water, which causes the pH of the cathode to rise [140]. In the MEC process, electrons are combined with free protons in the solution to generate hydrogen, which consumes the protons at the cathode, causing the cathode pH to increase [141]. Based on this assumption, MFC and MEC cathodes can be used to recover struvite [142,143].

Studies conducted by many researchers have shown that BES combined with struvite precipitation can successfully recover P from urine. Additionally, MFC electricity production can be utilized to reduce costs and improve resource utilization during P recovery. Zang et al. reported that 62.4 % P was successfully recovered from urine as MAP (struvite) precipitation using MFC, with a power density output of 0.9 W m^{-3} [129]. You et al. reported a three-stage MFC/struvite extraction process system that recovered 82 % of PO_4^{3-} -P in urine. During the operation, 14.32 W m^{-3} (absolute power: $358 \mu\text{W}$) and 11.76 W m^{-3} (absolute power: $294 \mu\text{W}$) were generated from the first and third stages of the system, respectively [144]. Tao et al. studied the performance of a single-chamber MFC and dual-chamber MFC for power generation and struvite recovery, and the results proved that the single-chamber MFC is more suitable for struvite recovery, while the dual-chamber MFC has a stronger power generation capacity [145]. Furthermore, the combination of MEC and struvite precipitation allows for the recovery of both P and H_2 . Almatouq et al. studied the rate of simultaneous generation of H_2 and recovery of P in a two-chamber MEC. The maximum P precipitation efficiency achieved by MEC was 95 % and the maximum H_2 generation rate was $0.28 \text{ m}^3\text{-H}_2/\text{m}^3\text{-d}$ [146].

The mobility in a cation exchange membrane (CEM) can be increased by using an external power supply, which enhances hydrogen production and other reactions in the cathode. Higher current density leads to better ammonia recovery, making externally powered MECs more effective than MFCs in ion recovery [147]. In MFCs and MECs with CEMs, the high pH near the cathode surface, due to proton depletion or hydroxyl release, may improve nutrient recovery by facilitating the conversion of ammonium to ammonia or struvite crystals [148].

As a novel recovery technology, BES enhances ammonia evaporation by increasing pH in the cathode area, offering an eco-friendlier method for nutrient recovery from urine without high temperatures or chemicals. However, the cost of components like electrodes and membranes still restricts its development. More importantly, many new types of BES combination systems use synthetic urine for research, which does not clarify whether the microbes in real urine compete with the system, or whether other components in real urine impact the recovery efficiency. Despite its potential, BES needs to be tested on a larger scale with real urine and could potentially integrate with or replace certain steps in traditional recovery processes for more efficient, environmentally friendly nutrient recovery.

5. Simultaneous recovery of P and K from urine by K-struvite precipitation

So far most nutrient recovering technologies focus on N and P elements, overlooking the abundant presence of K in urine. Therefore, it is necessary to extend the application of these technologies to the recovery of K in urine. The most widely applied technology for recovering specific K elements in urine is the K-struvite (magnesium potassium phosphate hexahydrate, MPP, $\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$), the allotrope of MAP [149], and an

effective slow-release fertilizer, similar to MAP [50]. The formation process of MPP and MAP also reveals their homologous relationship [150]. However, the presence of Na^+ , NH_4^+ , and Ca^{2+} ions in urine, especially NH_4^+ , significantly inhibits the formation of MPP [151]. The ion binding mechanism of MAP and MPP is illustrated in Fig. 4. Initially, the ions of K, P, N, and Mg elements exist freely in urine. Subsequently, Mg ions and H_2O molecules combined to form a dimer as $\text{Mg}(\text{H}_2\text{O})_6^{2+}$ with an octahedral structure. Hydrogen bonds linked the octahedra of $\text{Mg}(\text{H}_2\text{O})_6^{2+}$ and the tetrahedron of PO_4^{3-} groups together. Following that, there was a continuous polymerization of K^+ ion coordination with the four oxygen atoms of H_2O molecules and one oxygen atom of PO_4^{3-} . The interaction between single nuclei formed a new crystal seed nucleus [152]. In the chemical crystal structure, the ionic radius of NH_4^+ is greater than that of $\text{K}(\text{H}_2\text{O})$. The stability of MPP containing $\text{K}(\text{H}_2\text{O})^+$ with a smaller ion radius is lower than that of MAP containing NH_4^+ with a larger ion radius. In normal urine with the coexistence of NH_4^+ and K^+ , the precipitation tendency of MAP is higher than that of MPP. Therefore, the process of recovering K from urine needs to remove ammonia from urine, and it is required to be used in combination with other processes, such as ammonia stripping [153] and MAP precipitation [154].

Even in the absence of ammonia, co-precipitation of sodium magnesium heptahydrate (MSP) and MPP can occur in urine [155], since K content is higher than Mg and P in normal urine [156]. To efficiently recover K from urine, additional sources of P and Mg need to be added. However, the cost of recovering K-struvite is high, which is not conducive to the recovery and application of K-struvite [154]. As a result, more researchers are focusing on the cost-effective recovery of K-struvite, e.g., using cheaper Mg sources, etc. Huang compared the effects of different methods using lower magnesium oxide (LG-MgO) on the crystals of K-struvite. The results show that LG-MgO plus stabilizer and phosphoric acid (H_3PO_4) are more suitable for K and P [20]. Huang also compared the effects of different Mg sources including MgCl_2 , Mg sacrificial electrode, and $\text{Mg}(\text{OH})_2$ on the crystallization of MPP. Analysis showed that MgCl_2 is the best Mg source for K-struvite crystals and can obtain the highest K recovery [157].

Zhang et al. conducted a pilot-scale fluidized bed reactor (FBR) to recover K and P from source-separated urine using MPP crystallization [158]. Ultimately, high-purity microspheres ($86 \pm 2 \%$) with a maximum particle size of 4 mm were generated by operating at a surface flow rate of 350 cm/min. Removal rates of 20–35 % for K and 80–90 % for P were achieved. Similarly, Van Giang Le et al. used a fluidized bed homogeneous crystallization process, which resulted in total removal rates (TR%) of 98.4 % and 70.5 % for P and K, and crystallization rates (CR%) of 86.5 % and 62.3 %, respectively [152].

Other researchers have investigated the crucial influencing factors to increase the recovery of K-struvite. Xu et al. reported that the maximum recovery of P and K reached 99 % and 33 %, respectively, when a Mg source was added. It was found that the pH value and the molar ratio of Mg: P were the key factors determining the efficiency of nutrient recovery [19]. Gao et al. used a continuous flow MPP precipitation process to treat synthetic and real urine, effectively removing P and K from both types of urine. It was proved that adding the pre-formed MPP as a seed crystal to the solution helps to improve the K removal efficiency [151].

The concentrations of phosphates and magnesium, along with NH_4^+ , Ca^{2+} , and Na^+ , greatly impact MPP crystallization. Crystal size also affects its use as an agricultural fertilizer, necessitating further research into MPP crystallinity. In MPP precipitation research, most researchers use synthetic urine due to the high NH_4^+ -N concentration in real urine, which impacts K^+ recovery. Using real urine requires coupling with an N recovery process, which is often overlooked in research. However, real urine is essential for effective nutrient recovery. We suggest that future researches on MPP precipitation should include coupled N recovery to better simulate the conditions and processes of real urine recovery, thereby enhancing the scalability and commercial viability of this method.

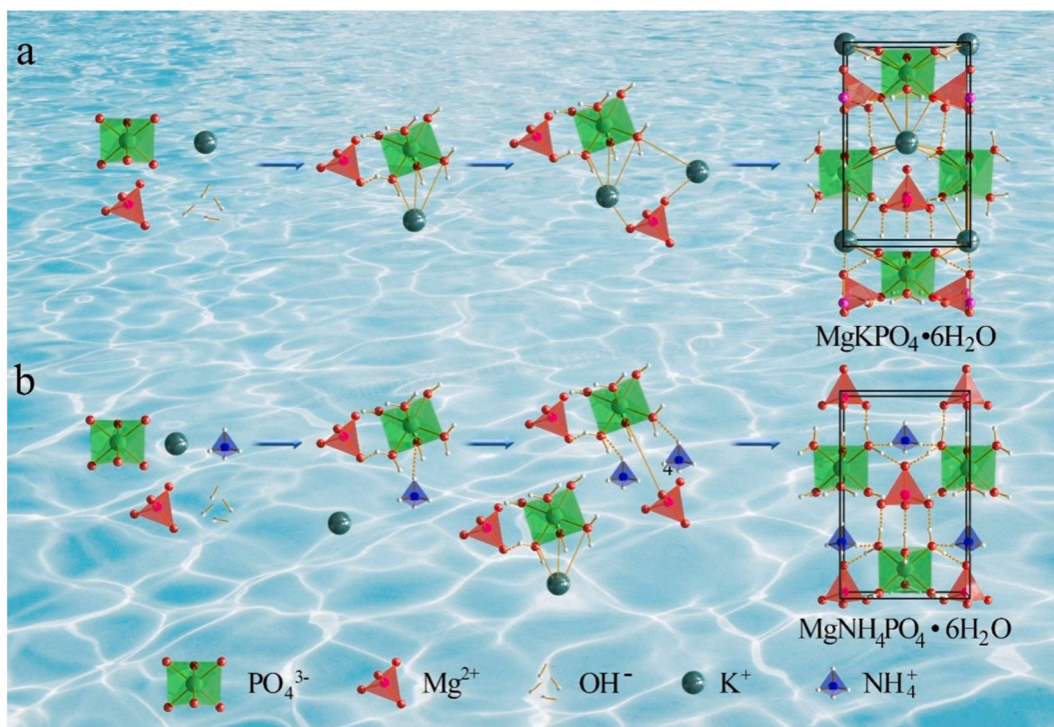


Fig. 4. Schematic diagram illustrating the competitive mechanism between K^+ and NH_4^+ in urine and the precipitation process of MAP ($MgNH_4PO_4 \cdot 6H_2O$) /MPP ($MgKPO_4 \cdot 6H_2O$). (a) MPP sedimentation mechanism diagram. (b) MAP sedimentation mechanism diagram. Diagram of MAP precipitation mechanism and K^+ and NH_4^+ competition mechanism.

6. Optimal recovery sequence of simultaneously recovering N, P, and K from urine through combined processes

6.1. The optimal process combination and statistical analysis

Urine contains valuable nutrients including N, P, and K. However, recovering these nutrients in one process is always challenging, requiring a combination of processes to maximize the recovery of resources. Consideration should be given to the order of N, P, and K recovery procedures and the selection of various processes to maximize recovery (Fig. 5). Researchers can choose a single recovery method if they want to obtain a blended fertilizer, but a combined recovery process is necessary for multi-N/P/K product. Table S2 lists the combined processes suitable for simultaneous N/P/K recovery. In terms of simultaneous recovery of N and P from urine, in addition to MAP precipitation, Pradhan et al. used ferrous precipitation and denitrification to recover nitrogen and P separately in the same process so that they can be used independently [159]. Over 98 % of the N and P can be recovered from the urine in 8 h at 30 °C.

To quantitatively express the constraints of current recycling technology, we conducted a statistical analysis of results from prior studies published in the literature (see Table S3). This analysis helps to more clearly identify and demonstrate trends and directions. The results of this statistical analysis are shown in Fig. 6. As can be intuitively seen from the figure, although struvite precipitation technology can recover most of the P in urine, it fails to recover N and K. Therefore, it is necessary to couple N and K recovery processes to synchronously recover N,P, and K elements. The air stripping method can efficiently recover N from urine without affecting the recovery of P and K. Distillation can recover nearly 100 % of the K salts in urine and also reclaim the water. Therefore, we consider “struvite precipitation → air stripping → distillation” and “air stripping → K-struvite precipitation” to be the two most optimal recovery process combinations.

Currently, some researchers have utilized these two process combinations for the simultaneous recovery of N, P, and K from urine. Xu et al.

utilized a laboratory-scale air stripping and K-struvite precipitation combined process to recover 78 % of N, 72 % of P, and 73 % of K from source-separated urine [43]. A study by Jagtap and Boyer demonstrated effective recovery of over 80 % of N, P, and K from urine using a combination of struvite precipitation, ammonia vaporization-acid uptake, and evaporation [160]. Jagtap et al. also explored distillation for K and non-potable water retrieval, achieving about 100 % K recovery and over 50 % urea water recovery [41]. This process resulted in higher K concentrations in potash than ion exchange methods. In an on-site study, Jagtap and Boyer collected urine and recovered nutrients and non-potable water through a multi-process approach [161]. This process exceeded 90 % in the quality of N, P, and K, and reclaimed water, producing value-added products like struvite, ammonium sulfate, and potash, surpassing commercial equivalents.

The stored urine displayed considerable variability in characteristics across samples. However, the N, P, and K levels in urine exhibited a trend of $P < K < N$, with N being the most abundant nutrient presence. Therefore, the optimal treatment sequence, as described in this review, involved recovering P first, followed by N and finally K, resulting in high nutrient recovery and minimal cost. Wu et al. integrated Na-chabazite adsorption with ammonia stripping and K-struvite precipitation [162]. Their experiments showed that both “stripping → K adsorption + struvite precipitation” and “stripping → K adsorption → struvite precipitation” were highly efficient. The first method yielded nutrient-rich charbarite, while the second produced ammonium sulfate, struvite, and K salts. Jang coupled ammonium distillation and struvite process studied the effect of different distillation gas-to-liquid ratios on the composition of struvite crystallization and the overall nutrient recovery rate from struvite [163].

6.2. Mass balance and cost analysis

To better reflect the changes in N, P, and K elements during the composite technology recovery process, we have established a comprehensive material balance model for two optimal recovery

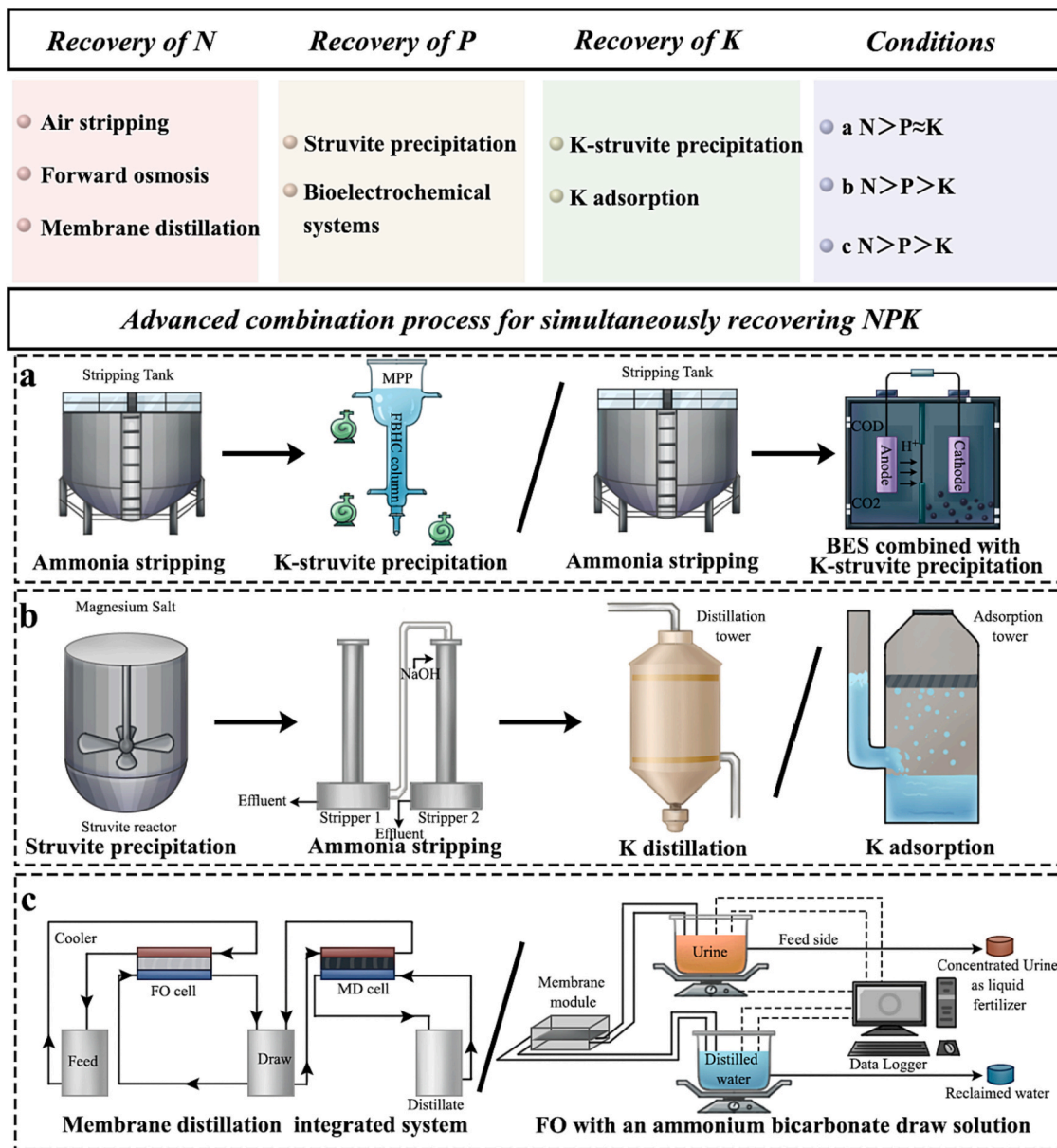


Fig. 5. Flow diagram of combined process and application scenarios for simultaneous recovery of N, P, and K from urine. (a) Two-step combined process for the recovery of ammonia and K-struvite; (b) three-step combined process for the recovery of ammonia, guano, and potassium salt; (c) single system for the recovery of concentrated fertilizer.

technology combinations based on the statistical data in Table S3. We used a machine learning MATLAB fully connected neural network to create an N, P, and K process recovery prediction regression model. The input layer has 1 unit, the hidden layer has 128 units, and the output layer has 1 unit. The maximum number of iterations is set to 1000. We built and trained the neural network using sklearn and numpy, with Python as the development language and PyCharm as the development software.

The neural network diagram is as shown in Fig. S2, and the material balance diagram is shown in Fig. 7. The recommended best recovery for urine in this study is storing urine and stabilizing urine. Urea has been hydrolyzed and ammonia nitrogen is fixed in the urine, so when calculating the material balance, we do not consider any additional ammonia losses except for certain research reaction processes. Since PO_4^{3-} changes very little or almost not at all during the gasification process, phosphorus loss can be neglected. Assuming that the ammonia in the gasification process can be completely absorbed by H_2SO_4 , most of the N, P,

and K elements can be recovered through the process combination of “struvite precipitation → air stripping → distillation,” (Fig. 7a). However, the “air stripping → K-struvite precipitation” process combination can only recover approximately 65 % of K (Fig. 7b), and this is also related to the incoming K concentration.

In order to evaluate the economic feasibility of selective nutrient recovery for N, P, and K, a preliminary economic analysis was conducted. Utilizing data simulated by the model, the theoretical economic costs and income for the recovery of N, P, and K from 1 m^3 of real urine were calculated (Table 3). The income from nutrient recovery from urine using the two process combinations is calculated based on the consumption of chemicals and energy, as well as the income from the final products. The final income for process “struvite precipitation → air stripping → distillation” is \$0.66 per cubic meter of urine, and for process “air stripping → K-struvite precipitation”, it is \$2.10 per cubic meter of urine. Due to the fewer process steps, the K-struvite precipitation combination has lower energy consumption. However, it has higher

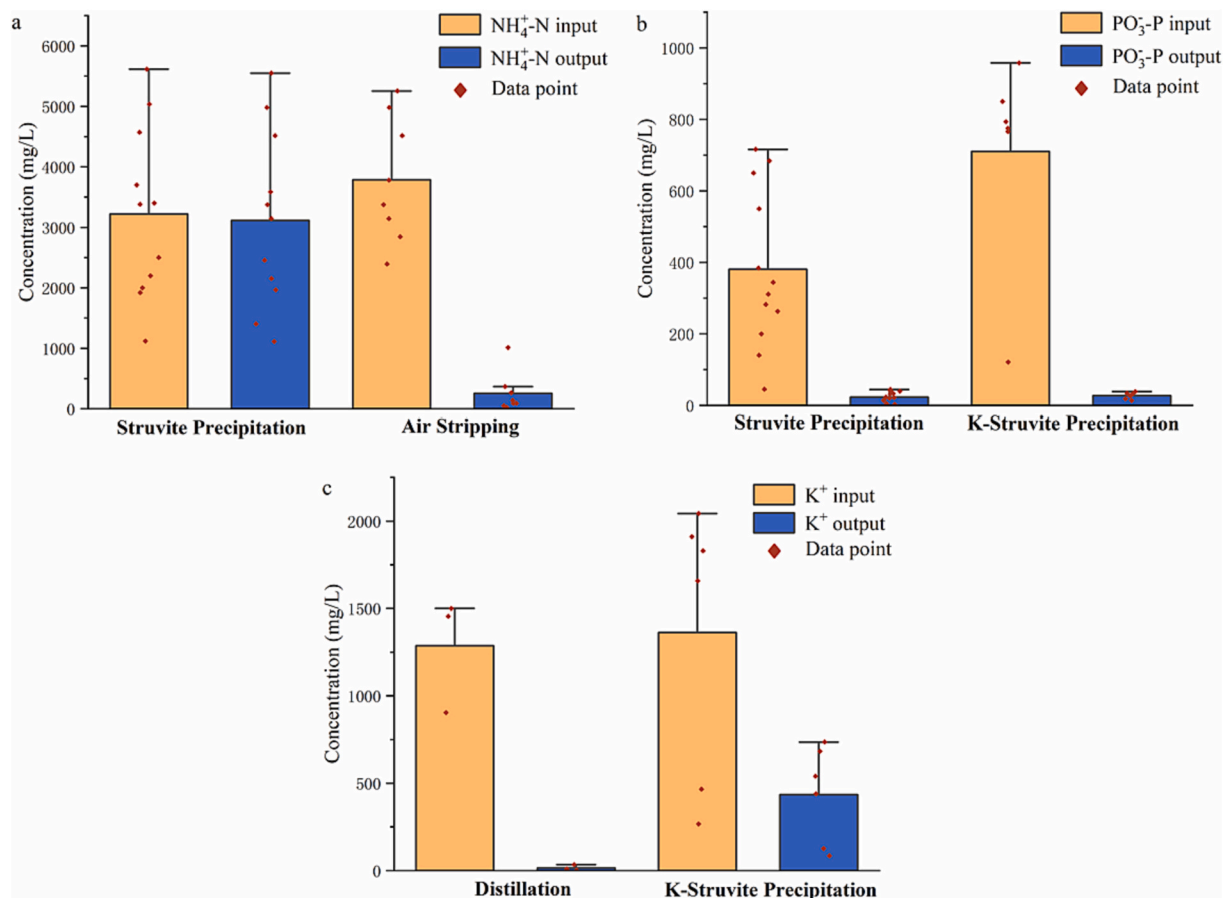


Fig. 6. Statistical diagram of N, P, and K concentration in the influent and effluent of different recycling processes. (a) shows the change in $\text{NH}_4^+\text{-N}$ concentration in the influent and effluent of struvite precipitation and air stripping; (b) shows the change in $\text{PO}_3^{3-}\text{-P}$ concentration in the influent and effluent of struvite precipitation and K-struvite precipitation; (c) shows the change in K^+ concentration in the influent and effluent of distillation and K-struvite precipitation.

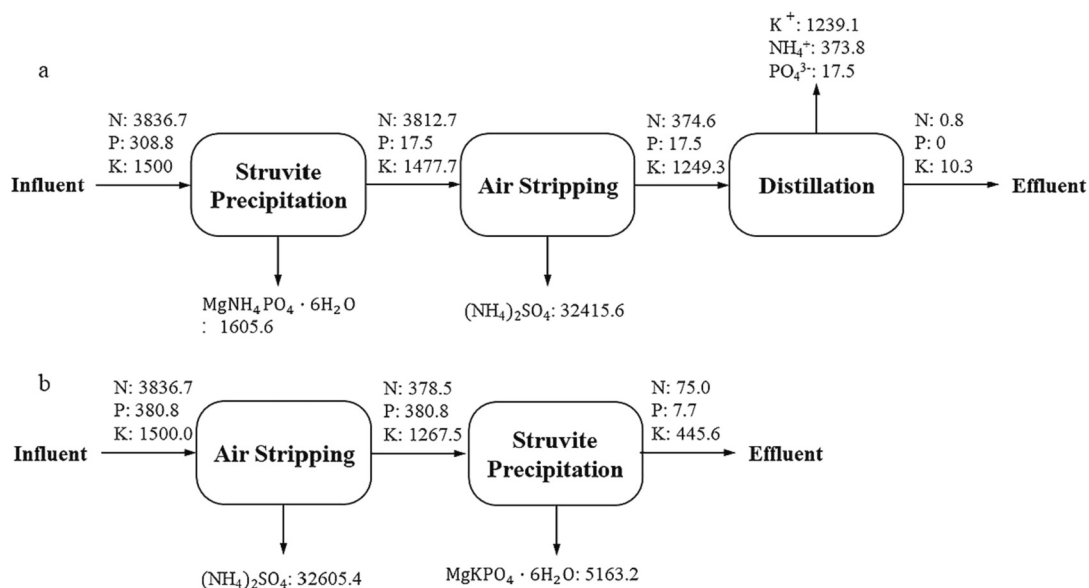


Fig. 7. Mass balance analysis diagram for N, P, and K content in combined process influent and effluent. (a) The mass balance of combination “struvite precipitation → air stripping → distillation”, (b) the mass balance of combination “air stripping → K-struvite precipitation”. Concentrations of the elements in the table are in mg L^{-1} .

Table 3Theoretical economic evaluation of N, P, and K recovery from 1 m³ of human urine through the combined process.

Parameter	Amount	Price	Data source	Parameter	Amount	Price	Data source
Struvite precipitation → Air stripping → Distillation				Air stripping → K-struvite precipitation			
MgCl ₂ ·6H ₂ O consumption	1.45 kg	\$ 0.25/kg	[164]	MgCl ₂ ·6H ₂ O consumption	3.65 kg	\$ 0.25/kg	[164]
H ₂ SO ₄ consumption	6 L	\$ 0.13/L	[164]	H ₂ SO ₄ consumption	6 L	\$ 0.13/L	[164]
Energy for drying the sediment	0.1kWh	\$ 0.07/kWh	[165]	Na ₂ HPO ₄ consumption	3.716 kg	\$ 0.31/kg	Alibaba.com
Energy for heating	239kWh	\$ 0.07/kWh	[164]	Energy for heating	239kWh	\$ 0.07/kWh	[164]
Energy for vacuum	3.7kWh	\$ 0.07/kWh	[164]	Energy for vacuum	3.7kWh	\$ 0.07/kWh	[164]
Energy for distillation	21kWh	\$ 0.07/kWh	[83]	Energy for water pumping	0.035kWh	\$ 0.07/kWh	[152]
(NH ₄) ₂ SO ₄ production	32.42 kg	\$ 0.50/kg	[164]	Energy for drying the sediment	0.035kWh	\$ 0.07/kWh	[152]
Struvite crystals Production	2.87 kg	\$ 0.50/kg	[164]	(NH ₄) ₂ SO ₄ production	32.60 kg	\$ 0.50/kg	[164]
Muriate of Potash production	3.97 kg	\$ 0.66/kg	[161]	K-struvite crystals production	5.63 kg	\$ 1/kg	[152]

The consumption of chemicals and product quantities in the table are calculated based on the material balance model for influent and effluent water. The prices of chemicals and products in the table are sourced from literature data.

chemical input costs, and the final crystallinity of K-struvite affects whether the end product has recovery value. The economic analysis shows that both combinations of technologies have positive income, but it has not accounted for labor and maintenance costs. Additionally, the economic feasibility of the processes can also be influenced by different on-site conditions. Using more cost-effective sources for chemicals or adopting green energy sources can help reduce costs and improve the economic viability of the process combinations.

7. Conclusion

We reviewed and evaluated methods to recover N, P, and K from urine. In pursuit of efficient resource recovery, we recommend the utilization of hydrolyzed urine due to the altered urine properties resulting from urea hydrolysis. The primary obstacles in urine recycling revolve around the modification of recovery conditions (e.g., adjusting pH, determining the appropriate molar ratio of N: P: K: Mg, etc.) and enhancing recovery efficiency. To maximize the efficacy of resource reclamation, it is imperative to identify the most suitable combination selection while the sequence of the recovery procedures is crucial. For instance, the recovery of K following the retrieval of ammonia, Mg, and P has the potential to yield higher returns at reduced costs. In the future, the focus should pivot towards the promotion and application of nutrient recovery technology in urine management. This entails not only the development and utilization of recovered products but also the conduction of pilot-scale tests. Furthermore, exploration into the feasibility of large-scale industrialization of urine recovery should be a central consideration, marking a critical stride towards sustainable and efficient resource management.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jwpe.2024.104975>.

CRedit authorship contribution statement

Qiting Liu: Writing – original draft. **Wenhui Sun:** Methodology. **Qianhao Zeng:** Investigation. **Haodong Zhang:** investigation. **Chengyang Wu:** Writing – review & editing. **Eric Lichtfouse:** Writing – review & editing. **Hongbo Liu:** Funding acquisition; conceptualization; supervision; writing-review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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