

Review

Physical Separation: Reuse Pollutants and Thermal Energy from Water

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Abstract: Conventional sewage treatment based on biological and chemical methods have made historical contributions to humans. However, it breaks the biogeochemical cycles of carbon, nitrogen, and phosphorus and cannot remove hazardous materials including viruses and nano/microplastics. Therefore, we rethought the conceptual revolution of principles of sewage treatment in the 1890s, that is, “the replacement of a philosophy that saw sewage purification as the prevention of decomposition with one that tried to facilitate the biological processes that destroy sewage naturally”. We proposed a promising sewage treatment system based on physical separation, which mainly consists of the source separators and the insoluble-pollutants separators, soluble-pollutants separators, and the wastewater heat recovery devices in wastewater treatment plants. By using the promising system, the carbon in wastewater will be recovered by sending biosolids directly into the soil after removing the hazardous materials and organic toxicity. The nitrogen and phosphorus in wastewater will be sent back into the soil or be used for hydroponics rather than be mineralized. The thermal energy in wastewater will be recovered and reused, and the hazardous materials will be removed. As a result, the promising system will turn the wastewater treatment system with high resource and thermal energy waste and high energy consumption into a no-chemicals, green factory. At present, nonetheless, it is still urgent to develop more advanced insoluble-pollutants separators and soluble-pollutants separators with high separation efficiency and low energy consumption, especially volume separators. Because the volume separators (e.g., functionalized sand filters) have the potential for replacing the surface separators (e.g., membranes).

Keywords: biogeochemical cycles; wastewater treatment; resource recycling; heat pump; carbon neutrality; sustainable development



Citation: Tian, J.; Chen, X. Physical Separation: Reuse Pollutants and Thermal Energy from Water. *Water* **2023**, *15*, 1196. <https://doi.org/10.3390/w15061196>

Academic Editor: Laura Bulgariu

Received: 30 January 2023

Revised: 5 March 2023

Accepted: 9 March 2023

Published: 20 March 2023



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1. Introduction

Since the industrial revolution, to meet demands for energy and food, people have moved a large amount of carbon, nitrogen, and phosphorus, respectively, from the underground to the atmosphere, from the atmosphere to the fields, forests, and rivers, and from the underground to the fields, forests, and rivers. As a result, the biogeochemical cycle found in 1926 [1] was broken [2]. Excess carbon, nitrogen, and phosphorus is polluting our environment [3,4]. The pollution of chemicals including carbon, nitrogen, and phosphorus is causing and exacerbating global climate change [5,6] (Figure 1) and many other global environmental issues and emerging environmental issues. Where the global environmental issues refer to the ozone layer depletion [7], biodiversity decline [8], marine oil pollution [9], eutrophication [10], persistent organic pollutants [11], and mercury pollution [12]. Whereas the emerging environmental issues refer to the pollution of pharmaceutical and personal care products [13], disinfection by-product pollution [14] (Figure 2), and microplastic pollution [15]. Ironically, nonetheless, mineable resources of many essential elements are limited [2]. The biogeochemical cycle of every element has a planetary boundary that should not be transgressed [16].

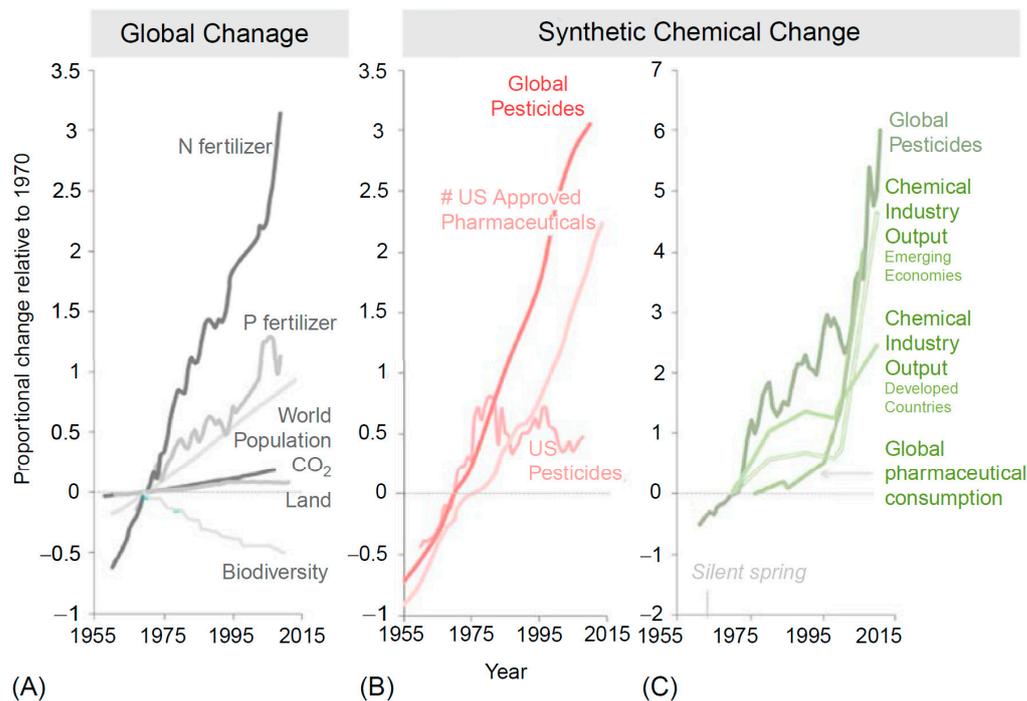


Figure 1. (A) Trajectories for drivers of global environmental change; (B) increases in the diversity of US pharmaceuticals and the application of pesticides within the US and globally; (C) trends for the global trade value (in USD) of synthetic chemicals and for the pesticide and pharmaceutical chemical sectors individually. To allow comparison, all trends are shown relative to values reported in 1970 (Reprinted with permission from Ref. [5]. 2017, The Ecological Society of America).

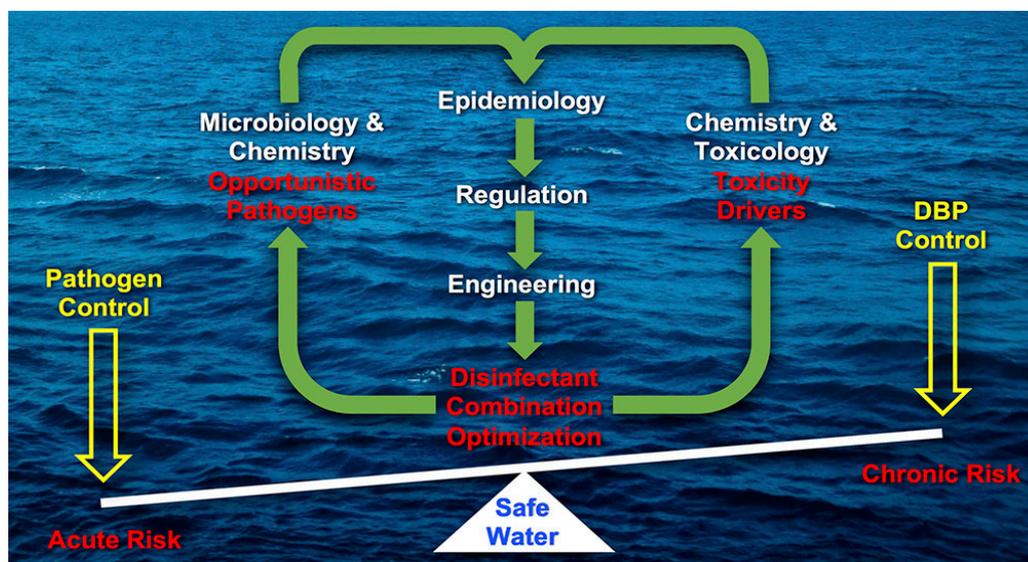


Figure 2. Drinking Water Disinfection Byproducts (DBPs) and Human Health Effects: Multidisciplinary Challenges and Opportunities. (Reprinted with permission from Ref. [14]. 2017, American Chemical Society).

2. Broken Carbon, Nitrogen, and Phosphorus Cycles

2.1. Broken Carbon Cycles

With respect to carbon, the amount of carbon dioxide produced by the burning of fossil fuels has increased rapidly. Global warming is becoming faster and faster [17,18], and hence the Antarctic Ice Sheet is losing mass at an accelerating rate [19,20]. Researchers [21]

recently found that the Antarctic Ice Sheet presents many temperature thresholds beyond which ice loss is irreversible. Considering the great harm of climate change (e.g., the effects on the amphibian [22], terrestrial ecosystems [23], and crop [24]), the rapid reduction of greenhouse gas emissions has become imperative and increasingly urgent, especially the realization of carbon neutrality [25]. It should be noted that many substances should be used as resources rather than energy sources. Otherwise, the greenhouse effect and the destruction of the carbon cycle will intensify and even become irreparable.

For example, the carbon stored in the top meter of the world's soil is more than three times [26] (or at least more than two times [27]) the amount of carbon held in the atmosphere, and two-thirds of it is in the form of organic matter [28]. In terms of the elemental composition of soil organic matter, soil organic carbon accounts for approximately 50% [29]. Nevertheless, the soil's organic carbon is vulnerable to carbon losses through biological degradation. This results are that the greenhouse gases are easy to be released to the atmosphere. As a result, global warming is accelerated [26], which inversely accelerates the soil organic carbon losses [30,31].

Since the 19th century, approximately 60% of the world's soil carbon has been lost due to the worldwide intensification of land use and conversion of uncultivated land for food, feed, fiber, and fuel production [27]. As a consequence, agricultural productivity and the ability to provide ecosystem services declined significantly. Thus, increasing soil organic carbon is crucial for carbon neutrality, food production, and the delivery of many interrelated ecosystem services. The soil organic matter containing soil organic carbon should be used as a resource rather than an energy source (e.g., methane converted from the soil organic carbon by anaerobic treatments [26]). To increase global soil organic carbon stocks by 4‰ per year as compensation for the global emissions of greenhouse gases by anthropogenic sources, the '4‰ Initiative: soils for food security and climate' was launched at the 21st Conference of the Parties to the United Nations Framework Convention on Climate Change [32].

2.2. Broken Nitrogen Cycles

With respect to nitrogen, there are two forms of nitrogen in nature: N_2 and all other nitrogen forms as "reactive nitrogen" or "fixed nitrogen" (N_r) [33]. Where N_2 maintains a stable atmosphere for life on earth, whereas N_r is important for all life on earth, especially for plant growth. Accordingly, in the past 100 years, people produce numerous nitrogen fertilizers by converting nitrogen in the air to ammonia [27], which accounts for 63% of anthropogenic nitrogen fixation [33]. Nevertheless, in terms of the whole food chain, only nearly 20 percent of the reactive nitrogen added in farming ends up in human food, and nearly 80 percent is wasted as excess reactive nitrogen and N_2 to the environment. Excess reactive nitrogen is polluting our environment: the water quality, ecosystems, biodiversity, soil quality, greenhouse-gas balance, and air quality [34]. For example, NH_3 causes eutrophication and decreases biodiversity. NO_2 and NO are the main air pollutants. NO_3 forms particulate matter in the air. Ammonium-based fertilizers make the soil acidic. The Global Warming Potential of N_2O is 265 times that of carbon dioxide according to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, and causes ozone layer depletion.

However, although reactive nitrogen has far exceeded the demand for use and is polluting our environment [16], the global consumption of nitrogen fertilizers are still increasing rapidly. For example, from 1980 to 2016, the consumption of all types of nitrogen fertilizers in South Asia increased from 4.7 to 21.8 million metric tonnes, and that in East Asia increased from 15 to 34 million metric tonnes [33]. Globally, nearly 200 million tonnes of reactive nitrogen are lost to the environment per year as excess reactive nitrogen and N_2 . If the fertilizer price is USD1 per kg N, the cash loss per year will be nearly USD200 billion. Therefore, it is urgent to convert current nitrogen cycle pollution to nitrogen circular economy by recovering and reusing reactive nitrogen through physical separation, rather

than continuously converting N_2 to reactive nitrogen with a high cost and then converting excess reactive nitrogen to N_2 .

2.3. Broken Phosphorus Cycles

With respect to phosphate, which is also usually used to produce fertilizers, the phosphate cycle is destroyed more severely. Unlike the aforementioned carbon and nitrogen, there are no substitutes for phosphate, and phosphate cannot be obtained from the air. Morocco controls most of the global phosphorus reserves, and Morocco, China, and Algeria control more than 85% of the known global phosphorus reserves [2]. The power concentration is significantly higher than that of oil, where a dozen members of the Organization of the Petroleum Exporting Countries hold 80% of the global oil reserves. What is worse, calculated by increasing the demand for phosphorus by 3% each year, the remaining phosphate rock will approximately run out in 38 years [35]. Phosphate rock may be a strategic material for many countries in the near future.

In summary, since the industrial revolution, the biogeochemical cycles of carbon, nitrogen, and phosphorus have been severely broken by human activities. Therefore, we should strive to establish a circular economy as early as possible to fix the broken biogeochemical cycles. Specifically, we should minimize the use of chemicals (e.g., carbon, nitrogen, and phosphorus) and the greenhouse gases emissions by recovering and reusing them. All wastewater treatment plants should reuse pollutants and thermal energy in water by making full use of physical separation without the use of chemicals [36], especially the physical separation technologies.

3. Conventional Sewage Treatment Based on Biological and Chemical Methods

At present, however, the treatment process of domestic wastewater is a largely one-way flow of nutrients and is almost down the drain [37]. Most domestic wastewater is treated utilizing the aerobic ‘activated-sludge process’, which mixes the domestic wastewater with the bacteria and air to degrade rather than reuse the pollutants including carbon, nitrogen, and phosphorus [38]. Thus, many researchers proposed to recover chemical energy contained in domestic wastewater by using anaerobic treatments [37,39,40] and microbial electrochemical cells [37] to produce energy-rich chemicals (e.g., methane [37,39,40] and hydrogen gas [37]) and electrical power [37] rather than to produce carbon dioxide (Table 1). Some researchers even suggested to use microbial electrolysis cells and microalgae cultivation to recover the carbon in domestic wastewater and to capture carbon by producing carbonates [41].

Nevertheless, the carbon-rich organic matter (i.e., the so-called “carbonaceous chemical oxygen demand” or “COD”) has a higher exergy content than methane, carbon dioxide [26,27], and carbonates [41], which should be maintained if possible [42] (Figure 3). Namely, the carbon-rich organic matter contained in the wastewater should be converted into organic products with highly valuable such as biopolymers, rather than being converted first into methane and then into carbon dioxide [42] or being converted into carbonates. Both methane and carbon dioxide are greenhouse gases, and the global Warming Potential of methane is 28 times that of carbon dioxide according to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. The product value of carbonates is too low.

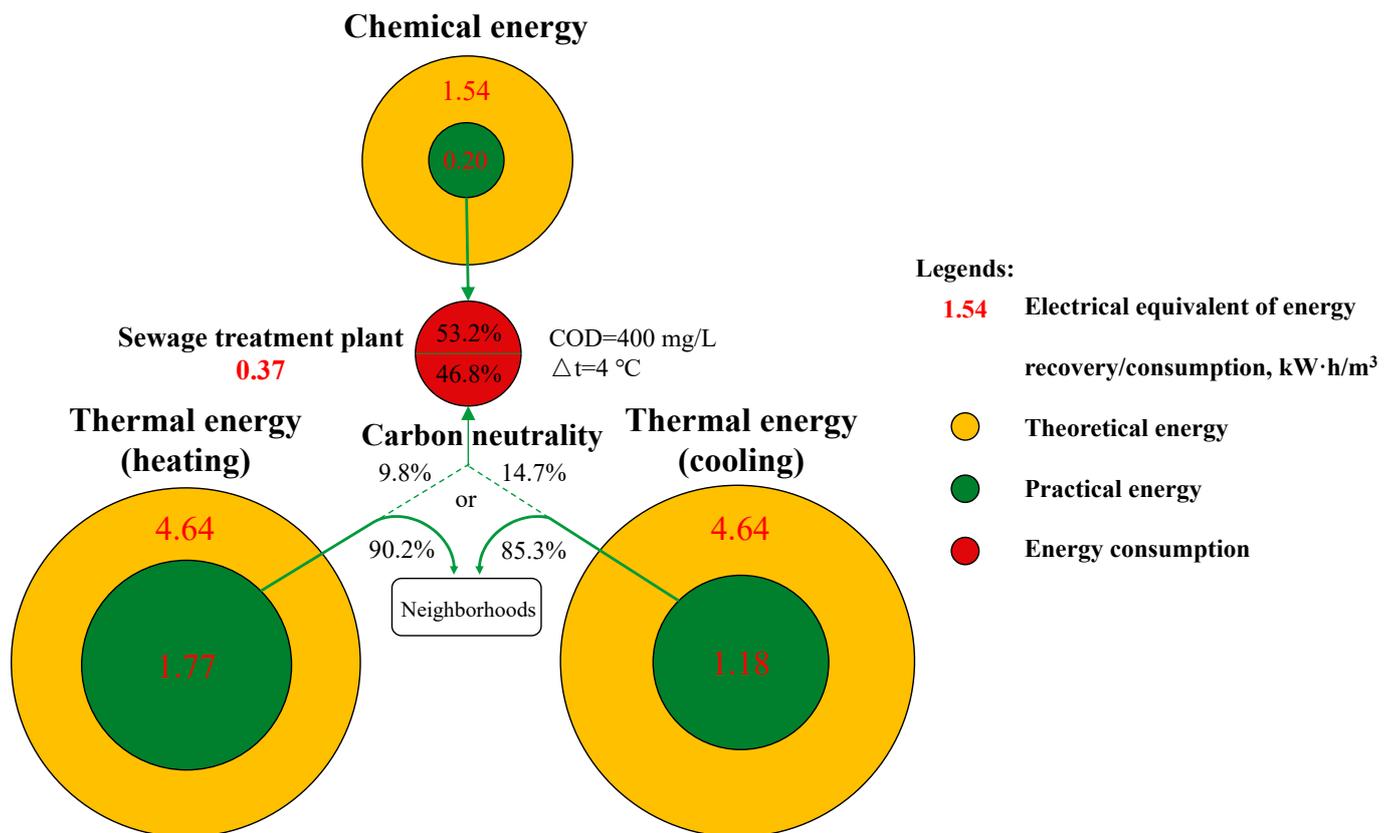
Based on the evaluation of a wastewater treatment plant with a feed COD of 400 mg/L, the recoverable potential of thermal energy in the effluent of the wastewater treatment plant is at least 6–8 times higher than that of the chemical energy [43]. Nonetheless, chemical energy is usually treated as the only recoverable energy contained in wastewater [37,41,42]. The thermal energy, which can be easily employed for the heating or cooling of buildings [44], the drying of dewatered sludge [42], the heating of biogas digesters [45], the agricultural greenhouses [42], the melting of snow [46], and the heating of domestic hot water [47], is usually ignored [42,48].

Table 1. Flow of nutrients and thermal energy in various wastewater treatments.

	Nutrients and Thermal Energy	Flow of Matter and Energy
Aerobic activated sludge process [38]	C	$\text{COD} + \text{O}_2 \rightarrow \text{CO}_2 + \text{H}_2\text{O}$
	N	$\text{NH}_4^+ + \text{O}_2 \rightarrow \text{NO}_3^- + \text{H}^+ + \text{H}_2\text{O}$ $\text{NO}_3^- + \text{COD} \rightarrow \text{N}_2\uparrow + \text{CO}_2 + \text{H}_2\text{O}$
	P	$\text{PO}_4^{3-} + \text{Al}^{3+} \rightarrow \text{AlPO}_4\downarrow$
	Thermal energy	Waste
“Denitrifying phosphorus removal bacteria + Anaerobic treatments” [39]	C	$\text{COD} \rightarrow \text{CH}_4$
	N, P	$\text{NH}_4^+ + \text{O}_2 \rightarrow \text{NO}_3^- + \text{H}^+ + \text{H}_2\text{O}$ $\text{NO}_3^- + \text{COD} + \text{PO}_4^{3-} \rightarrow \text{N}_2\uparrow + \text{CO}_2 + \text{H}_2\text{O} + \text{Phosphate}$
	Thermal energy	Waste
“Anaerobic treatment + Anammox process” [40]	C	$\text{COD} \rightarrow \text{CH}_4$
	N	$\text{NH}_4^+ + \text{NO}_2^- \rightarrow \text{N}_2\uparrow + 2\text{H}_2\text{O}$ $\text{NH}_4^+ + 1.5\text{O}_2 \rightarrow \text{NO}_2^- + 2\text{H}^+ + \text{H}_2\text{O}$ Together yield: $2\text{NH}_4^+ + 1.5\text{O}_2 \rightarrow \text{N}_2\uparrow + 2\text{H}^+ + 3\text{H}_2\text{O}$
	P	$\text{PO}_4^{3-} + \text{Al}^{3+} \rightarrow \text{AlPO}_4\downarrow$
	Thermal energy	Waste
“Anaerobic membrane bioreactor + Microbial electrochemical cells + Ion exchangers” [37]	C	$\text{COD} \rightarrow \text{CH}_4 + \text{Electricity}$
	N, P	N, P \rightarrow Fertilizer
	Thermal energy	Waste
“Microbial electrolytic carbon capture + Microalgae cultivation” [41]	C	$\text{COD} + \text{CO}_2 \rightarrow \text{Carbonates}$
	N, P	N, P \rightarrow Microalgae cultivation
	Thermal energy	Waste

What is worse, the domestic wastewater in China presents the characteristics of low organic contents (the average of which is just approximately 267 mg/L [49], which is markedly lower than the above-mentioned 400 mg/L [42]) and high inorganic solids. This results in the fact that anaerobic digestion cannot be well operated in China, and hence only less than 5% [49] (even 3% [50]) of wastewater treatment plants in China are equipped with anaerobic digestion facilities. On the contrary, if the thermal energy is extracted from the untreated wastewater rather than the above-mentioned effluent of the wastewater treatment plant, the recoverable potential of thermal energy will be significantly higher [44,47,51,52]. Because long-distance conveyance of wastewater from the source to the wastewater treatment plant and wastewater treatments will cause a lot of thermal energy loss.

In addition to the aforementioned squander of the carbon-rich organic matter and the thermal energy in wastewater, another huge squander is nitrogen and phosphorus (Table 1), which are valuable for making fertilizers. At present, more than 80% of nitrogen [37] and 90% of phosphorus [53,54] are squandered in the wastewater treatment processes. As a result, excess reactive nitrogen [33] and phosphorus [2] are polluting our environment, whereas we are still producing reactive nitrogen with a high cost to produce nitrogen fertilizer [33], and are still mining limited phosphate rock [2,35] to produce phosphorus fertilizer. In terms of the whole food chain, only nearly 20 percent of the reactive nitrogen added in farming ends up in human food [33].



Energy recovery from wastewater: Heat over organics

Figure 3. Energy balance towards the carbon-neutral operation of the evaluated sewage treatment plant. (Reprinted with permission from Ref. [42]. 2019, Elsevier).

Therefore, it is urgent to convert nitrogen cycle pollution into a nitrogen circular economy by recovering and reusing the reactive nitrogen and phosphorus as fertilizers to replace the production of concentrated fertilizer. That is, both the production of concentrated fertilizer and the conversion from reactive nitrogen to nitrogen gas should be minimized. Accordingly, the wastewater treatments that convert reactive nitrogen into nitrogen gas [38–40] are harmful to the formation of a nitrogen circular economy [33]. The wastewater treatments that recover and reuse reactive nitrogen and phosphorus [37,41] are better.

Besides, the “wet sludge” produced by wastewater treatment (5–10 kg/m³ of the treated water) needs drying and disposal (in landfill or on land) or incineration, which accounts for 30–50% of the treatment facility’s overall costs [37]. What is worse, a large number of hazardous materials (e.g., viruses, microplastics, bacteria, and heavy metals) are left in the sewage and sludge [36]. The hazardous materials further migrate around the world and cause the ozone layer depletion [7], biodiversity decline [8], marine oil pollution [9], eutrophication [10], persistent organic pollutants [11], mercury pollution [12], pollution of pharmaceutical and personal care products [13], disinfection by-product pollution [14], and microplastic pollution [15]. Their effects may be carcinogenic or even genotoxic [14,55].

4. Promising Sewage Treatment Based on Physical Separation

Therefore, we should rethink the conceptual revolution of the principles of sewage treatment in the 1890s [56], that is, “the replacement of a philosophy that saw sewage purification as the prevention of decomposition with one that tried to facilitate the biological processes that destroy sewage naturally”, and then combine the source-separation idea [57] to develop less-energy-consumption physical separation technologies to recover wastewater resources and thermal energy without the use of chemicals [58–60].

For example, we can use physical separation to replace both the conventional aerobic activated-sludge treatments and the anaerobic treatments [37]. The promising sewage treatment based on physical separation (Figure 1) could remove the hazardous materials (e.g., the viruses, bacteria, nano/microplastics, and heavy metals) contained in the wastewater, and could reverse the aforementioned nutrients and energy costs to form the circular economy. The corresponding physical treatment facilities can be classified into four types: source separators, insoluble-pollutants separators, soluble-pollutants separators, and wastewater heat recovery devices.

According to the difference in the thickness of the separation medium, the physical separation can be divided into surface separation (e.g., membrane separation) and volume separation (e.g., functionalized sand filtration). Unlike high-energy-consumption surface separators that mainly rely on the aperture, the volume separator has the potential for utilizing the adsorption function of the functionalized natural materials in the separators to achieve the separation of soluble and insoluble micro-nano contaminants. Researchers [61] have recently found that, by controlling the internal unevenness of the separation medium, the energy consumption of the nanofiltration membrane can be greatly reduced. This demonstrates that the separation efficiency and energy consumption of the separator not only depends on the aperture and the thickness of the separation medium but also depends on the internal unevenness of the separation medium. Therefore, volume separation has the potential for replacing surface separation, and we need to develop a high-efficient and low-energy-consumption volume separation theory, techniques, and equipment (e.g., functionalized sand filtration) that can achieve ultrafiltration, nanofiltration, and reverse osmosis.

If the promising sewage treatment based on physical separation (Figure 4) is applied, the chemical energy contained in the wastewater will be recovered by using the biosolids directly to the soil after removing the hazardous materials (e.g., viruses, nano/microplastics, bacteria, and heavy metals) and organic toxicity. That is, the carbon will be recovered as a carbon resource rather than chemical energy. This will definitely promote the remediation of the broken biogeochemical cycle by decreasing the movement of carbon from the soil to the atmosphere [2]. The emissions of greenhouse gases (e.g., carbon dioxide emitted by the aerobic activated sludge process and methane emitted by the anaerobic processes [37,41]) in wastewater treatment plant will decrease significantly.

The thermal energy contained in the wastewater, which is usually ignored and hence wasted, can be recovered and reused in all the areas that need thermal energy. For example, the drying of dewatered sludge [42], the heating or cooling of buildings [44], the heating of biogas digesters [45], the agricultural greenhouses [42], the melting of snow [46], and the heating of domestic hot water [47]. If we want to recover the thermal energy contained in the wastewater as much as possible, we need to comprehensively and selectively recover the thermal energy from both the raw wastewater and the wastewater in wastewater treatment plants according to local needs and conditions.

The reasons are as follows: if the wastewater in the wastewater treatment plant is chosen as the thermal source, owing to the existence of insoluble-pollutants separators, the blockage, fouling, and corrosion issues of the wastewater heat exchanger will be greatly released. Nonetheless, the supply distance and the thermal energy loss produced by long-distance transportation and wastewater treatments will become critical barriers [42]. If the raw wastewater is chosen as the thermal-energy source, the supply distance and the thermal energy loss produced by long-distance transportation and wastewater treatments will be avoided. Nevertheless, the blockage, fouling, and corrosion issues of the wastewater

heat exchanger will become critical barriers [51,52]. Fortunately, unlike the wastewater treatment plants mainly based on biological treatments [42], the thermal-energy recovery from the raw wastewater promotes the wastewater treatments in a promising system based on physical separations. In recent years, researchers [62,63] have developed an efficient de-foulant hydrocyclone with a reflux function for the thermal energy recovery from raw wastewater (Figure 5), and have reviewed the enhanced-separation hydrocyclone technologies [64,65].

The energy-saving technologies and renewable energy technologies will boost the energy recovery from wastewater and offset the energy consumption of wastewater treatments in the promising system proposed in this study. For example, the thermal energy recovered by the wastewater source heat pump can be used for drying the sludge, and the electricity produced by the solar photovoltaic system can be used for providing electricity for the wastewater source heat pump. Renewable energy technologies, such as photovoltaic technology, photothermal technology, wind power technology, and geothermal technology, should be selected according to local specific conditions. The energy-saving technologies should be further developed and applied, especially those used for the physical separation technologies (e.g., the hydrocyclone [66–69], functionalized sand filter [70] (Figure 6), fiber coalescer [71,72], gas cyclone [73,74] (Figure 7), and membrane [58]).

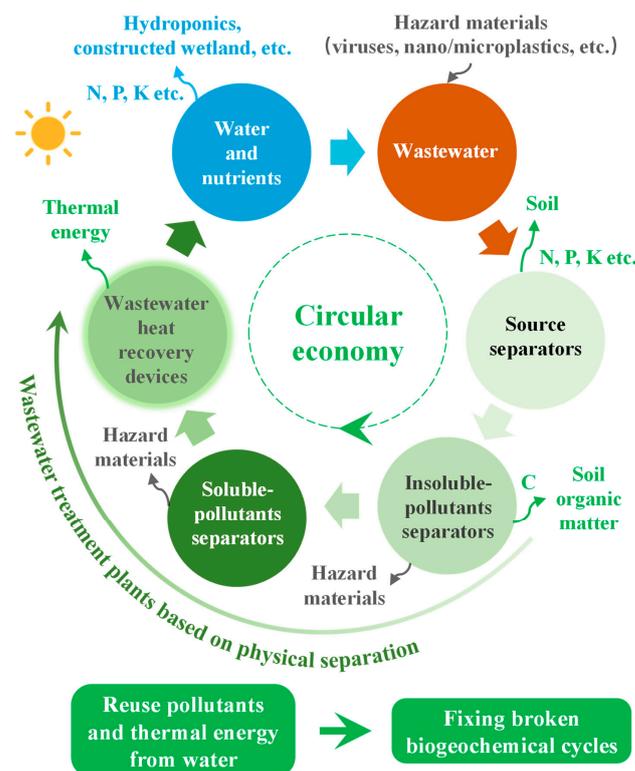


Figure 4. The promising sewage treatment based on physical separation.

Overall, owing to the fact that the promising system proposed in this study is mainly based on physical separation, the hazardous materials in the wastewater will be collected and removed. The carbon, nitrogen, and phosphorus will be recovered and reused. The broken biogeochemical cycle [2] will be fixed (Figure 8). The thermal energy contained in the wastewater will be recovered and reused, and renewable energy will be used. The emissions of greenhouse gases in wastewater treatment systems will decrease markedly. The promising system proposed in this study will turn the wastewater treatment system with high resource waste and high energy consumption into a no-chemicals, green factory, which can collect and remove hazardous materials, and recover and reuse pollutants and thermal energy.

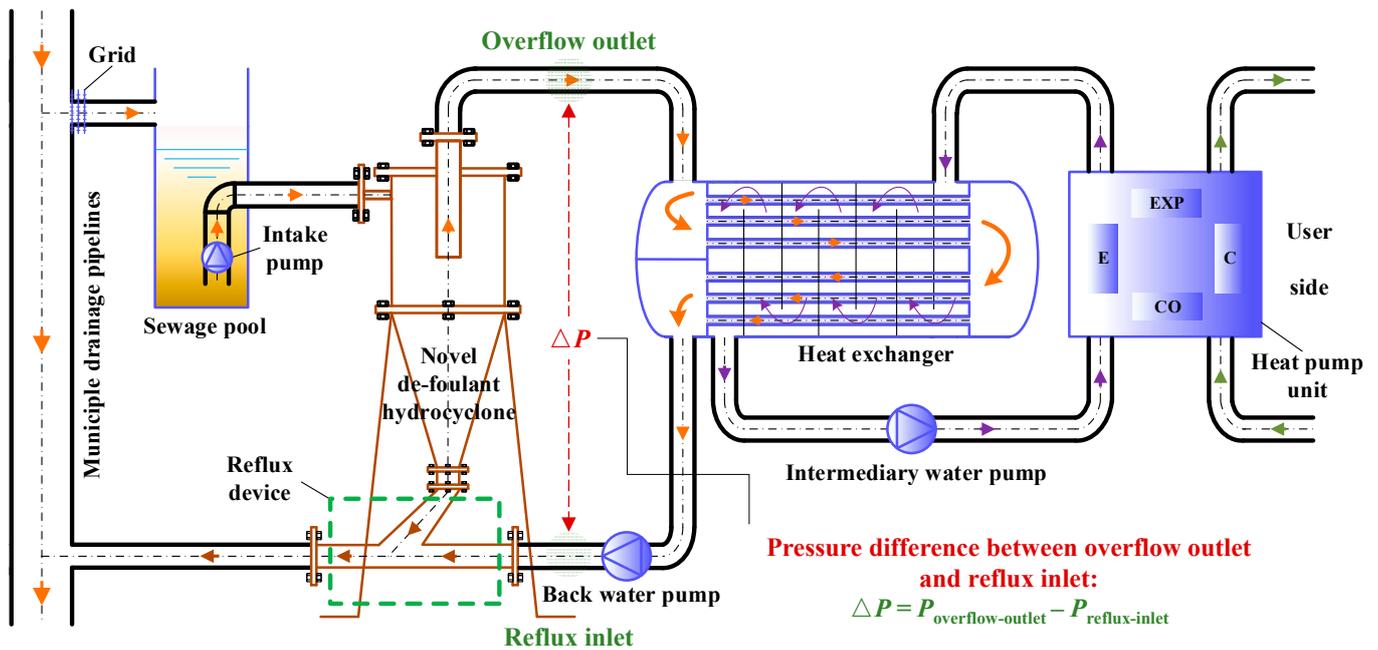


Figure 5. Schematic of a practical project of sewage source heat pump with the de-foulant hydrocyclone. (Reprinted with permission from Ref. [62]. 2021, Elsevier).

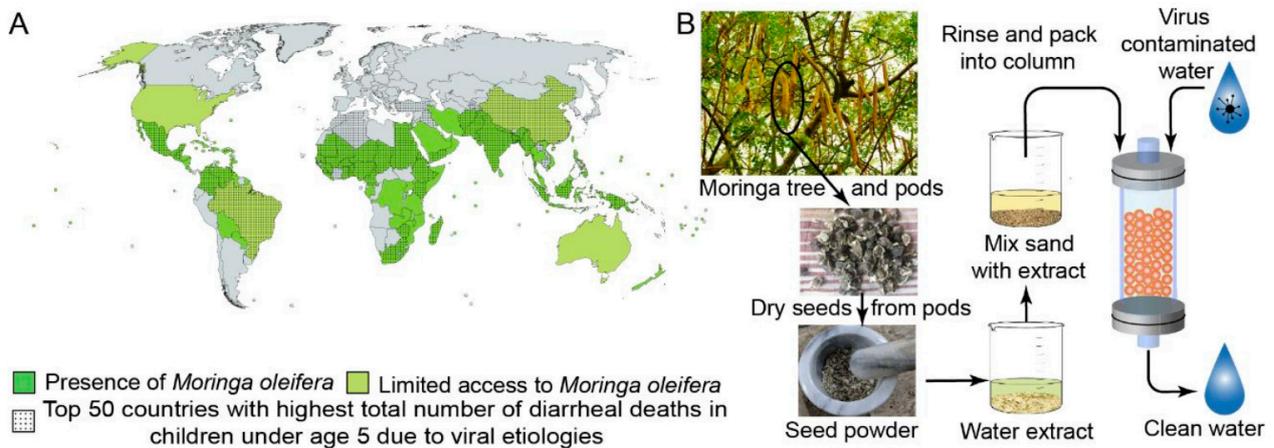


Figure 6. 7 Log Virus Removal in a Simple Functionalized Sand Filter. (A) The geographical distribution of *Moringa oleifera*; (B) A simple functionalization procedure using *Moringa oleifera* seed water extract was used to improve the pathogen removal efficiency of sand filters. (Reprinted with permission from Ref. [70]. 2019, American Chemical Society).

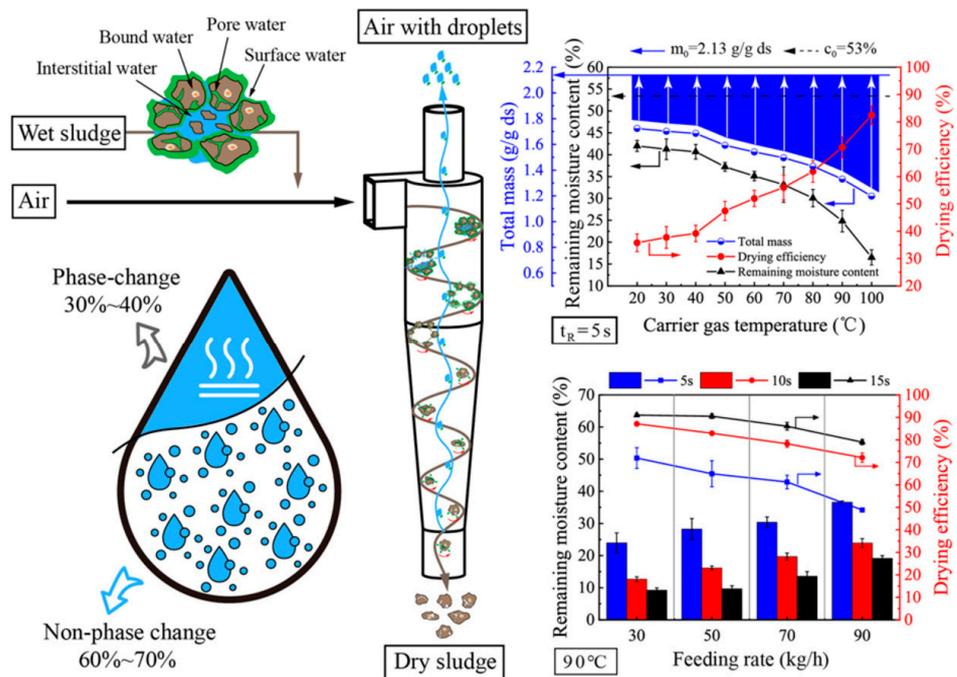
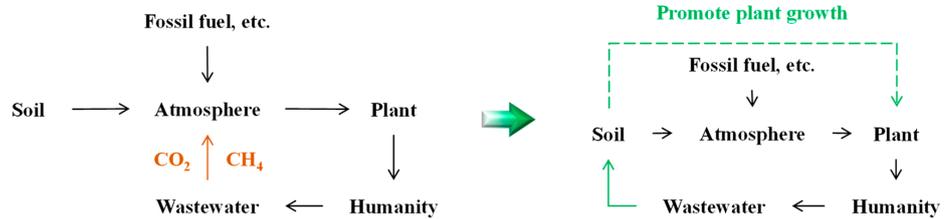
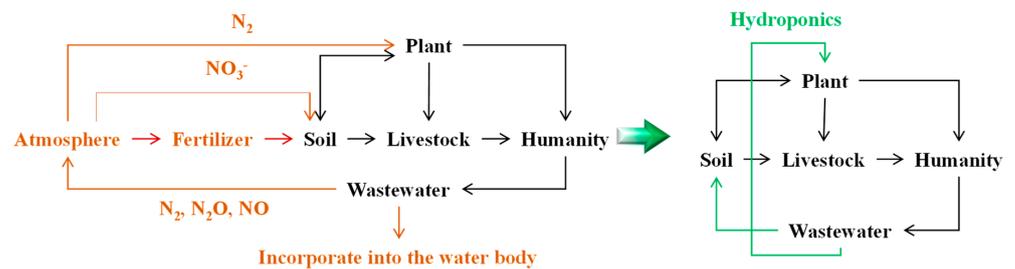


Figure 7. Sludge low-temperature drying with mainly non-phase change in mere seconds based on particle high-speed self-rotation in cyclone. (Reprinted with permission from Ref. [73]. 2022, Elsevier).

(a) Carbon cycle



(b) Nitrogen cycle



(c) Phosphorus cycle

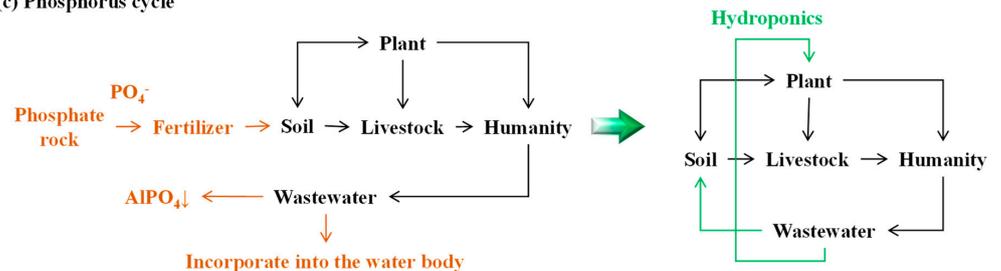


Figure 8. Carbon, nitrogen, and phosphorus cycles of the conventional sewage treatment based on biological and chemical methods (Left) and the promising sewage treatment based on physical separation (Right).

5. Conclusions

Based on the above discussion of the potential of reusing the pollutants and thermal energy from water with physical separation, several conclusions can be drawn.

- (1) The conventional sewage treatment based on biological and chemical methods breaks the biogeochemical cycles (e.g., the carbon, nitrogen, and phosphorus cycles) and cannot remove hazardous materials including the viruses, microplastics, bacteria, and heavy metals. Accordingly, we should rethink the conceptual revolution of the principles of sewage treatment in 1890s [56], that is, “the replacement of a philosophy that saw sewage purification as the prevention of decomposition with one that tried to facilitate the biological processes that destroy sewage naturally”.
- (2) The carbon in the wastewater should be sent back to the soil rather than be used to produce carbon dioxide, methane, or carbonate. The nitrogen and phosphorus in the wastewater should be sent back to the soil or used for hydroponics rather than being mineralized. The thermal energy in the wastewater should be recovered and reused; whereas the chemical energy in the wastewater should be maintained rather than be recovered by producing methane and carbon dioxide. The hazardous materials should be removed.
- (3) The proposed promising sewage treatment system based on physical separation mainly consists of the source separators and the insoluble-pollutants separators, soluble-pollutants separators, and the wastewater heat recovery devices in the wastewater treatment plants;
- (4) The proposed promising sewage treatment system based on physical separation has the potential to replace conventional sewage treatment based on biological and chemical methods to fix the broken biogeochemical cycles (e.g., the carbon, nitrogen, and phosphorus cycles).
- (5) It is urgent to develop more advanced insoluble-pollutants separators and soluble-pollutants separators with high separation efficiency and low energy consumption [58–60], especially volume separators. Because the volume separators (e.g., functionalized sand filters) have the potential for replacing the surface separators (e.g., membranes).

Author Contributions: Conceptualization, J.T. and X.C.; writing—original draft preparation, J.T.; writing—review and editing, X.C.; supervision, X.C.; project administration, J.T.; funding acquisition, J.T. All authors have read and agreed to the published version of the manuscript.

Funding: This work was financially supported by the National Natural Science Foundation of China (52000070).

Data Availability Statement: No new data were created.

Conflicts of Interest: The authors declare no conflict of interest.

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