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Review

Recent Advances in Resource Utilization of *Huangshui* from Baijiu Production

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Abstract: *Huangshui* is a typical organic wastewater in Chinese Baijiu production, with high pollution and valuable ingredients. Conventional wastewater treatment leads to resource-wasting and environmental pollution. It is urgent that the demand for effective *Huangshui* treatment with the development of the Baijiu-making industry. This review systematically summarizes recent studies, revealing the main characteristics and application of *Huangshui*, focusing on the application of the rich microbial resources and flavor substances, which provides a practical approach to cascade and full use of *Huangshui* in medicine, cosmetic, functional food, fertilizer, and wastewater treatment fields. Further research suggested that *Huangshui* can also be used as an external carbon source for the denitrification system or as an organic liquid water-soluble fertilizer for more fruits and grains. The applications favor improving production efficiency and lowering pollutant emissions and introduce novel concepts for the sustainable development of related industries. Thus, Chinese Baijiu plants can achieve the near-zero emissions of wastewater and cleaner production.

Keywords: Chinese Baijiu' *Huangshui*; *Huangshui* resources; *Huangshui* property; functional components; cascade and full utilization

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1. Introduction

Chinese Baijiu, one of the six most famous distilled liquors in the world, is widely consumed in East Asia due to its unique flavor and long history. Compared with other distilled spirits, the production of Chinese Baijiu is a unique brewing process, including saccharification, natural solid-state fermentation, and flavor formation simultaneously, being described in Figure 1. Daqu powder from the fermented wheat inoculates with various microbes, which can serve as microbial agents for Baijiu production [1]. The microbes undergo enrichment and growth for 30–40 days in the solid-state fermentation. The pollution and energy consumption from Baijiu fermentation mainly come from grain, brewing processes, and transportation, as well as wastewater treatment and solid waste disposal [2]. Additionally, during the fermentation process, free water gradually seeps into the bottom of the fermentation pit along with the dissolution of different substances, ultimately forming *Huangshui* [3]. Such organic water is one of the main by-products of the solid fermentation of Baijiu, which is a brown, viscous, and odorous liquid. In the Baijiu industry, about 3500 tons of *Huangshui* will be formed when 10,000 tons of Baijiu is produced [3]. According to the above data, the output of Baijiu is 8 million kiloliters, in which 2.4–3.2 million tons of *Huangshui* will be produced by 2025 in China. However, the large amount of organic matter in *Huangshui* is not fully utilized to form high-value-added products due to the limitations of solid-state fermentation and conventional distillation processes. In 2020, the effluent of brewing wastewater accounted for 1.83%

of the total industrial wastewater effluent in China, and the effluent of chemical oxygen demand (COD_{Cr}) accounted for 3.46% of the total industrial wastewater COD_{Cr} effluent (China). The reduction in wastewater pollutants and improvement in resource recycling are important ways to affect the sustainable development of the brewing industry.

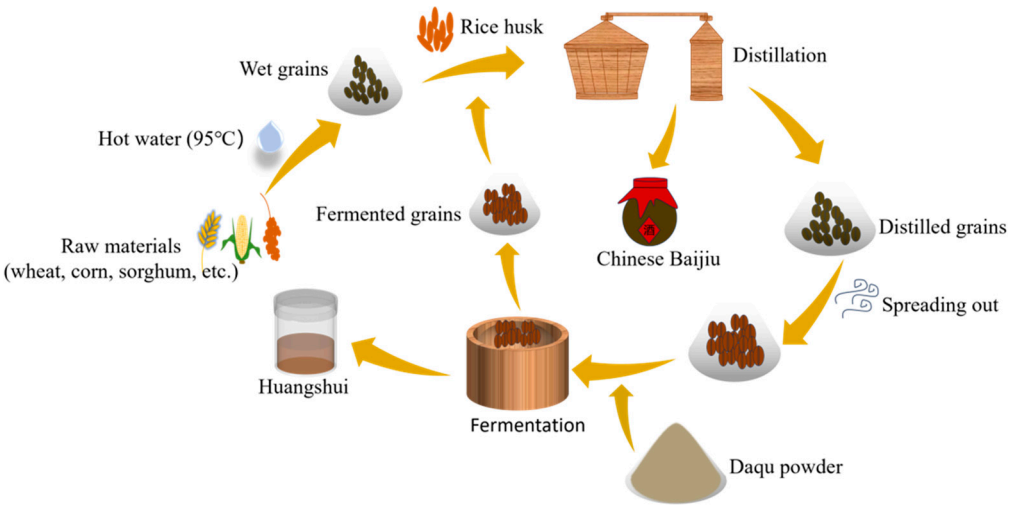


Figure 1. Chinese Baijiu and its by-products production process.

Huangshui has complex components and contains a large amount of organic matter. Based on the huge yield and high organic content of *Huangshui*, its treatment is difficult and costly. A large amount of *Huangshui* causes high treatment costs and great pressure on Baijiu factories. If not properly treated, it will result in high environmental risk and waste of resources. Although the anaerobic process of treating *Huangshui* is effective, it generates a large number of harmful gases (e.g., CH_4, H_2S, and CO_2) and fermentation residue. In addition, *Huangshui* is also rich in many nutrients such as starch, polysaccharides, protein, ammonia nitrogen, organic acids, alcohols, and esters [4,5].

Waste treatment and environmental protection are the two main concerns for the sustainability of the Baijiu industry. Consequently, many studies have focused on how to recycle and utilize distilled spent grain (DSG), *Huangshui*, and waste alcohol [2,6,7]. Most studies have explored the applications of DSG, *Huangshui*, and waste alcohol in food fungi cultivation, livestock feed, energy generation, organic acid, and organic solvent preparation, as well as biochar and organic fertilizer production [2,8]. Recently, other reports mainly paid attention to the extraction of polysaccharides from *Huangshui* and their applications in health, food preservation, and wastewater treatment [3,9]. He et al. [6] employed a culture medium mixed with DSG and *Huangshui* to produce bacterial cellulose via *Gluconacetobacter xylinus*. The yield of bacterial cellulose from the DSG feed was 3.72 times that from the conventional Hestrin–Schramm feed, obtaining 7.42 g/L. Moreover, its structure and characteristics were similar to those of the Hestrin–Schramm feed. Ma et al. [2] proposed the use of DSG and *Huangshui* to produce fermentable monosaccharides. The treatment process could lower solid waste emissions while obtaining good economic benefits [2]. It avoids the drying process and fully utilizes the components of DSG and *Huangshui* to produce applicable products [2]. Although such a technique contained less organic matter, more wastewater was generated, which could be further treated [2]. This needs to be built and operated together with a wastewater treatment station, which requires the high costs of equipment, thereby raising initial investment.

Recycling organic matter and nutrients from *Huangshui* is currently an effective method for its resource utilization, but it is vitally important to explore a low-cost, highly efficient, and large-scale treatment process and widely applicable utilization models. Nonetheless, there are few reviews on studies associated with *Huangshui*. Jiang et al. [10] only briefly introduced the complex components contained in *Huangshui* and their application areas,

thus existing reports require more systematic elaboration. This review focuses on the advances of *Huangshui* recycling in nutrition, energy, and materials. It proposes advanced approaches and future progress for the full utilization of *Huangshui*. Proposing a full utilization strategy of the total resource reuse of *Huangshui*, which includes process design and integration, is intended to minimize *Huangshui* and carbon emissions, and extend its value chain.

2. *Huangshui* Main Property and Its Resource Value

2.1. Huangshui Main Components and Property

Huangshui is a byproduct of Chinese Baijiu production and is rich in abundant microbes, small molecular sugars, polysaccharides, short-chain fatty acids, amino acids, polypeptides, esters, and alcohols [5]. In addition to containing a large amount of flavor components such as alcohols, aldehydes, acids, and esters, *Huangshui* also contains some nutrients such as sugars, amino acids, humus, yeast autolysates, and a certain number of live functional microbes. Due to the influence of raw materials used in Chinese Baijiu fermentation and their proportion, pit quality, pit age, and operational factors, the chemical composition of *Huangshui* produced by each pit or distillery is quite different, and its chemical component and content are described in Table 1 [4,10–12]. Generally, *Huangshui* is constituted of complex ingredients and includes a large number of organic substances and microbes with 25,000–40,000 mg/L COD_{Cr} and 25,000–30,000 mg/L five-day biochemical oxygen demand (BOD_5) [12]. If it is disposed of improperly, it easily causes secondary pollution.

Table 1. Physicochemical index of different *Huangshui* samples [4,10–12].

Index	Content	Index	Content
pH	3.15–3.77	Lactic acid (mg/100 mL)	3688.08–4863.51
Acidity (mmol/100 g)	35.13–44.93	Succinic acid (mg/100 mL)	73.21–388.07
Starch content (%)	1.41–3.71	Malic acid (mg/100 mL)	247.53–382.34
Alcohol content (% vol)	3.13–5.37	Acetic acid (mg/100 mL)	270.51–416.45
Reducing sugar (%)	0.28–0.86	Citric acid (mg/100 mL)	59.25–95.44
Ester (%)	0.13–0.27	Tartaric acid (mg/100 mL)	47.93–70.64
Tannin and pigment (%)	0.12–0.27	COD_{Cr} (mg/L)	25,000–85,300
Glycerol (mg/100 mL)	537.09–627.36	BOD_5 (mg/L)	25,000–30,000

Under a low pH environment, *Huangshui* could contain potent bacterial inhibitors, whereas, under an alkaline environment, the inhibition was lowered by reducing toxicity, which could originate from the action of organic acids [13]. Similarly, pH is an important indicator of the quality of Baijiu, where the presence of organic acids can lower the pH of *Huangshui*. For example, lactic acid bacteria can synthetize organic acids, which in turn lowers *Huangshui* pH [14], causing its high acidity. The lactic acid of *Huangshui* was the dominant organic acid, accounting for 3688.08–4863.51 mg/100 mL (Table 1). Similarly, Ling et al. [11] described that lactic acid concentration in *Huangshui* was 1001.1–1749.8 mg/100 mL, along with soluble COD_{Cr} of 53,300–85,300 mg/L and pH 3.2–3.8. Other acids such as acetic and malic acids ranged from 270.51 to 416.45 and 247.53 to 382.34 mg/100 mL, respectively (Table 1). Some organic acids have antimicrobial activity, including acetic acid, succinic acid, and lactic acid [11,15]. Low contents of starch, reducing sugar and aldehyde were 1.41–3.71%, 0.28–0.86%, and 1.50–3.60%, respectively. Additionally, the glycerol and ethanol contents accounted for 537.09–627.36 mg/100 mL and 3.13–5.37% vol, respectively. These reducing sugars, glycerol, and ethanol were produced during the fermentation process, which were important components of *Huangshui*. The esters in *Huangshui* are generally produced by two pathways chemical reactions and microbial fermentation, where the fermentation is the main pathway for ester production in *Huangshui*. Microbes such as *Candida albicans* and *Hansenula* yeast have strong ester production ability, converting the organic substances (e.g., aldehydes and ketones) in *Huangshui* into esters via ketone acid

decarboxylation, amino acid deamination decarboxylation, and alcohol oxidation [16]. Moreover, Baijiu *Huangshui* is rich in plant active components, having antioxidant capacity and enhancing immunity [17]. Thus, high contents of organic matter, nutrients, and chemicals in *Huangshui* should be used to explore high-value products and achieve their full utilization, thereby avoiding air and water pollution, and reducing resource loss.

2.2. Huangshui Resource Value

As shown in Figure 2, there are large quantities of flavor components and varieties of beneficial microbes in *Huangshui*, which can be used as the raw materials to produce lactic acid, flavor substances, artificial pit mud, liquid protein feed, vinegar, food preservatives and polysaccharides [18]. Kang et al. [4] also pointed out that *Huangshui* was rich in carbohydrates, organic acids, alcohols, aromatic flavor compounds, and nitrogen-containing matters as well as different functional microbes. As described, *Huangshui* had been used for Baijiu blending after esterifying enzyme treatment, preparing calcium lactate by using the extracted lactic acid, extracting the flavoring components for the compounding of essences, culturing the artificial pit mud for Baijiu production, processing and producing liquid protein feed. In addition, it has been used to produce vinegar and preserve food after simple treatment due to its good antiseptic effects and high safety. Nonetheless, appropriate evaluation and utilization of *Huangshui* are crucial to reducing carbon and pollutant emissions, along with recovering resources. A previous study focused on *Huangshui* composition and its sustainable use, revealing new methods to realize the full utilization of the high-value components of *Huangshui* and the sustainable development of the Baijiu industry [19].

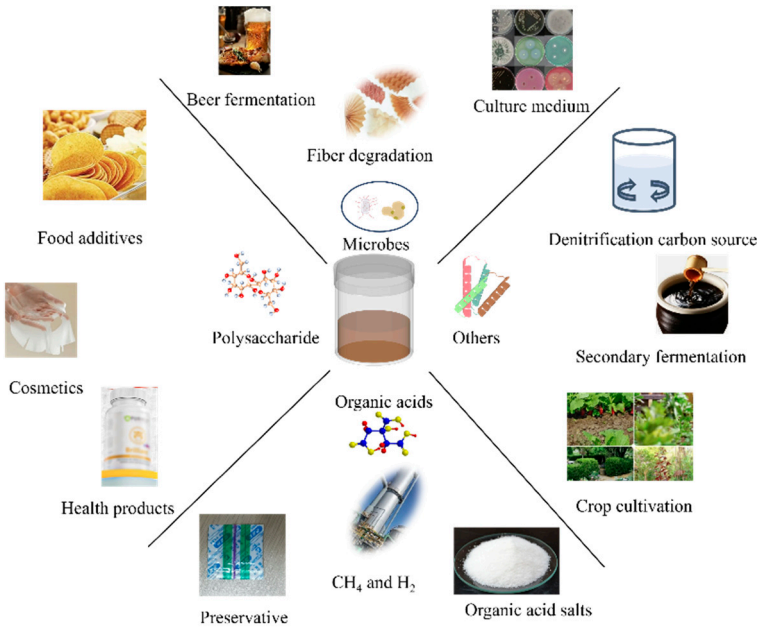


Figure 2. *Huangshui* resourcing: microbes, polysaccharides, organic acid, and others.

2.2.1. Some Microbial Resources

The production of Chinese Baijiu involves the joint action of a diversity of microbes from pit, mud, and fermented grains [20]. The long-term contact between *Huangshui* and the pit mud and fermented grains at the bottom of the pit enriches the essential nutrients for microbial growth, forming an extremely complex micro-ecological system [14,21]. However, due to the complexity of the system, it is difficult for traditional microbial isolation methods to fully reflect the community composition and succession patterns of microbes in *Huangshui*. Therefore, molecular biology techniques have been used in the study of microbial systems. The total number of microbes in *Huangshui* remains at 10^5–10^6 cells/mL, with the majority being bacteria, which can be classified into six phyla: Firmicutes, Actinobacteria, Lentisphaerae, Bacteroidetes, Tenerictes and Actinobacteria (Figure 3a). Except for bacteria,

there are a small number of yeasts. There are many functional microbes in *Huangshui* that have evolved via long-term domestication, such as lactic acid bacteria, bacillus, and yeast [14,20,22], which can become the main microbial resources. They had been screened and identified and proven to play important roles in nutrient metabolism and the synthesis of components including various acids and alcohols [14,20,22]. Gao et al. [3] depicted that the microbial structures of *Huangshui* samples at the phylum level were composed mainly of *Actinobacteria*, *Bacteroidetes*, *Chloroflexi*, *Firmicutes*, and *Proteobacteria* (Figure 3a). While the microbial structure of archaea, the Euryarchaeota phylum dominated in all samples, whose relative abundance ranged from 65.3% to 99.5% [3]. For genus level, *Lactobacillus* possessed an advantage in *Huangshui* samples, which showed a high abundance of *Lactobacillus* [3]. Moreover, the *Lactobacillus* with a high proportion favored exploring functional strains for the high value-added use from *Huangshui* [3]. *Clostridium* and *Caproiciproducens* were positively related to the occurrence of *Methanosaeta*, while *Lactobacillus* revealed a negative relationship with *Methanoplasma*, *Methanobacterium*, and *Methanolobus* [3]. Considering archaea, *Methanosarcina*, *Methanobrevibacter*, *Methanoculleus*, and *Methanobacterium* became dominant genera in *Huangshui* (Figure 3b) [3], being mainly divided into acetoclastic and hydrogenotrophic methanogens for producing CH_4. Synergistic interspecies H_2 transfer-based interactions between hexanoic acid bacteria and methanogenic archaea for the regulation of fermentation in full-flavored Baijiu.

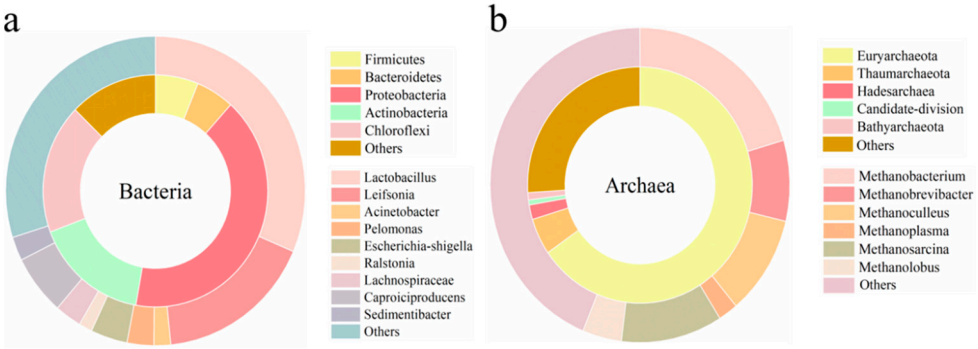


Figure 3. Common microbes in *Huangshui*: The inner circle represents the phylum level, and the outer circle describes the genus level (relative abundance/%) (**a**) bacteria; (**b**) archaea [3].

During the production of Chinese Baijiu, lactic acid bacteria produced organic acids, which lowered the pH of *Huangshui*, which was between 3.15 and 3.77 [4]. This phenomenon was explained by the following fact that *Lactobacillus* showed negative correlations with the occurrence of *Methanobacterium*, *Methanolobus*, and *Methanoplasma*, causing lactic acid accumulation that lowered pH, thus restraining methanogenic activity [23]. This indicates that lactic acid bacteria dominate in *Huangshui*. Kang et al. [4] observed that for all bacterial communities in the *Huangshui* sample, the bacteria dominated by Firmicutes had 18 amplification sequence variants cross lactobacilli, among which the relative abundance of *Lactobacillus acetotolerans* was the highest among three *Huangshui* samples. Gao et al. [14] revealed that with the extension of fermentation time, the main flavor components between *Huangshui* and pit mud obviously rose, and the common microbes gradually increased. Among common microbes, lactic acid bacteria accounted for 56.96% [14]. Moreover, Zhao et al. [24] isolated and purified three strains of lactic acid bacteria from *Huangshui*, which could convert lactic acid into ethanol. In particular, the extracted lactic acid bacteria were able to produce bacteriocins, organic acids, and other compounds, which could be used as biofertilizers in plant production [25].

The most widely applied yeast species is brewing yeast, being the earliest strain to achieve industrial-scale manufacturing. It has been used in some fields such as food and medicine, light industry, energy and chemical industry, and life sciences. Yeasts could conduct carbohydrate metabolism and support flavor development. During the process of Chinese Baijiu fermentation, yeast strains have two functions, alcoholic fermentation

and esterification, participating in the formation of ethanol and esters. Lai et al. [22] isolated three yeast strains from *Huangshui*, being *Candida humilis*, *Saccharomyces cerevisiae*, and *Saccharomyces exigua*. Among them, *Saccharomyces cerevisiae* was the dominant strain, accounting for 70.59% of the total isolated strains. This indicated that the fermentation environment was suitable for the growth of *Saccharomyces cerevisiae*, which was capable of inhibiting the growth of pathogens. In addition, the brewing yeast also produced aromatic compounds, giving the wine a rich pineapple and carambola aroma. Therefore, the screened yeast strains with acid resistance or aroma-raising ability from *Huangshui* provide new ideas for their application in the brewing industry. Many studies mainly focused on bacterial communities. For example, Gao et al. [3] found that *Lactobacillus*, *Methanobacterium*, *Methanobrevibacter*, *Methanoculleus*, and *Methanosarcina* were the dominant genera in *Huangshui*. In addition, hexanoic acid bacteria showed some important characteristics in the cultivation and maintenance of fermentation cellars [14]. It is known that *Huangshui* is rich in lactic acid bacteria, which are able to convert sugars into lactic acid through the glycolysis pathway. Meanwhile, lactic acid can be used to synthesize ethyl lactate through the alcohol acyltransferase pathway, which adds flavor to the body of Baijiu [3]. A previous report had shown that the *Clostridium* genus was able to form a variety of flavor substances including fatty acids and alcohols, which in turn made Baiju have more aromatic taste [23]. However, there has been relatively little report on the composition and screening utilization of the yeast in *Huangshui*. It is necessary to conduct a systematic analysis of fungal community structure from *Huangshui* in the future.

Bacillus plays a crucial role in nutrient metabolism, forming flavor components such as esters, acids, ketones, and aldehydes. Compared with other microbes that do not form buds, Bacillus has the advantages of good environmental adaptability and high survival ability. It is one of the most important microbes to form the main flavor compounds (e.g., ethyl caproate) of strong flavor Baijiu, converting starch, protein, and purine into organic acids such as acetic acid, butyric acid, and caproic acid as well as alcohols [26]. Xie et al. [20] isolated a Gram-positive, strictly aerobic rod-shaped bacteria (*Bacillus aquiflavi* sp. nov.) from *Huangshui*, which could grow under the conditions of 20–45 °C, pH 6.0–10.0, and low NaCl (below 2%). Its main feature was the formation of endophytic spores, resisting external adverse conditions [20]. Desvaux et al. [27] and Sasaki et al. [28] found that the cellulase-producing bacteria in *Huangshui*, such as *Clostridium* spp. and *Bacillus* spp. had the ability to degrade cellulose in mixed raw materials. The bacteria could disrupt the skeletal structure of the cell wall, and release starch from it, which was beneficial for the action of saccharifying enzymes, and further improved raw material utilization, increased fermentation rate, and shortened reaction time [29]. So, the technology of isolating Bacillus from *Huangshui* and applying it to degrade cellulose in crude raw materials effectively expands the biological resources for producing cellulase and promotes cellulase production to meet industrial needs.

2.2.2. Flavor Substances

As described in Figure 4, *Huangshui* is rich in a large quantity of flavor substances, which has attracted considerable attention. It was reported that *Huangshui* contained 33 flavor substances, which mainly included acetic acid, butyric acid, caproic acid, and ethyl caproate [3]. These flavor compounds are related to the flavor components of Baijiu and play a vital role in the formation of the flavor of finished liquor. For Baijiu fermentation, *Lactobacillus* spp. is active in transcribing genes related to the synthesis of flavor substances and their precursors [30]. Bacteria in the *Clostridium* genus can produce various flavor substances such as alcohols, phenols, and organic acids that enable Baijiu's strong flavor [23]. Wang et al. [31] pointed out that the most important odor characteristics of the strong flavor Baijiu, such as taste, sweetness, and flower fragrance, mainly came from esters, which included ethyl hexanoate, ethyl butyrate, ethyl lactate, ethyl acetate, ethyl valerate, butyl hexanoate, and ethyl octanoate. Gao et al. [32] investigated the sources of esters and observed that they were mainly multifunctional volatile fatty acids and linear chain

alcohols produced by microbial fermentation of grains, which promoted the synthesis of esters under the catalysis of enzymes. Kang et al. [4] revealed that *Huangshui* contained 45 volatile compounds, including ethyl hexanoate, hexanoic acid, and 1-butanol, and 224 nonvolatile metabolites like carbohydrates, benzenoids, nucleic acids, and organic acids. And the volatile compounds of esters (13.6–21.7%) and acids (41.4–58.2%) were absolutely dominant in *Huangshui* [3], which could use *Huangshui* to produce esterification solution and seasoning cooking wine [3]. Zou et al. [5] reported *Clostridium* could produce short-chain fatty acids, including acetic butanoic and caproic acids, which could act as main precursors to many flavor compounds. Additionally, the concentration of phenols in *Huangshui* was between 17.9% and 28.3%, implying that *Huangshui* had antibacterial activity and would not deteriorate in a short period of time [4]. In fact, *Huangshui* originated from Baijiu with different flavors containing different contents of acetic acid, butyric acid, caproic acid, ethyl caproate, and ethyl lactate. The differences in flavor substances were consistent with microbial diversity and community structure observed in *Huangshui* [3,4]. In addition, *Huangshui* is rich in sugar because of the presence of bioactive polysaccharides, potentially showing an immunomodulatory role. This favors *Huangshui* being developed as an active composition in functional foods [9].

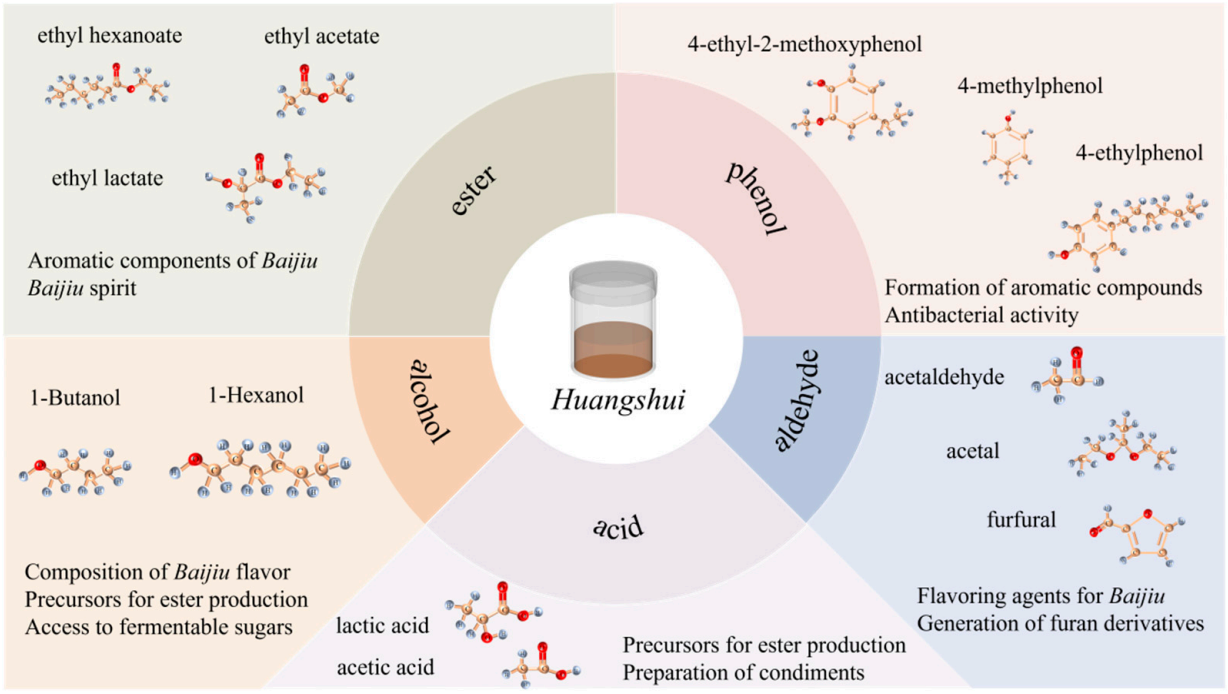


Figure 4. Flavor substances in *Huangshui* [3,10,14].

3. *Huangshui* Utilization Approaches and Techniques

3.1. Exploration and Utilization of Functional Microbes

There are various methods for extracting the high-value components in *Huanghsui*, including physical, chemical biological and combined methods [20]. The extraction process generally involves extraction, purification, and identification [20]. The plate delineation method can isolate strains in *Huangshui*, in which acidic gradient differential culture can isolate parthenogenetic acid-tolerant strains. Hanging-drop method can be used to determine microbial viability. Polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) can further analyze the evolution of microbial community and high-throughput sequencing technology can evaluate the microbial community [14]. The synergism of filamentous fungi, yeasts, and bacteria largely determines the successful liquor fermentation. There are a large number of functional microbes in *Huangshui*, mainly bacteria, among which lactic acid bacteria and *Bacillus* have been proven to be dominant bacteria (Table 2).

The high acidity, viscosity, and organic matter content of *Huangshui* make it difficult for ordinary microbes to survive in it. Therefore, functional microorganisms screened from *Huangshui* naturally adapt to extreme environments and have better tolerance compared to general strains under high acid and salt conditions. As depicted in Table 2, there are a variety of microbes in *Huangshui* that have high application value. Du et al. [33] found that lactic acid bacteria could be eliminated. Zhao et al. [24] used lactic acid bacteria to convert lactic acid into ethanol. Yang et al. [34] observed that the co-culture of *Bacillus licheniformis* A5 and *Bacillus subtilis* B2, increased the cellulase activity by 30–70%, which could provide a new microbial source for the production of cellulases. Daqu is a saccharifying agent and starter in the production of Chinese Baijiu, which plays a vital role in the fermentation of Baijiu and is one of the factors that ultimately determine the flavor quality of Baijiu. The fermentation ability of Baijiu starter mainly depends on the microbes, and many functional probiotics in *Huangshui*, such as yeast and aroma-producing bacteria (e.g., *clostridium*), are not only the main microbes for fermentation and production of liquor but also the main undertaker of liquor flavor substances. It is difficult to obtain them through long-term cultivation. Therefore, *Huangshui* can be used as a source of bacteria to add aroma and ester-producing microbes to Daqu to improve the quality of Daqu. Kang et al. [4] confirmed that *Lactobacillus* could produce and use non-volatile compounds, containing amino acids, sugar alcohols, and biogenic amines, while volatile compounds in *Huangshui* mainly evolved from dominant fungal genera. Interestingly, the *Huangshui*-adapted yeasts respond to some stress conditions, such as pH, nutrient availability, oxidative stress, and microbial interaction, while both isolation and identification of functional microbes in *Huangshui* still face enormous challenges [4].

Table 2. Screening and applications of functional microbes from *Huangshui*.

Bacterial Strain	Results and Applications	References
Bacillus subtilis, *Candida utilis*	They could use to ferment distillers' grains for feed production, raising the total amino acid by 17.74%.	[2]
Lactic acid bacteria	The bacteria could use starch and reducing sugars in *Huangshui* to produce lactic acid.	[4]
Acid tolerant yeast Strain	The strain showed high acetic acid tolerance and grew normally at 12 g/L acetic acid.	[13]
Lactic acid bacteria	They could convert lactic acid into ethanol.	[24]
Lactobacillus	The Lactobacillus screened from *Huangshui* could improve flavor and eliminate ethyl carbamate.	[30]
Lactic acid bacteria	They could competitively degrade arginine with yeasts cooperation.	[33]

3.2. Baijiu Blending

Huangshui has various functional microbes, organic acids, carbohydrates, nitrogen-containing matters, alcohols, and flavor substances (e.g., ethyl acetate and ethyl butanoate) [5,10]. In addition to microbial use, *Huangshui* can also be utilized in a variety of uses, which is acted as a raw material for secondary fermentation, extraction of proteins, soluble starches, organic acids, polysaccharides, tannins, and pigments [9,35]. *Huangshui* is simply treated, which can be utilized to blend Baijiu, produce calcium lactate, extract flavor components, culture the artificial pit mud, make solid and liquid feed, prepare vinegar and polysaccharides, and preserve food [10,18]. *Huangshui*, as the main by-product of Baijiu brewing, contains rich aromatic substances and nutrients [3]. If the substances continue to be esterified and blended into Baijiu, it can not only increase the flavor of the original liquor body but also give the liquor body a soft and harmonious taste [36]. For example, water-absorbing resin was used to concentrate *Huangshui* for extracting flavor substances, such as acetaldehyde, ethyl formate, and others, which could be further mixed to blend ordinary Baijiu into superior full-flavored Baijiu [37,38]. Recently, a large amount

of study has focused on how to optimize process parameters, promote the esterification reaction of abundant alcohol substances and organic acids in *Huangshui*, and produce high-quality liquor with rich ester content. Xia et al. screened *Monascus purpureus* SICC 3.19 and made it an esterifying agent to be added to *Huangshui* [37]. The reaction was carried out under optimal process conditions, and it was found that the content of the total ester was increased by 1.5 times [37]. The filamentous fungus Rhizopus chinensis can produce whole-cell lipases and exhibits strong tolerance under acid and alcohol stress, making it a promising catalyst in ester synthesis [39,40]. The esterase produced by the *Monascus* genus is pivotal in the ester synthesis process, thereby affecting the formation of Baijiu aroma [41]. Esterification could be achieved by microbes via several bio-reactions [42]. The concentration of esters in *Huangshui* that promote the aroma formation of Baijiu is relatively low, making *Huangshui* have other odors causing uncoordinated aroma. The content of esters in *Huangshui* needs to be improved by the esterification reaction. Organic acids and alcohols in *Huangshui* are used as substrates to produce major aroma and flavor substances in Baijiu such as esters by modern microbiological technology and fermentation engineering process. Presently, there are two commonly employed esterification techniques, including microbial enzyme method and artificial synthesis. The microbial enzyme method is widely used in the production of esterification solution with *Huangshui* due to its mild reaction conditions, fast reaction rate, high conversion rate, and better product purity [43]. In addition to enzymatic esterification reactions, the addition of *Monascus* enzyme- and ester-producing yeast to *Huangshui* substrate is also a widely used esterification method. Some of the enzymes from Baijiu microbes can catalyze ester synthesis in the aqueous phase, being different from classical enzyme-catalyzed esterification [44]. Xia et al. [37] screened a strain of *Monascus purpureus* and added it into *Huangshui* as an esterification agent for producing an esterification solution. They found that the total ester concentration increased from 4.86 to 7.31 g/L under the optimal conditions (10% alcohol and pH 3.5) after esterification for 16 days [37]. Moreover, *Huangshui* is enriched with aromatic substances, which can be used to produce biological functional blending liquid or other essence to improve Baijiu flavor and quality [10]. The supercritical CO_2 fluid extraction potentially promotes esterification reactions to form aromatic compounds such as ethyl butyrate and ethyl hexanoate [45]. Excess ethanol and inorganic salts in esterification solution can form a biphasic aqueous system for ester synthesis for blending acidified-base liquor, optimizing liquor flavor [46]. Furthermore, the *Huangshui* after esterification improved the rate of Baijiu's excellent and first grade by string evaporation. However, many reports on *Huangshui* recycling for the utilization of flavor compounds are non-quantitative and random.

3.3. Organic Acids

Huangshui contains abundant sugars, proteins, organic acids, and various aromatic components of alcohol esters along with high COD_{Cr}, which can increase the load of subsequent biochemical treatment. It is important to know how to convert the high-content components in *Huangshui* via microbes to high-valuable products and apply them to the food industry. This favors the comprehensive utilization value of *Huangshui*, which achieves zero emissions of brewing by-products. Such high organic wastewater can also serve as a starter culture for yeast and microbial cultures for beverage, condiment, and biomass energy production [10,47]. Organic acids are a type of functional compounds enriched with various biological activities and health-protecting properties, largely due to their carboxyl groups [48]. The natural antioxidant qualities in organic acids facilitate them to eliminate free radicals and repair cellular injury caused by oxidative stress [49]. Organic acids can be extracted by concentration and crystallization methods [10]. At present, the methods of extracting organic acids include the calcium salt method, ion exchange resin adsorption, bipolar membrane separation, distillation, solvent extraction, supercritical CO_2 extraction, and others. The lactic acid and reducing sugars in *Huangshui* are used as substrates by propionic acid bacteria to produce propionic acid. Meanwhile, propionic acid has different industrial uses in mold inhibition, fruit flavoring, grain, and feed preservation,

as perfume bases in cosmetics, in conjunctivitis, and in anti-arthritic drug production of the pharmaceutical industry [50]. In addition, most organic acids have anti-microbial activity, in which the minimum inhibitory dosages of acetic acid and lactic acid for *Shigella* species are 200 mg/L and 5000 mg/L, respectively [51]. The relatively high contents of organic acids in *Huangshui* partially reveal the antimicrobial phenomenon. Organic acids are also utilized in cosmetic production such as exfoliants and skincare products [10]. The use of organic acids extends to the food industry, in which they act as acidity regulators, improving the texture and taste of foods. They favor extending the shelf life of food via inhibiting microbial growth and avoiding food spoilage [10]. Organic acids and their salts from some fruits had been applied in meat preservation. Butyrate plays a vital role in gastrointestinal health, while its concentration affects microbial and host functions [10]. In addition, organic acid salts (e.g., calcium L-lactate) are used as a calcium source with better bioavailability and solubility compared to other organic salts, intervening in pharmaceutics to cure calcium deficiency [52].

Biotechnological and chemical techniques necessitate increased energy consumption and chemical usage, yet enable a comprehensive retrieval of valuable constituents [53]. Despite many studies on organic acids in different industries, their comprehensive uses in *Huangshui* are limited. The organic acids include formic acid, acetic acid, hexanoic acid, lactic acid, oxalic acid, citric acid, and succinic acid in *Huangshui* [7]. Nonetheless, their overall development and utilization in *Huangshui* have still been unexplored. Revealing the full potential of organic acids from *Huangshui* can obtain environmental and economic benefits, lower operational costs, and favor sustainable development.

3.4. Polysaccharides

Polysaccharides are an important biological macromolecule, which have different bio-activities and health-improving effects [54]. Polysaccharides can be extracted by water-ethanol extraction and purified by column chromatography. The extracted polysaccharides can be purified and crystallized to obtain high-purity polysaccharides [45]. It contains a higher proportion of glucuronic acid, galacturonic acid, arabinose, and galactose, which will lead to better antioxidant activity. The lower molecular mass and purity of *Huangshui* polysaccharides as well as their high protein content will also improve their antioxidant properties. Polysaccharides act as antioxidants, which help to neutralize free radicals, lower oxidative stress, and protect cells from oxidative damage [55]. Polysaccharides improve immunity against infection and exhibit antitumor activity [10]. The α-D-glucan is isolated from *Huangshui* and has antioxidant and immunoregulation activity [45]. Heteropolysaccharides with relatively complex structures have been isolated as well. They act as a mediator to stimulate THP-1 cells and upregulate messenger ribonucleic acid, which in turn enhances cellular engulfment and phagocytosis. Yang et al. [56] depicted that the healthcare function of polysaccharides can extend to relevant use fields in feed, food, medicine, and cosmetics as well as industrial products. Polysaccharides not only help diabetes patients stabilize blood sugar levels but also favor probiotic bacterial growth, which promotes intestinal health [57]. Polysaccharides are used to produce medicines and health products for treating some diseases and boosting physical immunity [10]. Considering the functional effects of polysaccharides, they act as food additives, including gelling agents, thickeners, and stabilizers [58]. Using the multifunctional polysaccharides from food by-products can decrease resource waste and provide potential development opportunities for the related areas. The various utilizations of polysaccharides highlight their potential value in many fields. Such approaches comply with low carbon emissions, the environment, and a circular economy.

3.5. Proteins and Amino Acids

Huangshui comes from microbial metabolism and fermented cereals, containing generous nutrients, for instance, organic acids (12.5–737.58 mg/100 mL), sugars (11.01 mg/mL), proteins (0.08–0.27% of the total *Huangshui*), amino acids (5.11 mg/mL) and a variety

of microorganisms [3,9]. Proteins are critical biomolecules in organisms with several functions, including structural support of cells, tissues, and organs, catalyzing chemical reactions, meanwhile delivering various substances to living organisms, which play a vital role in the immune mechanism. Proteins can be extracted under acidic and alkaline conditions or obtained by wet extraction and purified by precipitation, filtration, and crystallization [7,10,59]. Moreover, proteins are capable of producing bio-active peptides with antibacterial, antioxidant, and anti-inflammatory properties [10]. The antioxidant effects of proteins have been shown to be associated with their amino acids [60]. Proteins in *Huangshui* can be applied in many areas, including food, feed, beverages, and industrial products [59]. It is usually used by liquor factories for cellar raising, cultivating artificial cellar mud, and mixing back into the cellar for fermentation [18]. The peptides from *Huangshui* exhibit the potential to lower inflammatory responses and blood pressure, which helps to alleviate some symptoms related to inflammatory and high blood pressure diseases [10,59]. Nonetheless, the research on proteins in *Huangshui* is limited, while its potential utilization still needs to be explored in the future. In addition, amino acids are the smallest basic unit of the protein molecule, one of the nutrients necessary for life activities, playing a central role as components of proteins and metabolic intermediates [4]. It can be obtained by enzymatic digestion of residual proteins and further purified by chromatographic separation [45]. *Huangshui* contains many free amino acids, such as alanine, glutamic acid, and isoleucine. Meanwhile, amino acids, as essential substances can enter the human body as food or medicine, thus acting as an important part of the human body. Alanine can protect and improve liver function, serine can lower blood cholesterol, and isoleucine can repair muscles and control blood sugar levels [61]. Amino acids, protein decomposition products, and nitrogenous compounds produced by bacterial autolysis in *Huangshui* provide a nice material basis for microbial growth and metabolism.

3.6. Aldehydes, Flavonoids, Ketones and Phenols

In addition to esters, organic acids, and polysaccharides, *Huangshui* contains other organic compounds, including aldehydes, flavonoids, ketones, and phenols. They affiliate with secondary metabolites, which are often observed in natural plant compounds, having different biological activities [10]. Solvent, Soxhlet, and extraction techniques such as ultrasound-assisted and subcritical water extraction were used to obtain the substances. Thin-layer chromatography or high-performance liquid chromatography allowed their separation and purification [10]. The compounds possess excellent antioxidant capacity, eliminating free radicals, and thus lowering oxidative stress [10]. Moreover, the organic compounds can also mitigate inflammatory reactions by controlling associated pathways and maintaining anti-inflammatory mediators [62]. Considering flavonoids, they not only have anti-inflammatory, anti-hypertensive, heat-clearing and detoxifying, anti-oxidant, and diuretic effects but also possess significant effects in anticancer and lipase inhibition. Sasaki et al. [28] reported that flavonoids exhibited anticancer potential by inhibiting tumor growth and metastasis. Aldehydes are important organics, whose unique structure and properties make them widely used in some fields such as chemical synthesis and biology. Ali et al. [63] pointed out that aldehydes could lower the interaction between protein and glucose, which could avoid diabetic complications. There are many potential benefits in the aforementioned organic compounds, but there are still limited reports on their deep development and use in the pharmaceutical industry. Phenolic ketones show antibacterial effects against fungi and bacteria, which enable them to be utilized in pharmaceutical production and food preservation [10]. Polyphenols are capable of regulating gut microbiota, autophagy, inflammation, and oxidative damage [64]. In addition, polyphenols and phenolic ketones have anti-aging and lifespan extension properties, which can be used in health products [10,64]. Hu et al. [65] investigated that quercetin could serve as a bio-based anti-aging agent, and revealed that quercetin with a high dosage of phenolic hydroxyls showed excellent anti-aging properties.

Nonetheless, the positive effects of the organic compounds from *Huangshui* have been investigated and applied to the food, pharmaceutical, medicine, health products, and cosmetics industries. However, the availability of *Huangshui* is low due to the microbial diversity and its metabolic complexity in *Huangshui*. Furthermore, the complete extraction of useful components can be obtained from *Huangshui* with some biotechnological and/or chemical processes, likely requiring more chemicals and energy, causing secondary pollution and wasting water resources. Compared with the process of using dried DGS directly as livestock feed, it is more difficult to treat *Huangshui* and obtain its full application. Therefore, it is urgent to seek a simple, low-cost, near-zero-emission process for producing a series of bulk products using *Huangshui* as raw material.

3.7. Full Utilization of Huangshui in Different Fields

The high concentration of organics of *Huangshui* with some microorganisms can be used as a substrate or inoculum to treat certain types of organic wastewater for organic pollutant degradation and biofuel production. Wang et al. [66] utilized the DSG containing *Huangshui* to co-produce biobutanol and bioCH_4, having relatively high costs. Ling et al. [11] observed that the antimicrobial activity of *Huangshui* could promote the oleaginous yeast to become the dominant microbes over undesirable microbes for treating mixed wastewater to produce lipids. The undistilled alcohol in *Huangshui* by fermentation and distillation can be extracted to produce bioethanol for use as biofuel, but the residual wastewater still needs to be treated [10]. In addition, for most wastewater treatment plants (WWTPs), conventional treatment processes cannot solve the technical problems of the imbalanced carbon-to-nitrogen (C/N) ratio and low-carbon source in wastewater. WWTPs usually choose organic compounds such as acetic acid, ethanol, and glucose as external carbon sources, with an average market price of around USD 350 per ton. External carbon sources, as substrates for biochemical reactions and microbial growth, affect the structure and abundance of microbial populations, and different carbon sources can selectively enrich different high-efficiency denitrifying bacteria [67]. The anammox process reveals the prospect of obtaining energy balance and even electricity generation in WWTPs [67]. When certain organic wastewater is high-concentration wastewater, its key parameters of carbon source utilization, including B/C $\geq$ 0.5, C/N $\geq$ 6, and COD $\geq$ 100,000 mg/L, can be used as an effective alternative carbon source for feeding into WWTPs. Interestingly, *Huangshui* fully meets the indicators of such wastewater and can be directly used as an external carbon source for the denitrification system of WWTPs (Figure 5). This method has significant economic and environmental benefits compared to other technologies that utilize *Huangshui* to produce H_2 and CH_4 with fermentation.

In addition, the organic matter in *Huangshui* can improve soil quality, providing nutrients for plants [7]. After appropriate treatment of *Huangshui*, it can be served as a soil conditioner or organic fertilizer in agriculture [10]. If *Huangshui* is used to irrigate or improve soil without proper treatment, its high concentration of organic matter will cause soil contamination, influencing soil quality and vegetation health [68]. Halvorson and Gonzalez [69] concluded that the contents of soluble soil carbon and nitrogen in soil significantly decreased after being mixed with tannin solution, implying that tannin could damage the two elements of carbon and nitrogen conversion. Some calcium salts of organic acids, including Ca-acetate, Ca-citrate, and Ca-lactate, can be formed when $CaCO_3$ or $Ca(OH)_2$/CaO is added to *Huangshui*. These solubilities of Ca-acetate, Ca-citrate, and Ca-lactate are 300 g/L, 0.85 g/L, and 79 g/L, respectively [70]. The prepared calcium lactate can meet the market demand for lactic acid in the pharmaceutical, chemical, and food industries. Moreover, the solubility of Ca-amino acid is 2.24 g/L. Interestingly, these Ca-containing compounds that exist in liquid or solid forms can be biologically effectively utilized, which lays a good foundation for the cascade and full utilization of *Huangshui*. The Ca-containing matter can be used as feed additives to improve symptoms of calcium deficiency in animals. The liquid organic fertilizer with the above-mentioned organic acid calcium, such as Ca-lactate and Ca-amino acid is sprayed onto crops, increasing crop

quality and yield, and enhancing their stress resistance. Prospectively, the component extraction methods can be used before fertilization and fermentation to enable cascade and full application of *Huangshui* subsequently (Figure 5). The extraction process parameters for various target ingredients should be optimized by low costs and eco-friendly technology. Furthermore, the extracted ingredients and residues can be used in other products to enable the *Huangshui* cascade and full utilization.

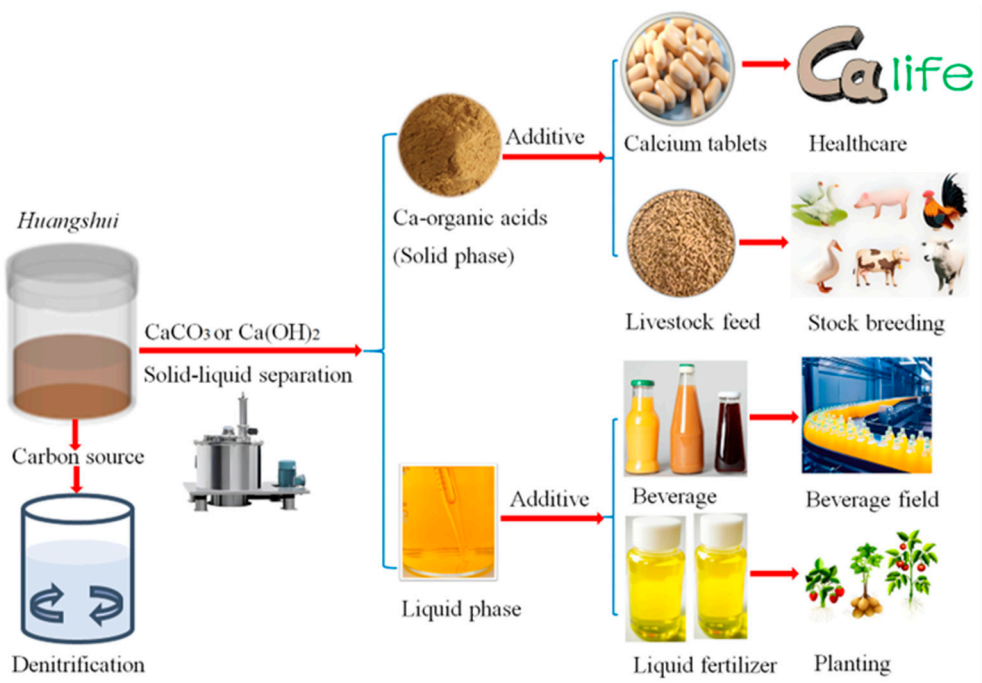


Figure 5. Processes for cascade and complete utilization of *Huangshui*.

4. Future Challenges and Prospects

4.1. Challenges

Recently, with the continuous growth of gross domestic product, the Baijiu (liquor) industry has visibly expanded. The quantity of wastewater generated from Baijiu production significantly increases. Conventional methods of managing and treating DSG and *Huangshui* likely cause the risk of soil and air pollution [7] and potential harm to agriculture and human health [71,72]. The comprehensive and full utilization of *Huangshui* is helpful for lightening environmental burdens and reducing wastewater treatment costs [2]. Although high-value separation can expand the industrial chain and obtain the value of derivative products, the treatment and use efficiency are limited. *Huangshui* contains complex ingredients, some of which are likely difficult to separate, and need complex processes, thereby causing many difficulties in practical application. The separation of some beneficial ingredients from *Huangshui* involves high costs, such as investment and operational costs, which will influence economic viability. Secondary pollution and carbon emissions also should be highlighted in beneficial ingredient separation. On the other hand, some reuse techniques lack stable and mature processes, resulting in technical difficulties, low operational stability, and quality control challenges [10]. Considering the challenges and difficulties, evaluation and surveys are required when *Huangshui* is reused, which can favor resolving technical and market barriers and enabling low carbon, complete application, and sustainable management of reuse.

Huangshui has many reuse values, including microbial screening, secondary brewing, biofuel, and fermentation conversion, their application potential can be improved by isolating bioactive components and utilizing them in the functional food field [10]. As described in Table 1, *Huangshui* contains beneficial matters of proteins, peptides, polysaccharides, organic acids, aldehydes, and phenols [7], being effectively extracted from *Huangshui*, and

applied in beverage, healthcare, and medicine fields (Figures 2 and 5). The beneficial ingredients in *Huangshui* can be obtained by simple chemical reactions and physical separation as well as processing processes, in which chemical reagents such as $CaCO_3$, CaO, and $Ca(OH)_2$ can be food and pharmaceutical grades for subsequent different application areas (Figure 5). These reused ingredients can be applied in food-processing fields (e.g., beverages, condiments, and food additives), healthcare (e.g., calcium lactate and calcium amino acid chelate), cosmetics (skincare), animal husbandry (feed additives), agriculture (fertilizers, brewing yeast), and medicine production [10].

4.2. Prospects

Promoting industrial technological innovation and developing new productive forces are inevitable requirements for China to promote high-quality economic development and accelerate modernization construction. The increasing consumer demand for innovation, sustainability, and health is encouraging enterprises to develop the integration of *Huangshui* to meet market demands. The prospect of full utilization of *Huangshui* will have a promising future, which is helpful for achieving near-zero pollutant emissions and maximizing congregational resources, further meeting market requirements in different fields [7,10,73]. In addition, it is essential to advance the extraction and utilization of the components in *Huangshui*. The methods employed in extraction should be inexpensive and environmentally safe. For example, simple physical methods such as membrane separation can separate microorganisms from *Huangshui* [74]. It can also separate components in *Huangshui* according to the size of molecular mass, which can be targeted for the utilization of *Huangshui* at a larger stage (Figure 5). Membrane separation equipment is cost-effective and has strong continuous mechanized operation. The extracted target components are used as an additional carbon source for denitrification systems or as organic fertilizer to improve soil quality. Therefore, the combination of a simpler, more efficient, and cleaner treatment process with modern separation and extraction technology is one of the future development directions of *Huangshui* resource utilization. Moreover, microbes have strong adaptability, decomposition, and metabolism advantages, making them suitable for application in *Huangshui* treatment. Using abundant organic substances to synthesize products with high value can obtain the material virtuous cycle in the Baijiu industry [11]. Through innovating food products, optimizing fertilization structure, and paying attention to healthy foods and integrated water and fertilizer, along with eco-friendly management and reutilization strategies, the full utilization of *Huangshui* can be achieved, with its maximum value, ultimately impelling the sustainable developments and prosperities of the food industry and agriculture fields [10,66]. With the intensification of energy and food crises and water shortages, as well as the increasing demand for environmental protection, the full utilization of high-strength organic wastewater can alleviate the above difficulties.

5. Conclusions

This review explores the main physicochemical properties of *Huangshui* and its potential applications in some related fields like food and beverage, healthcare, wastewater treatment, and agriculture. The significance of the efficient utilization of beneficial ingredients, including polysaccharides, organic acids, phenols, ketones, amino acids, and peptides in *Huangshui* is highlighted. Specifically, the low-carbon, complete, and sustainable mode of reusing *Huangshui* will be the focus of future research, such as the denitrification of carbon sources, and water and fertilizer integration. Meanwhile, the effect of product application should require a long-term monitory.

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