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System Analysis of Bio-Natural Gas from Large-Scale High Organic Load Wastewater in China

Chenxuan Wang[a], Xiaoping Jia[a], Zhiwei Li[b], Raymond R. Tan[c], Fang Wang[d,*]

[a]School of Environment and Safety Engineering, Qingdao University of Science and Technology, Qingdao 266042, China
[b]School of Chemistry and Chemical Engineering, Hefei University of Technology, Hefei 230009, China
[c]Department of Chemical Engineering, De La Salle University, Manila 0922, Philippines
[d]Sino-German Engineering College, Qingdao University of Science and Technology, Qingdao 266061, China
wangf@qust.edu.cn

The demand for renewable alternatives to natural gas as a fuel for industries and residential consumers has been growing fast. Bio-natural gas is expected to play an important role in green energy transition and climate change. Large amounts of high organic load wastewater are discharged from coking, pharmaceutical, papermaking, printing and dyeing, petrochemical, and food processing industries. Bio-natural gas could be produced from high organic load wastewater using anaerobic digestion or membrane bio-reactor and biogas upgrading technologies. This study performs a comprehensive analysis of bio-natural gas produced from high organic load wastewater (BNGWW) in China. The status quo is compared with the potential use of this alternative, coupled with a techno-economic analysis of production technologies, energy policies, and opportunities and challenges. A model is proposed to estimate the provincial BNGWW demand in China from 2016 to 2020 and the decomposed demand across 42 sectors from 2016 to 2019. Factors influencing BNGWW potential are analyzed under the changes of time, region, and sectors. The result shows that BNGWW demand is affected mainly by natural gas prices, industrial scale, and chemical oxygen demand of organic wastewater. Some provinces in East China, Central and Southern China, and Xinjiang have high BNGWW potential. The agricultural and food processing industry has the highest production potential. Challenges and barriers to the development of BNGWW have also been highlighted.

1. Introduction

At present, China's energy structure is dominated by non-renewable energy, such as coal, oil, and natural gas. Petroleum and natural gas resources are scarce, mainly dependent on imports. As the world consumes more petroleum, its reserves are dwindling, and prices are soaring, China urgently needs to find alternative energy sources (Zeng et al., 2021). The coal-dominated energy structure has added to environmental pressures in China. China declared that it would achieve a "carbon peak" by 2030 and that it aims to be carbon neutral by 2060. Under the pressure of resource and emissions limits, it is urgent to realize the low-carbon transformation of the energy system.

Bioenergy is derived from biomass, which in turn comes from solar energy through photosynthesis. Biomass can improve the efficiency of energy use and economic benefits, reduce carbon emissions, and pressure on the environment. There are a variety of treatment methods to convert biomass, and anaerobic digestion has already been implemented in the industry, representing the most mature biogas production technology. Biomass in China is an abundant energy resource. In particular, there is huge potential for biogas, with a theoretical annual output of 73.6×10^9 m^3 (Xue et al., 2020). Bio-natural gas (BNG) is a renewable natural gas substitute mainly containing methane produced by the purification or methanation process. Generally, the concentration of methane in biogas after anaerobic digestion is about 50 % - 70 %. After CO_2 and impurities are removed by the purification process, BNG with a methane content of no less than 90 % can be obtained. BNG is the same as conventional natural gas in composition and caloric value and can be used as a direct substitute using the same distribution and end-use technologies. It is a green and low-carbon clean energy, which can replace fossil energy for cogeneration, heating, and vehicle fuel. Zhao (2020) classifies and

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quantifies the potential of BNG production from urban organic waste, and takes Changshu City as a case to analyze the potential of BNG production from organic waste and the scheme of purification and grid connection. Liu et al. (2023) estimated and analyzed agricultural waste and biogas potential in Hubei province from two aspects of crop yield and animal husbandry output by geographical methods. Liu et al. (2016) estimated the theoretical and practical biomethane potential in China and calculated that the total efficiency of the existing anaerobic fermentation process was 35.59 %. Mittal et al. (2019) studied the biogas potential of biomass energy and predicted the gas production potential of different raw materials in 2040.

Industrial wastewater is the main source of water pollution in China. Compared with the stable characteristics of domestic wastewater pollutants, industrial wastewater has a wide variation in composition depending on the source. If discharged into the environment without treatment, industrial wastewater will cause serious environmental pollution, which is the focus of water pollution control. High organic load wastewater discharged mainly from coking, pharmaceutical, papermaking, printing and dyeing, petrochemical, and food processing industries, can be used as a raw material for BNG production. Biogas can be produced from organic wastewater containing high concentrations of toxic and harmful pollutants by anaerobic digestion technology, and BNG was obtained by biogas upgrading technology which can remove the CO_2. This technology has a high utilization rate, is relatively mature, and can be integrated with wastewater treatment. It can simultaneously address environmental and energy resource concerns (Zhang, 2018). Tan et al. (2018) indicated that the electricity from POME biogas is expected to save up to 9.46 % of the total energy supplied for palm oil mill processes.

There are few papers on the energy potential of BNG from industrial organic wastewater (BNGWW). An estimation calculation model of BNGWW was proposed to calculate the provincial-wide BNGWW in China from 2012 to 2019 and the BNGWW of 42 industries from 2016 to 2019 based on guidelines of the IPCC (2006). Based on the geographic information system (GIS) spatial analysis method, the spatial and temporal distribution characteristics of BNGWW theoretical potential in China from 2012 to 2019 were studied. Under the changes of time, region, and sectors, the BNGWW potential and its influencing factors were analyzed.

2. Method

2.1 Study area and data sources

The geographic scope of this paper is China, with 23 provinces (without Taiwan), 5 autonomous regions and 4 municipalities. According to the physical geographical division of China, the 31 provinces are divided into seven regions: North China, Northeast China, East China, Central China, South China, Southwest China and Northwest China.

The data of chemical oxygen demand (COD) discharge of industrial wastewater come from the Environmental Statistics Data (National Bureau of Statistics, PCR, 2012-2014), the China Statistical Yearbook (National Bureau of Statistics, PCR, 2012-2022), and the China environmental protection database (China Environmental Protection Database, 2023).

2.2 Estimated calculation model of BNGWW

The calculation formula of BNGWW (IPCC, 2006) is as follows:

$$\mathrm{M_{COD}} = \mathrm{T_{COD}} * \varepsilon \tag{1}$$

$$\mathrm{M_{CH_4}} = (\mathrm{M_{COD}} * \mathrm{MCF} * \beta) - \mathrm{R} \tag{2}$$

$$\mathrm{V_{BNG}} = \frac{\mathrm{M_{CH_4}}}{\rho_{\mathrm{CH_4}} * \gamma} \tag{3}$$

where M_{COD} is COD removal (t); T_{COD} is COD emissions (t); ε is COD removal efficiency (97 %); M_{CH_4} is methane production (t); MCF is methane conversion factor, refer to the IPCC guidelines for the anaerobic reactor value of 85 %; β is the methane production rate, 0.25 kg_{CH_4}/kg_{COD} is taken for industrial wastewater treatment; R is the methane recovery amount, which is 0 for anaerobic reactor without considering methane recovery; V_{BNG} is the total potential of BNGWW (Mm^3); ρ_{CH_4} is the methane density of 0.67 m^3/kg at 20 °C standard atmospheric pressure; γ is the methane content in BNGWW.

In 2021, there were 68,150 sets of wastewater treatment facilities in China's industrial enterprises, with a COD removal rate of 97.3 % based on 2021 data. In order to enable BNGWW to meet the requirements of incorporation into natural gas pipeline network, the methane content in BNGWW is calculated according to the requirements of Classification and Basic Characteristics of Urban Gas (GB/T 13611-2018) (MHURD, PRC, 2018). Most types of natural gas in China are 12T in GB/T 13611-2018. Under the assumption, the biological

natural gas only contains CH_4 and CO_2. In order to meet the requirements of high calorific value and high white number of 12T natural gas in GB/T 13611-2018 at the same time, the content of CH_{co} should be greater than 94.36 % (Zhao, 2020), and the methane content γ in the above formula is 95 %.

3. Result

3.1 Total potential of BNGWW in China

The national BNGWW potential is proportional with the COD discharge of industrial wastewater. From 2012 to 2021, with the COD discharge of industrial wastewater decreased every year, the national potential of BNGWW decreased from 1096.05 Mm^3 in 2012 to 136.99 Mm^3 in 2021, fell by 87.5 % (Figure 1). The highest reduction rates were recorded in 2016 and 2020, with a decrease of 58.14 % and 35.55 %.

In 2015, the Chinese government issued a series of water pollution prevention and control documents, including the Action Plan for Prevention and Control of Water Pollution (MEE, PCR, 2015). In order to strengthen the prevention and control of water pollution and ensure national water security, to improve water pollution in key industries in China. The "Water Ten" policy has significantly reduced the intensity of water pollution, improved the urban industrial structure, increased the proportion of the tertiary industry, promoted the development of low-energy and environment-friendly industries, and reduced the discharge of COD in wastewater (Lu et al.2021). According to the Ministry of Ecology and Environment of China, by June 2017, 1,762 enterprises in the six industries of paper making, steel, printing and dyeing, pharmaceutical, leather, and nitrogen fertilizer had completed the transformation of industrial pollution, with a completion rate of 84.6 %. In industrial clusters at the provincial level and above, 1,968 have built centralized sewage treatment facilities, and 1,746 have set up online monitoring devices, with completion rates of 80.6 % and 71.5 %. Due to the influence of water pollution control policies, COD discharge of industrial wastewater has decreased significantly. The national potential of BNGWW decreased to 432.75 Mm^3 in 2016, which was 601.55 Mm^3 and 58.2 % lower than 2015 with the highest potential change.

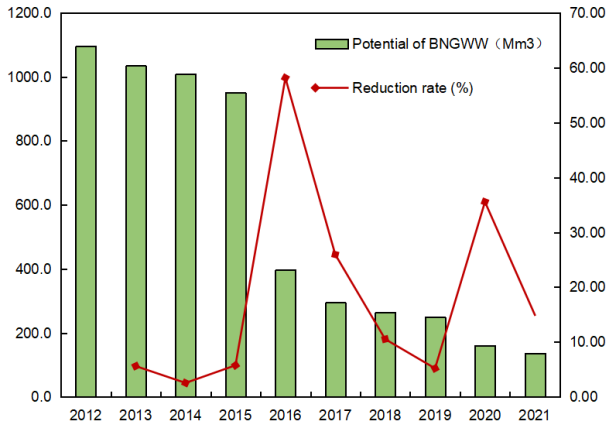

Figure 1: Total potential of BNGWW and reduction rate in China from 2012 to 2021

During the COVID-19 pandemic in 2020, most cities took quarantine measures, shutting down some transportation and businesses. During the quarantine period, major industries operated at much lower levels than normal. Wang and Su (2020) studied changes in industrial activity and energy consumption in some parts of China, showing that refinery operating rates in Shandong province, a major refining hub, fell to their lowest level since the fall of 2015. Industrial activities decreased significantly, COD emission decreased, and BNGWW gas production potential decreased by 35.6 % from 272.05 Mm^3 in 2019 to 175.26 Mm^3.

In 2012, the potential of BNGWW in East China was 304.25 Mm^3, accounting for 25.51 % of the total BNGWW potential in China, which was the highest among the seven regions in China (Table 1). The northeast region has the lowest potential of BNGWW, which is 97.52 Mm^3, accounting for 8.18 % of China. By 2019, the gas production potential of East China has decreased to 106.42 Mm^3, accounting for 40.5 % of the total. North China has become the region with the lowest gas production potential, accounting for 18.00 Mm^3, accounting for 6.85 of the national gas production potential. The BNGWW potential in North China decreased by 6.87 % in 8 y. East China has superior natural environmental conditions, rich natural resources and complete industrial categories. In 2021, there will be 208,175 industrial enterprises above designated size in East China,

accounting for 47.15 % of the country. It is a region with the highest comprehensive technology level and the most developed economy in China.

Table 1: Potential of BNGWW in different regions in 2012 and 2019

Regions	2012		2019	
	BNGWW（Mm^3）	Proportion（%）	BNGWW（Mm^3）	Proportion（%）
North China	141.68	11.88	18.00	6.85
Northeast China	97.52	8.18	22.13	8.43
East China	304.25	25.51	106.42	40.52
Central China	163.48	15.46	30.15	11.48
South China	158.82	13.32	33.09	12.6
Southwest China	141.95	11.9	29.73	11.32
Northwest China	185.03	15.51	49.43	8.81

3.2 Provincial-wide BNGWW in China from 2012 to 2020

The collected data covers 31 provinces in China, excluding Taiwan Province (Figure 2). From 2012 to 2014, the highest gas production was in Guangdong Province, with gas production of 11.24 Mm^3, 75.87 Mm^3, and 76.27 Mm^3, accounting for 7 % of the total potential of BNGWW in China. In 2015, it accounted for the lowest proportion, accounting for 5.48 % of the national potential, and ranked third in gas production. From 2016 to 2018, the proportion of annual gas production increased year by year. By 2019, the potential of BNGWW was 20.44 Mm^3, accounting for 8.18 %, ranking second in China. From 2015 to 2019, Jiangsu Province had the highest gas production potential in China. In 5 y, the gas production decreased from 65.19 Mm^3 to 28.43 Mm^3, a decrease of 56.39%, and the proportion increased from 6.86 % to 11.38 %. From 2012 to 2015, the potential of BNGWW in Xinjiang Province accounted for 5-6 % of total gas production, which decreased after 2016, while the potential in Shandong Province, which became the third largest province in China, increased after 2016. After 2016, the regions with high gas production potential are Jiangsu, Zhejiang, Jiangxi, Shandong, Hunan, and Guangdong. Gas production is mainly related to the industrial, economic, and technological development of the regions. The discharge of industrial wastewater is closely related to regional economic and technological development. The high discharge of industrial wastewater is mainly in some cities of Jiangsu, Zhejiang, Shandong, and Shanghai. The main reason is that the industrial structure is dominated by the textile industry and manufacturing industries, and the industrial level is high

In 9 y, Tibet has the smallest BNGWW potential, with annual gas production of 0.30 Mm^3 in 2012, accounting for only 0.03 % of the national total. In 2019, its annual gas production potential reached 0.42 Mm^3, accounting for 0.17 % of the national total. Low industrial wastewater discharge is distributed in the northwest, central and northeast regions, including some cities in Shaanxi, Gansu, Ningxia, Sichuan, Henan, Inner Mongolia, Liaoning, and Heilongjiang. The main reason is that industrial wastewater source industries are few, and the industrial level is relatively backward.

3.3 BNGWW potential of different industries from 2016 to 2019

Figure 3 shows the seven industries with the highest BNGWW potential from 2016 to 2019: Agricultural and sideline food processing industry, chemical raw materials and chemical products manufacturing industry, paper and paper products industry, textile industry, wine, beverage, and refined tea manufacturing industry, food manufacturing industry, and pharmaceutical manufacturing industry. The total potential of these seven industries in 4 y accounted for more than 70 % of the national gas production. The total amount of pollutants discharged from the wastewater of the agricultural and sideline food processing industry is large, and the content of toxic substances is less. Most of the wastewater with high COD concentration is the remaining raw materials, fats, starch, sugar, bacteria, and organic matter in the production process.

Over the course of 9 y, the BNG from the agricultural and non-staple food processing industry was the highest, with gas production of 81.38, 57.96, 50.51, and 51.38 Mm^3, accounting for about 20 % of the national average gas production. Chemical raw materials and chemicals manufacturing, textiles, paper, and paper products industry are similar, accounting for about 12 % of the country's total gas production in 4 y. The wastewater from these three industries is complex in composition and may also contain toxic pollutants, which can inhibit reactions during anaerobic digestion and reduce actual biogas production.

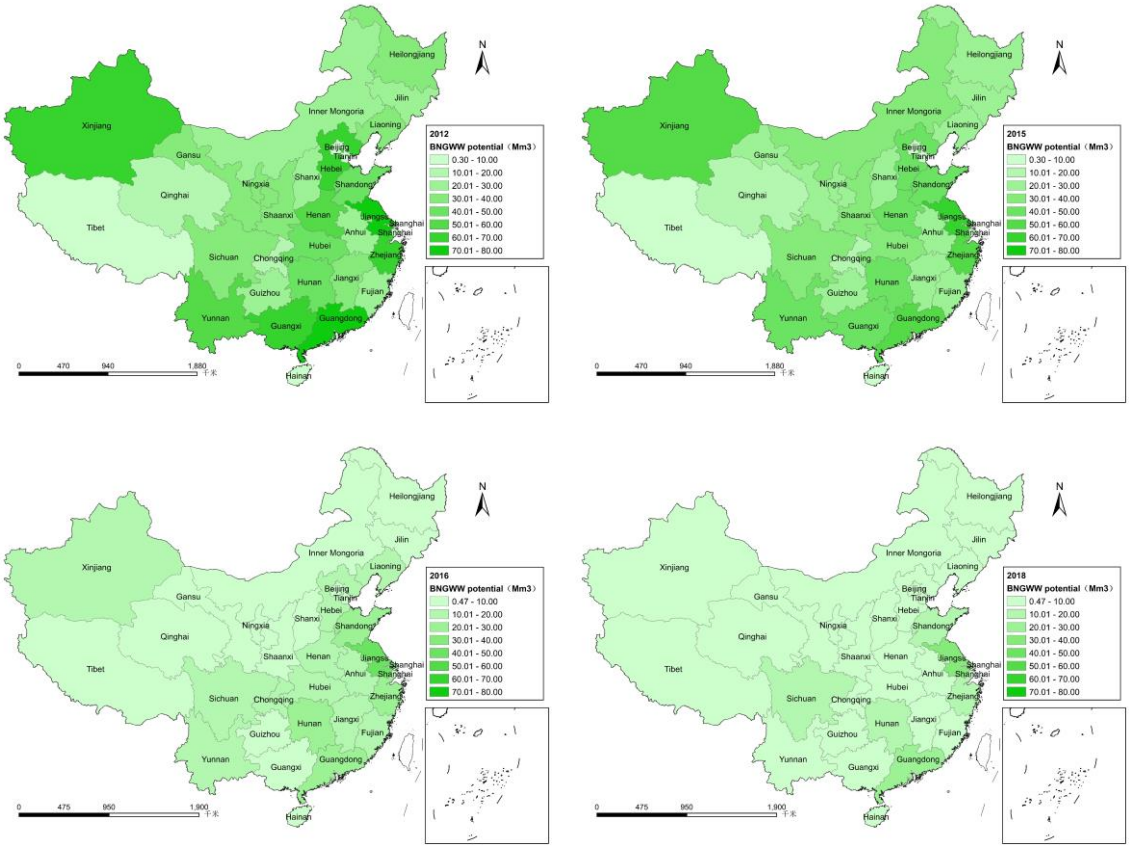

Figure 2: Provincial-wide BNGWW in China (2012, 2015, 2016, 2018)

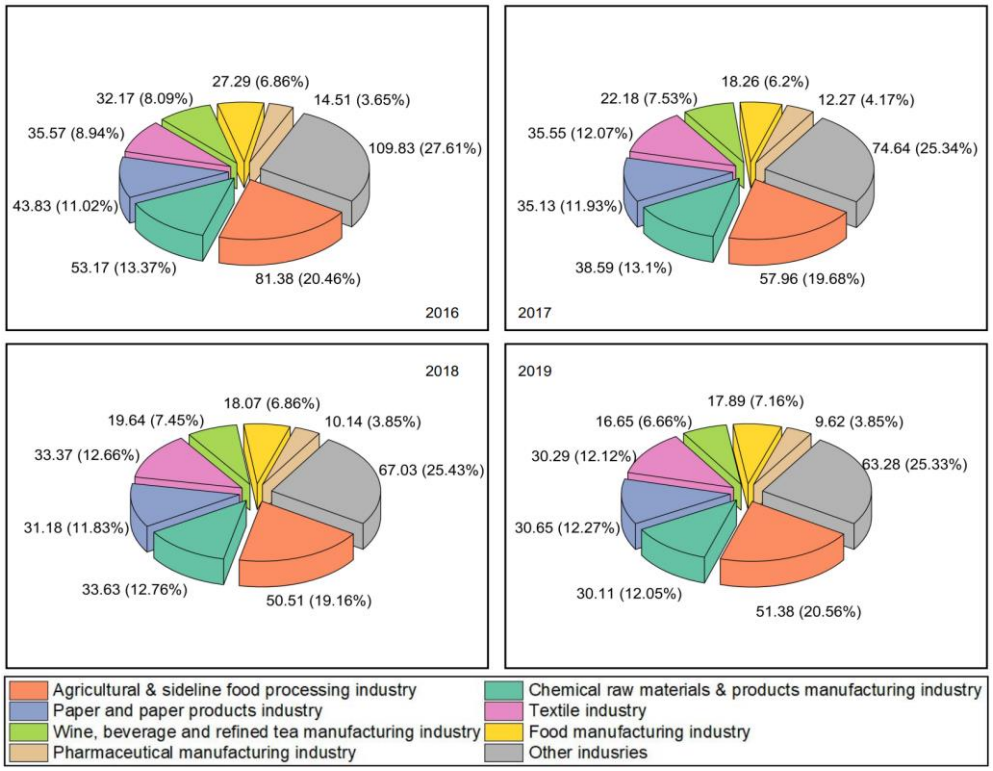

Figure 3: BNGWW potential of different industries from 2016 to 2019

4. Conclusions

Based on the IPCC guidelines, an estimated calculation model of BNGWW was proposed to calculate the provincial-wide BNGWW in China from 2016 to 2020 and the BNGWW of 42 sectors from 2016 to 2019. The spatial and temporal distribution characteristics of BNGWW's theoretical potential in China from 2012 to 2019 were studied by using the geographic information system (GIS) spatial analysis method, and the gas production potential of BNGWW and its influencing factors were analyzed. The BNGWW potential of China decreased year by year from 2012 to 2021 and decreased significantly in 2015 and 2020 due to the impact of policies and COVID-19. Regarding space, the provinces with the greatest gas production potential are Guangdong, Jiangsu, and other coastal areas. These areas have developed an industrial economy and produce more industrial organic wastewater, which can be used as key areas for BNGWW. The organic wastewater of the agricultural and sideline food industry is the industry with the highest BNGWW potential among 42 industries in China, accounting for about 20 % of the national potential. The biggest factors affecting the potential of BNGWW are COD discharge from industrial organic wastewater and different natural gas manufacturing paths. National policies and industrial activities also indirectly affect the BNGWW potential. Future work can focus on optimizing BNGWW production and distribution systems over an extended timeframe in the context of China's carbon neutrality target.

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