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Life cycle environmental and economic assessment of Phosphogypsum utilization in China

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ABSTRACT

Phosphogypsum (PG), a byproduct of the phosphate fertilizer industry, poses significant environmental challenges due to its huge production and complex composition. Efficient and large-scale utilization of PG remains a global issue. China, as the world's largest producer and discharger of PG, faces a relatively low utilization rate and urgently needs effective management strategies. This study evaluates the environmental and economic performance of the main PG treatment pathways in China using life cycle assessment (LCA). The results reveal notable benefits for all three utilization technologies, with sulfuric acid co-production cement (PSC) offering the greatest environmental advantages and β-building gypsum (PBG) providing the highest economic benefits. Scenario analysis suggests PSC as the most effective strategy for achieving dual environmental and economic gains, making it a strong candidate for large-scale implementation. These findings provide critical insights for future PG management strategies in China.

1. Introduction

Phosphogypsum (PG) is a byproduct generated during the wet processing of phosphoric acid production from phosphate rock (Baolin et al., 2022). It is estimated that 4.5–5.5 tons of PG are produced for per ton of phosphorus pentoxide produced (El-Didamony et al., 2012). PG has a complex, with more than 80 %−90 % consisting of $CaSO_4{\cdot}2H_2O$. However, due to the presence of impurities such as phosphorus, fluorine, organic matter, oxide and trace amount of heavy metals and radioactive substances (Attallah et al., 2019; Rutherford et al., 1996), along with its highly acidity, the comprehensive utilization of PG remains a global challenge. According to statistics, the annual world production of PG is about 200–250 million tons (Saadaoui et al., 2017). Over 85 % of PG is either deposited in large stockpiles or discarded into the sea, without any treatment, leading to severe environmental pollution and high managing costs (Cánovas et al., 2018; Jalali et al., 2019). Therefore, the comprehensive utilization and disposal of PG has become the key to the green, healthy and sustainable development of phosphorus chemical industry.

According to the *Phosphogypsum Leadership Innovation Partnership* report published by the International Fertilizer Association (IFA) in 2020, the primary pathways for the comprehensive utilization pathways of PG in major PG-producing countries are focus on three sectors: agriculture, construction, and roads (Hilton et al., 2020). Due to differences in national conditions, the emphasis on utilization varies. In China, the annual PG are approximately 80 million tons, and the cumulative stockpile has exceeded 870 million tons, accounting for 14.5 % of the global stockpile (Ma, 2019). With the continuous strengthening of environmental protection policies and the implementation of solid waste management measures, China's comprehensive utilization rate of PG significantly increased, reaching 45.6 % in 2021 (Cui, 2022).

However, the utilization of PG still mainly relies on basic utilization methods, such as being used as a cement retarder, in building materials, for road construction, or mine filling, currently (Cui et al., 2022). Due to factors such as limited regional market demand, low product added value, and restricted transportation radius, relying solely on PG-based building materials is insufficient to meet the demand for the consumption of newly produced and accumulated PG. Therefore, there is an urgent need to explore new technologies for the large-scale and high-value-added utilization of PG.

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In recent years, the academic community has been continuously exploring the novel valorization routes of PG (Agrawal et al., 2023; Cánovas et al., 2018). These include recovering minerals such as rare earth elements (Laurino et al., 2019; Virolainen et al., 2019), producing lithium sulfate and calcium sulfate (Hammas-Nasri et al., 2020), developing high-strength gypsum powder (Guan et al., 2022), employing it for CO_2 sequestration (Contreras et al., 2015), and incorporating it as fillers in papermaking (Mechi et al., 2017). However, most of these studies remain in the experimental phase or exhibit relatively low utilization rates. The PG-based sulfuric acid–Portland cement co-production technology adheres to circular economy principles by recycling the calcium and sulfur elements in PG, thereby establishing a self-circulating process within the phosphoric acid production cycle. This technology can be directly implemented within phosphoric acid production enterprises, enabling on-site resource utilization. The sulfuric acid produced is directly reused in the phosphoric acid production unit, reducing the need for externally purchased sulfuric acid and avoiding environmental risks associated with sulfuric acid transportation. The Portland cement produced can be supplied to the extensive cement market, reducing the demand for limestone quarrying and conserving valuable calcium resources (He et al., 2022; Qin et al., 2023). Against the backdrop of high demand for sulfuric acid, continuous increases in sulfur prices, and clear decarbonization targets in the cement industry, this technology offers distinct advantages and represents as an effective pathway for maximizing the utilization of PG resources.

Life cycle assessment (LCA) is a widely used method for comprehensively analyzing the environmental impacts of waste management strategies and technologies, providing critical technical support for promoting waste optimization management and developing a circular economy (Dong et al., 2024). In the context of PG recycling and utilization, previous studies have applied LCA to evaluate specific management methods. For instance, Kulczycka et al. (2016) assessed the environmental impacts of PG landfilling and sulfuric acid leaching for rare earth element recovery in Poland. Mohammed et al. (2018) evaluated the conversion of PG into paper and fertilizer. Tsioka and Voudrias (2020) compared the environmental impacts of PG management methods in brick production, soil amendment, road construction, and stockpile treatment, indicating that using PG in agriculture as a soil amendment and in road construction as a subbase had lower environmental impacts compared to conventional methods. However, some common methods of resource utilization have not been thoroughly assessed. In China, the resource utilization of PG primarily focuses on cement retarders and construction materials, but information on these practices is relatively scarce. Currently, China is accelerating the development of a resource recycling-based industrial system. Strengthening the large-scale comprehensive utilization of PG and other bulk industrial solid wastes is a crucial measure to enhance resource efficiency. Nonetheless, the future management plans for PG and their associated environmental and economic performance remain unclear.

To address this research gap, this study compared the environmental and economic impacts of PG stockpiling treatment and resource utilization technologies using a life cycle assessment (LCA) method, based on extensive investigation into the PG disposal in China. Furthermore, variations in environmental and economic impacts under different scenarios of resource utilization technologies were explored. Additionally, the potential for improving the overall environmental and economic impacts of PG treatments in China was discussed. The scientific findings of this study will provide support for decision-making in the management of PG in China.

2. Material and methods

Life Cycle Assessment is a fundamental methodology in international environmental management and product design. According to ISO 14040 (ISO, 2006), LCA comprises four main steps: goal and scope definition, life cycle inventory, life cycle impact assessment, and interpretation.

2.1. Goal and scope definition

The objective of this study is to analyze the environmental and economic impacts of PG disposal in China and to support the development of sustainable strategies for PG resource utilization. This study examines the primary management methods for PG in China, including stockpiling (PS) and three resource utilization technologies: production of cement retarder (PCR), production of construction materials (using β-building gypsum powder as an example) (PBG), and co-production of sulfuric acid with cement (PSC). Firstly, a comparative analysis of the environmental and economic impacts associated with different PG disposal methods was conducted from a lifecycle perspective, and critical processes and substances influencing these impacts were identified. Subsequently, the environmental and economic benefits of PG resource utilization under different configuration scenarios were explored based on the utilization of 1 ton of PG. Finally, an assessment of the total environmental and economic impacts of PG treatment in China was conducted based on actual processing volumes and modes, and optimization scenarios were established to evaluate the potential for environmental and economic improvements of PG disposal.

The functional unit provides a reference for the relevant inputs and outputs of the product (ISO, 2006). In this study, the disposal of one ton of PG (moisture content about 20 %) was defined as the functional unit for comparing the life cycle impacts of different PG disposal methods. All materials, energy consumption, and direct emissions are presented based on the functional unit. The system boundaries were established via a cradle-to-gate approach, as illustrated in Fig. 1.

2.1.1. PG for cement retarder technology

The substitution of natural gypsum with PG as a cement retarder is a common application. However, PG typically contains various impurities, with phosphorus (including soluble phosphorus and lattice-bound phosphorus) and fluorine impurities significantly impacting cement setting time and strength (Costa et al., 2022; Huang et al., 2019). Therefore, the modification treatment of PG is necessary. The most representative methods for PG modification include washing, neutralization, calcination, and combined methods. Among these, alkali neutralization treatment is a simple process with low investment, significant effects, and minimal secondary pollution, making it widely used in cement retarder production (Qin et al., 2023).

This study selects alkali neutralization treatment to modify PG, using quicklime as the modifier. According to on-site studies at typical Chinese firms, the specific process is as follows: after weighing, the PG is sent to the crusher through the feeding system for crushing, while quicklime is mixed with PG in a certain proportion and thoroughly stirred. The mixed materials are then sent to the finished product warehouse for aging, with an aging time generally around 4 to 7 days. This allows the quicklime to fully react with harmful substances such as free acids (phosphoric acid, hydrofluoric acid) in the PG, transforming them into inert salts, thereby reducing their adverse effects on the cement retarder performance.

2.1.2. PG for β-building gypsum technology

PG can be used to produce construction materials, generally consisting of two parts. The first part involves preparing PG into β-building gypsum powder, and the second part involves using the building gypsum powder as raw material, processed and molded into gypsum products such as gypsum blocks, gypsum boards, gypsum bricks, gypsum mortar, etc. The second part follows the same process as natural gypsum powder. Therefore, in this study, only the process of producing β-building gypsum powder from PG is investigated. The specific process is as follows: pre-treatment for iron removal, crushing, drying, screening, calcination, cooling homogenization, grinding, aging, and final packaging. Both drying and calcination heat sources are provided by coal-fired hot air

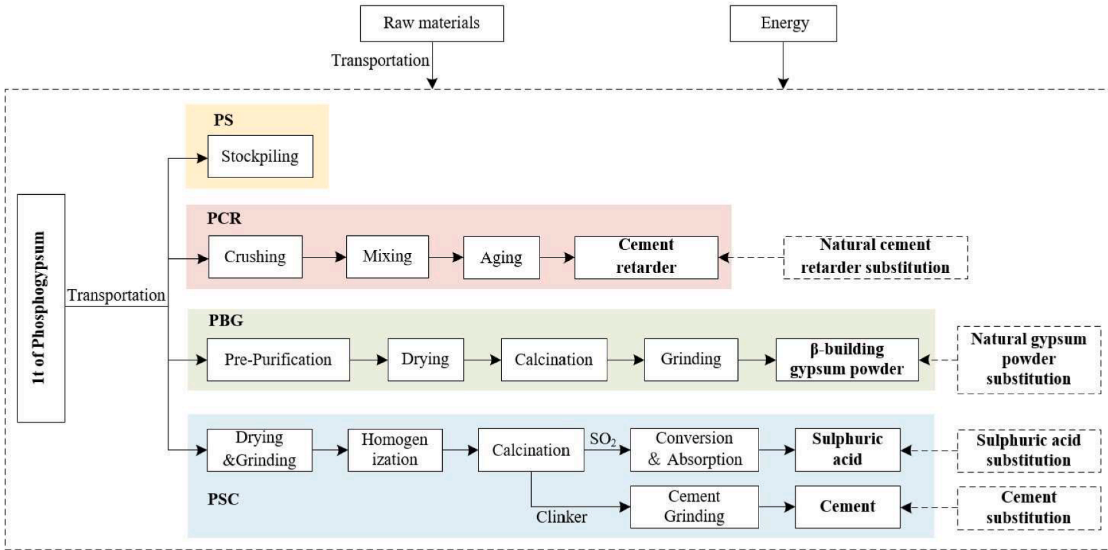

Fig. 1. System boundaries of PG disposal methods. Notes: PS: PG stockpiling; PCR: PG for cement retarder; PBG: PG for β-building gypsum powder; PSC: PG for sulfuric acid co-production cement.

furnaces. In the drying process, coal-fired flue gas (waste heat flue gas) directly contacts the material for drying, controlling the material temperature at around 100 °C, and ensuring that the free moisture content of the material after drying is below 3 %. The calcination process utilizes a fluidized bed calciner, employing a low-temperature calcination process. High-temperature coal-fired flue gas enters the calciner indirectly through heating tubes, controlling the material temperature at 150–160 °C, whereby the PG loses its crystalline water and transforms into β-hemihydrate gypsum. The drying exhaust gas purification system adopts SNCR (Selective Non-Catalytic Reduction) + cyclone dust removal + bag dust removal + dual-alkali desulfurization, and the exhaust gas is discharged when meeting air quality standards after denitrification, dust removal, and desulfurization; other waste gases are discharged after meeting dust removal standards. This process description is derived from on-site research at representative enterprises in China.

2.1.3. PG for sulfuric acid co-production cement technology

The PSC technology involves a thermochemical decomposition reaction conducted in a reducing atmosphere, converting $CaSO_4$ into CaO and SO_2 (Zheng et al., 2014). The sulfur dioxide is further utilized to produce sulfuric acid, while the calcium oxide is used for Portland cement production. Based on field investigations conducted in typical enterprises in China, this technology includes five processes: drying of raw materials, homogenization, preheat and calcination, sulfuric acid preparation and cement grinding. Preheat and calcination, sulfuric acid preparation process are further described below.

Preheat and calcination. A novel dry suspension preheater kiln decomposition calcination technology is employed. The raw materials, after undergoing drying and homogenization processes, are prepared into a raw mixture. The raw mixture enters the suspension preheater, where it is in a suspended state and undergoes indirect heat exchange with SO_2 gas from the kiln tail, resulting in an extremely rapid heat transfer rate. The preheated raw mixture then enters the rotary kiln calcination stage, where the temperature is controlled within the range of 1200–1400 °C. The primary reaction involving PG and coke is represented by Formula 1. Subsequent to calcination, the materials enter the clinker-forming zone, where mineralization reactions occur, leading to the formation of the main mineral components of cement clinker: tricalcium silicate (C3Si), dicalcium silicate (C2Si), tricalcium aluminate (C3A), and tetracalcium aluminoferrite (C4AF). The resultant kiln gas, after undergoing electrostatic dust removal, is directed to the sulfuric acid production facility.

$$2CaSO_4 + C \rightarrow 2CaO + 2SO_2\uparrow + CO_2\uparrow \quad (1)$$

Sulfuric acid preparation. The double-conversion and double-absorption technology was adopted, including dilute sulfuric acid washing and purification, gas drying, the first sulfur dioxide conversion, SO_3 intermediate absorption, the second sulfur dioxide conversion, and final absorption. The catalyst for conversion was a vanadium-based catalyst with a SO_2 conversion rate of 99.9 %. The steam generator supporting the high-temperature absorption tower recovers the waste heat of sulfuric acid flowing out at the bottom, and the amount of steam (0.8 MPa) recovery was 0.4t/t acid.

2.2. Life cycle inventory (LCI) and data sources

The Life Cycle Inventory (LCI) of PG integrated utilization technologies is detailed in Table 1. Three representative companies were selected to provide the inventory for PCR, PBG, and PSC technologies. Foreground data, including the consumption of materials and energy, waste emissions and concentrations, transportation distances and modes, and product outputs, were obtained from annual statistics and monitoring data of these representative enterprises and published literature. The background inventory data were sourced from the Chinese Core Life Cycle Database (CLCD), which offers comprehensive data specific to Chinese industrial processes and materials (Chen et al., 2019). More detailed sources of the LCI are provided in Tables S1-S3. Since there is no inventory data for PG stockpiling in the CLCD database, the LCI dataset named "treatment of waste gypsum, sanitary landfill, Switzerland (CH)" in the Ecoinvent v3.1 databases is the best available proxy data for it.

It is important to note that the CLCD database does not contain background inventory data for the production of cement retarders and β-building gypsum powder using conventional methods. Therefore, we constructed models for the conventional production of these materials based on natural gypsum. The foreground data for these models were obtained by investigating several typical enterprises, representing the average level of the Chinese market. Background data for these models were sourced from the CLCD database. The LCIs for natural cement retarder and β-building gypsum powder are presented in Table S4.

Table 1
Life cycle inventory analysis of different PG resource-utilization technologies.

Categories	Substances	Unit	PCR	PBG	PSC
Resource consumption	Clay	kg			52.00
	Natural plaster	kg			16.00
	Fly ash	kg			79.50
	Slag powder	kg			37.10
	Catalyst	kg			1.80 E-3
	Quicklime	kg	30.93	1.84	
	Sodium hydroxide (30 %)	kg		0.62	
	Ammonia (20 %)	kg		0.44	
	Water	kg	3.09	8.57	1332.00
	Coke	kg			52.00
Energy consumption	Coal	kg		41.43	120.00
	Natural gas	m^3			0.35
	Electricity	kWh	2.06	10.71	90.00
Emissions to air	Particulates	kg	3.09 E-3	6.90E-02	8.80 E-2
	SO_2	kg		4.80E-02	1.96 E-1
	NO_x	kg		1.40E-01	7.47 E-1
	Sulfuric acid mist	kg			8.00 E-3
	NH_3	kg		7.10E-02	8.00 E-3
	HCl	kg			5.80 E-3
	Fluoride	kg		6.40E-03	1.37E-02
	CO_2	kg		108.46	408.16
Emissions to water	Wastewater	m^3	100 % reused	100 % reused	100 % reused
Emissions to soil	Solid waste	kg	100 % reused	100 % reused	100 % reused
Product	Sulphuric acid (98 %)	kg			408.00
	Cement	kg			533.00
	Cement retarder	kg	1030.00		
	β-building gypsum powder	kg		714.00	
	Steam (0.8Mpa, 200 °C)	kg			124.00

2.3. Life cycle impact assessment (LCIA)

The ReCiPe 2016 V1.1 Midpoint (H) model was chosen in this study to evaluate the environmental impacts of PG treatment. This model is widely recognized for its comprehensive coverage of environmental impact categories and its applicability to various industrial processes, and it has been extensively validated and used in many studies (Chen et al., 2019; Chen, Z. et al., 2022; Liu et al., 2022). The 16 environmental impact indicators are selected from three aspects: resource and energy consumption, human health and ecological toxicity, environmental impact. Resource and energy consumption categories include fossil resource depletion potential (FDP), metal depletion potential (MDP), freshwater consumption potential (FWC) and land use (LU), which are drivers of environmental impacts. Human health and ecological toxicity indicators include human toxicity (cancer) potential (HTP), human toxicity (non-cancer) potential (NHTP), marine ecotoxicity potential (METP), terrestrial ecotoxicity potential (TETP) and freshwater ecotoxicity potential (FETP). Common environmental damage indicators include freshwater eutrophication potential (FEP), terrestrial acidification potential (TAP), ionizing radiation potential (IRP), global warming potential (GWP), photochemical oxidant formation potential: ecosystems (EOFP), particulate matter formation potential (PMFP) and ozone depletion potential (ODP).

2.4. Techno-economic analysis models

Besides assessing the environmental impact of different PG treatment methods, a techno-economic analysis (TEA) was also conducted to evaluate their economic performance. The stockpiling of PG, due to the absence of product output, only considers input costs, mainly including: capital depreciation for PG stockpile facility construction, labor, routine maintenance, and post-closure restoration. This paper does not consider the solid waste environmental protection tax for PG stockpile. According to relevant regulations, facilities or sites storing or disposing of solid waste in compliance with national and local environmental protection standards are not required to pay environmental protection taxes. The economic impact of PG resource utilization technologies is the comprehensive value of technology costs and product revenue. Lifecycle costs include five main cost categories: raw materials, energy, depreciation, labor, and environmental protection taxes, while economic income comes from product revenue generated by PG resource utilization. The costs of raw materials, energy, labor, and product revenue are all based on the average values in the current Chinese market, derived from sources such as market price monitoring data of the National Bureau of Statistics, industry reports, and relevant literature. Depreciation is derived from enterprise data and literature reports. Routine maintenance and post-closure restoration are estimated based on the current Chinese market. Environmental tax is calculated based on *Environmental Protection Tax Law of the People's Republic of China* (2018). Detailed information on these sources is provided in the Supplementary Information (Tables S11-S13).

The total cost of PG stockpile can be calculated as follows:

$$P_C = P_D + P_L + P_{RM} + P_R \tag{2}$$

Where, P_C is total costs, P_D is capital depreciation costs, P_L is labor costs, $P_{RM}$is routine maintenance costs and P_R is post-closure restoration costs.

The total economic benefits of PG resource utilization technologies can be calculated as follows:

$$P = P_P - P_C \tag{3}$$

$$P_C = P_M + P_E + P_D + P_L + P_{ET} \tag{4}$$

Where, P represents the total economic benefits, P_P is products revenue, P_C is total costs, P_M is material costs, P_E is energy costs, P_D is capital depreciation costs, P_L is labor costs, and P_{ET} is environmental protection taxes.

2.5. Scenario analysis

To elucidate the potential improvement in the environmental and economic impacts of PG treatment in China, a scenario analysis was conducted to evaluate the enhancements in future PG treatment modes. Based on the current status of PG treatment in China, reasonable predictions were made for improvement measures. To simplify calculations, this study only analyzed scenario changes in stockpiling and three utilization technologies. The proportions of other disposal methods, such as external sales, ecological restoration, and mine filling, remained unchanged; and accordingly their environmental and cost impacts were not accounted for. We used the 2021 production volume of PG in China as the baseline. Currently, the comprehensive utilization rate of PG is 45.6 %, with the proportions of PS, PCR, PBG, and PSC being 54.4 %, 12.7 %, 10.8 %, and 1.4 % respectively (see Fig. S1). With the continuous tightening of China's environmental policies and the implementation of policies to strengthen the comprehensive utilization of industrial solid waste, the PG utilization rate is expected to increase. According to the *Implementation Plan on Accelerating the Comprehensive Utilization of Industrial Resources* issued by the Chinese government, the goal is to achieve a 73 % utilization rate for industrial gypsum by 2025 (MIIT, 2022). As PSC technology is an important development direction for large-scale application of PG, its proportion is expected to gradually increase. Based on the growth of PG utilization efficiency and the development intensity of PSC technology, three scenarios for future PG treatment modes were established: conservative, moderate, and optimistic (see Table S5). Under these scenarios, the utilization rate of PG is expected to increase by 6.4 %, 15.4 %, and 27.4 % respectively. The proportion of PBG technology is expected to increase by 1.0 %, 2.5 %, and 4.5 % respectively, while the proportion of PSC technology is expected to increase by 5.0 %, 12.0 %, and 21.5 % respectively.

2.6. Uncertainty and sensitivity analysis

To assess the uncertainty of the LCA results, Monte Carlo analysis with 5000 iterations was conducted. This method, widely utilized in the field of LCA, allows for the propagation of uncertainty through the model (Bai et al., 2022; Wang et al., 2024). In this study, the coefficient of variation (CV) was employed to quantify the uncertainty across various impact categories. Sensitivity analysis is a crucial component of LCA, reflecting how small variations in input parameters propagate and affect the magnitude of LCA results (Liu et al., 2015). It helps identify sensitive parameters and potential sources of uncertainty. In this study, we also performed a sensitivity analysis to determine how a 5 % variation in each input parameter, when changed individually, affects the LCA results of different PG utilization technologies in China.

3. Results and discussion

3.1. LCA results of different PG disposal methods

We conducted a lifecycle inventory analysis of four PG treatment/utilization technologies, obtaining midpoint impact categories for treating 1 ton of PG, as depicted in Fig. 2. Overall, all impact categories of PG stockpiling show positive values, indicating the highest environmental burden, consistent with previous studies (Tsioka and Voudrias, 2020; Zhang, 2016). On one hand, the stockpiling process requires significant land occupation; on the other hand, it releases pollutants into water bodies, soil, and air. In contrast, most impact categories for the three utilization technologies show negative values, with a few exceptions being positive values but still lower than those for stockpiling. This suggests these technologies have positive environmental effects, reducing potential threats to human health and the environment.

In terms of resource and energy consumption categories, both PBG and PCR technologies have relatively minor impacts on FDP (−16.7 kg oil-Eq and −16.2 kg oil-Eq, respectively), attributed to the substitution effects of the products generated by these two technologies. These technologies can produce recycled products that replace those produced from raw materials, thereby reducing fossil resource consumption. Most savings come from avoiding energy consumption associated with the extraction and crushing processes of natural gypsum mines, which strongly inhibits fossil resource depletion. Conversely, PSC technology has the highest impact on FDP (20.2 kg oil-Eq), indicating a greater requirement for fossil fuel input. This is due to the greater fossil energy consumption associated with drying and the reductive decomposition of PG, which hampers the widespread application of this technology. However, compared to the other two technologies, PSC technology can save more copper-equivalent metal resources and water resources (−15.8 kg Cu-Eq and −2.0 m^3, respectively).

In terms of human health and ecological toxicity categories, all three utilization technologies exhibit excellent environmental performance. This is mainly because the products generated by these technologies avoid the extraction of mineral resources typical in conventional production, thus reducing emissions from mineral resource extraction. For instance, Zn discharged into freshwater from metal ore mining is the primary influencing factor for NHTP, FETP, and METP, while hexavalent chromium discharged into freshwater is the main influencing factor for HTP, and Cu and V discharged into the atmosphere are key substances for TETP.

In terms of environmental impact categories, PSC technology outperforms the other two utilization technologies in TAP, GWP, PMFP, and ODP. It can significantly reduce emissions of SO_2-Eq, CO_2-Eq, $PM_{2.5}$-Eq, and CFC-11-Eq, bringing considerable environmental benefits. Notably, as mentioned in the previous section, although PSC technology consumes the most fossil resources, its impact on global warming potential (GWP) is the smallest (−83.9 kg CO_2-Eq). According to the calculations, the greenhouse gas emissions generated by PSC technology are reduced by 13.3 % compared to conventional production processes. This reduction is primarily because the process reduces CO_2 emissions compared to the traditional method of carbonate calcination used in the cement industry (Zheng et al., 2014). Furthermore, PSC technology also reduces the use of pyrite, limestone, and other auxiliary raw materials, indirectly reducing carbon emissions. In contrast, PCR technology achieves relatively fewer carbon emissions reductions (−4.75 kg CO_2-Eq), because this technology requires quicklime to neutralize acidic impurities in PG. The preparation of quicklime involves the high-temperature calcination of limestone, which emits a considerable amount of CO_2. Additionally, it can be seen from the PMFP that PCR technology emits more particulate matter than natural gypsum, which is also related to the use of quicklime.

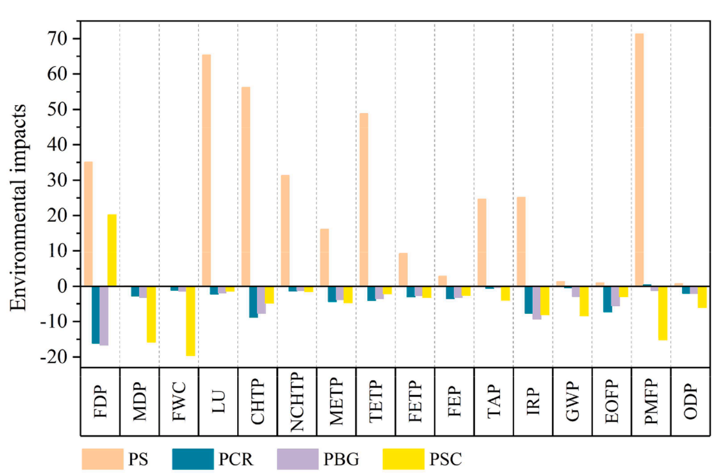

Fig. 2. LCA results for different PG disposal methods. Notes: The units used in this figure are as follows: FDP kg oil-Eq, MDP kg Cu-Eq, FWC 10^{-1} m^3, LU 10^{-2} m^2·yr annual crop land-Eq, CHTP 10^{-2} kg 1,4-DCB-Eq, NCHTP kg 1,4-DCB-Eq, METP 10^{-2} kg 1,4-DCB Eq, TETP kg 1,4-DCB-Eq, FETP 10^{-2} kg 1,4-DCB-Eq, FEP 10^{-4} kg P-Eq, TAP kg SO_2-Eq, IRP 10^{-4} kBq Co-60-Eq, GWP 10 kg CO_2-Eq, EOFP 10^{-1} kg NO_x-Eq, PMFP 10^{-1} kg $PM_{2.5}$-Eq, and ODP 10^{-5} kg CFC-11-Eq.

3.2. Process-specific environmental impact analysis

The contribution of each process and material flow to the environmental impacts for each technology is illustrated in Fig. 3. For PCR technology, the consumption of the neutralizing modifier quicklime is identified as the primary contributor to environmental burdens, accounting for 78.8 % to 99.9 % of FDP, MDP, LU, TAP, GWP, EOFP, PMFP, and ODP. Previous studies (Chen, B. et al., 2022; Wu et al., 2022) have confirmed the feasibility of utilizing industrial alkaline waste, such as calcium carbide residue, instead of quicklime for neutralization and modification of PG. Therefore, the application of industrial alkaline waste is crucial for reducing environmental impacts associated with PCR technology.

For PBG technology, the main negative impact is caused by energy consumption, particularly coal and electricity. The production and combustion of raw coal contribute 80.6 % of FDP and 75.9 % of GWP respectively. The consumption of electricity mainly contributes to human health and ecological toxicity, as well as TAP, IRP, EOFP, PMFP and ODP, with a contribution rate of 46.3-89.4 %. The substitution of building gypsum powder can completely offset all categories of environmental burdens, resulting in significant environmental benefits.

For PSC technology, the intensive consumption of fuel coal and reducing agent coke, both integral to its production, is the primary contributor to FDP. The calcination process of PG is the main stage of coal consumption, accounting for approximately 60 % of total coal use, while the drying of raw materials, especially PG with high moisture content, accounts for nearly 40 % of coal use. Correspondingly, the emissions of carbon dioxide from coal combustion and coke reduction dominate GWP. The emissions of nitrogen oxides from coal combustion are the main contributors to EOFP. Electricity consumption mainly contributes to IRP and the toxicity potentials, with the most critical substances being Cr (VI), Cu, Zn discharged into freshwater and Cu discharged into the atmosphere during electricity production. Notably, the environmental impact of the transportation of raw materials, especially raw coal and coke, cannot be ignored, with contributions to human health and ecological toxicity potentials, LU, FEP, and ODP accounts for 12.7–25.4 %. Therefore, reducing energy consumption and future clean energy substitution are key to minimizing the environmental impact of this technology. Traditional sulfuric acid, cement and steam industries are energy-intensive and high-emission industries. By substituting these products with PSC technology, substantial environmental benefits can be achieved, fully compensating for the environmental harm caused by this technology, with the exception of FDP.

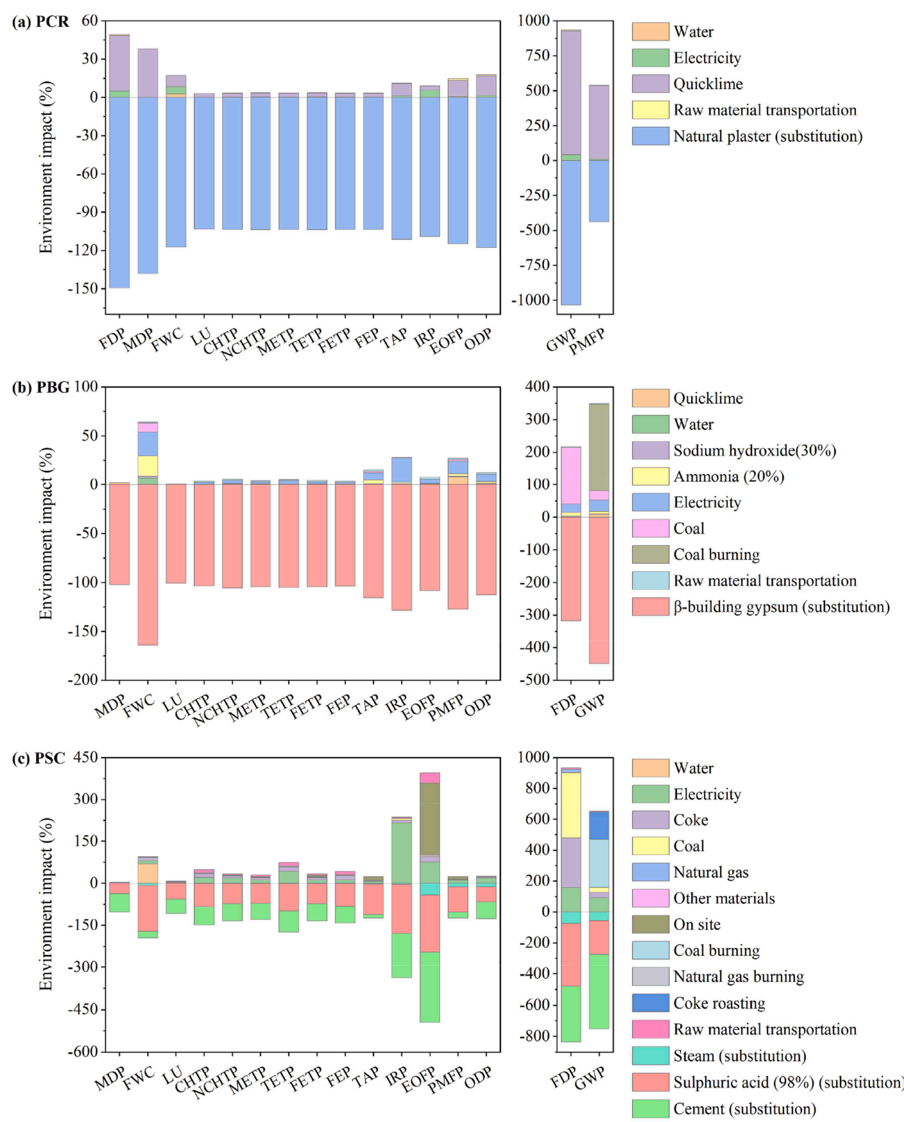

Fig. 3. Environmental impact contribution analysis of different PG resource-utilization technologies.

3.3. Sensitivity and uncertainty analysis

Tables S8-S10 list the highly sensitive parameters that significantly influence the LCA results of the three PG utilization technologies. A highly sensitive parameter is defined as one where a 5 % variation in input data results in a change in LCA results exceeding 1 %. The sensitivity analysis shows that the LCA results of the PCR technology are significantly affected by lime consumption and the substitution rate of cement retarders. For the PBG technology, the sensitive inventory data include electricity consumption, coal input, and product yield. In the case of the PSC technology, in addition to raw materials, energy, and product yield, the sensitive inventory data also include environmental emissions (NO_x) data. To ensure the reliability of our results, it is important to note that most of the prospective data in this study were derived from field research, which generally has low uncertainty. The data on raw material consumption, energy use, and product yield for the three utilization technologies were mainly obtained from investigations and published literature, ensuring good quality of the original data. As for the pollutants emitted into the atmosphere, the data come from on-site monitoring reports of enterprises, which are also of sound quality.

Following the sensitivity analysis, the uncertainty of the LCA results was further analyzed, with findings presented in Table S7. For most impact categories across the three PG utilization techniques, the coefficient of variation (CV) values were relatively low, with values less than 10 %. This indicates minimal data variability for these categories. However, certain individual metrics exhibited higher CVs, such as 33.6 % and 17.2 % for GWP and PMFP in the PCR technology, 19.8 %, 15.6 %, 29.2 % and 10.6 % for FDP, TAP, GWP and PMFP in the PBG technology, and 34.2 %, 24.4 %, 38.4 % and 19.3 % for FDP, IRP, GWP and EOFP in the PSC technology. These higher CV values suggest greater variability in these specific impact categories.

3.4. Techno-economic assessment

The economics of each treatment technology are analyzed and discussed in terms of cost and yield over its life cycle, as shown in Fig. 4. The specific cost and product revenue details can be found in Tables S11 to S13. The total lifecycle cost of PG stockpiling is 31 CNY per ton of PG. Capital depreciation for PG stack construction constitutes the largest proportion, accounting for approximately 61 % of the total cost. With China's increasing requirements for solid waste management - especially following the issuance of regulations such as the *Safety Technical Regulation on Phosphogypsum Stack* (SAWS, 2017) and the revised *Pollution Control Standard for Storage and Disposal Sites of General Industrial Solid Waste* (MEE, 2020), which impose more specific and stringent requirements on the impermeable system and closure of PG stacks - the construction and closure restoration costs of PG stacks have significantly escalated, accounting for about 34 % and 9 % of the total costs, respectively.

For all three PG utilization technologies, PG is provided free of charge. From the perspective of total cost, the PCR technology has the lowest cost at 33 CNY, as its process is the simplest and requires less raw material. In contrast, the PSC technology has the highest cost at 383 CNY, due to its higher consumption of raw materials and energy, particularly coke, coal, and electricity, which account for 39 %, 23 %, and 15 % of the total cost, respectively (Table S13). The cost of the PBG technology is slightly higher than that of the PCR technology, at around 57 CNY, with energy consumption being the primary cost driver, constituting 80 % of the total cost.

In terms of revenue, the income generated by the three utilization technologies comes from the value of recovered PG-based products. The PSC technology yields the most diverse range of products with the highest economic value. For every ton of PG, approximately 0.41 tons of 98 % sulfuric acid, 0.53 tons of cement, and 37.6 GJ of low-pressure steam can be recovered, with an average value of around 495 CNY. In

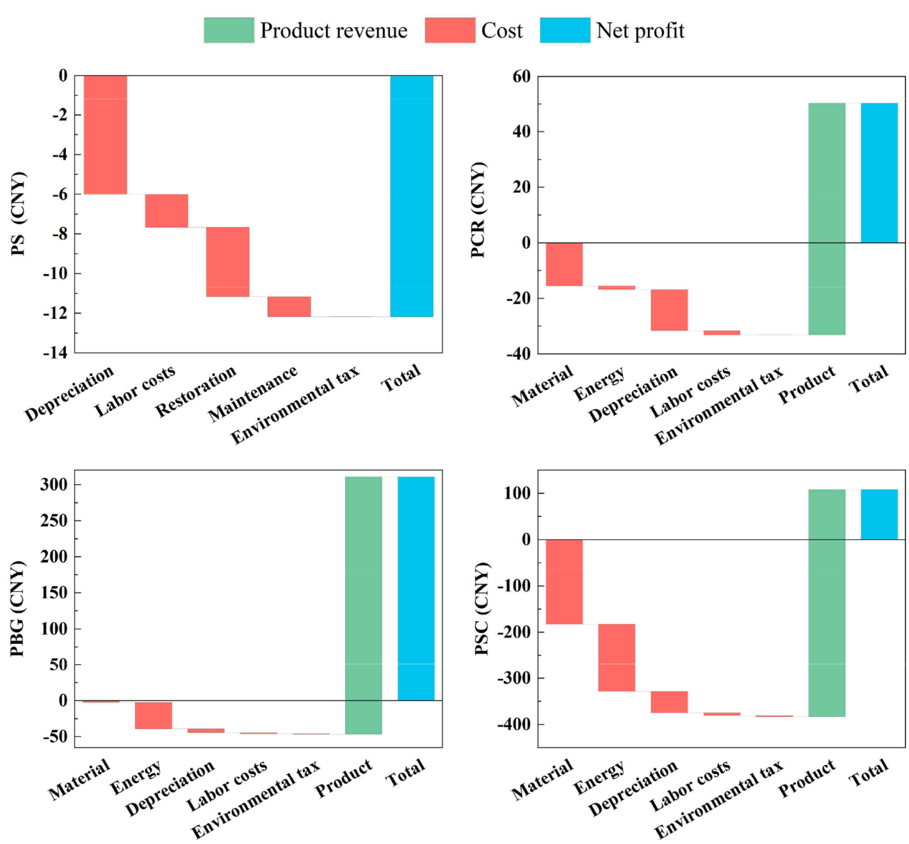

Fig. 4. Economic evaluation of different PG disposal methods.

the case of PBG technology, the recovered β-building gypsum powder serves as a raw material for other gypsum products such as gypsum boards, blocks, and mortar. With the continuous growth in China's construction market demand, the demand for building gypsum is increasing, leading to a rise in the price of gypsum powder, reaching approximately 500 CNY per ton of PG. Hence, the economic income from PBG technology is also significant. On the other hand, the revenue from PCR technology, which produces cementitious materials, is relatively lower, as it belongs to low-value-added products. The income from this technology is only 23 % of that from PBG technology and 17 % of that from PSC technology. However, if the recovered products from all three technologies enter the market circulation, their prices will also be influenced by the market prices of raw materials.

Overall, the net economic profit of the PBG technology is the highest, at 311 CNY per ton of PG. The PCR technology has the lowest net profit, only 50 CNY. The PSC technology's net profit is approximately 108 CNY. All three utilization technologies can generate positive economic benefits in the current market.

3.5. Scenario analysis

To explore optimal utilization patterns for PG resources, this study focuses on 1 ton of PG, adjusting the proportions of the three utilization technologies to assess the environmental and economic benefits under different scenarios. As shown in Fig. 5 and Fig. S2, among the three resource utilization technologies, when the proportion of PBG or PCR technology remains constant and the proportion of PSC technology increases, it is not conducive to reducing the potential depletion of fossil resources. When the proportion of PSC technology exceeds approximately 0.44, FDP becomes positive, indicating the need to deplete more fossil resources. For MDP and FWC, when the proportion of PBG or PCR technology remains constant and the proportion of PSC technology increases, more metal and water resources can be saved. When the PSC technology accounts for the largest proportion, the benefit of metal and water resources saving reaches its peak, being 5.7 times and 17.2 times the minimum savings, respectively. In terms of human health and ecological toxicity potentials, PCR technology is more conducive to increasing the environmental benefits of CHTP and TETP, while PSC technology is more conducive to increasing the environmental benefits of NCHTP, METP, and FETP. However, the difference between the maximum and minimum environmental benefits is not significant, ranging from 19 % to 47 %. Regarding TAP, GHG, and PMFP, when the proportion of PSC technology increases, the reduction in SO_2-eq acid gases, CO_2-eq greenhouse gases, and $PM_{2.5}$-eq particulate matter increases. The benefit of associated emission reduction reaches its peak when PSC technology accounts for the largest proportion. Overall, when the proportion of PBG or PCR technology remains constant, increasing PSC technology can achieve more environmental benefits. Fig. 5(e) shows the economic benefits of utilizing 1 ton of PG under different scenarios. When the proportion of PCR or PSC technology remains constant and the proportion of PBG technology increases, the economic benefits increase. When the proportion of PBG technology is at its maximum, the economic benefits reach their peak.

In China, the production and distribution of PG are significantly influenced by the distribution of phosphate ore resources, exhibiting clear regional concentration (Chuan et al., 2018). According to data from 2021, the top five provinces for phosphate fertilizer production are Hubei, Yunnan, Guizhou, Sichuan, and Anhui, which together account for approximately 89.5 % of the national total PG production. Among them, Hubei Province leads with the highest PG production,

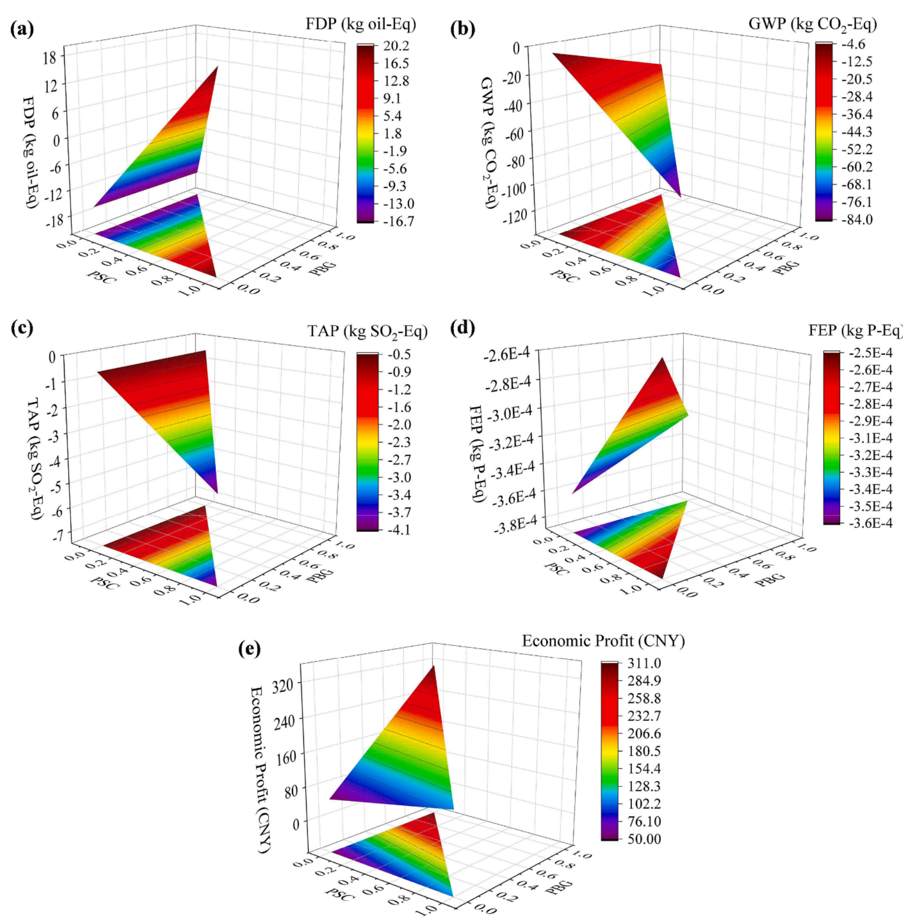

Fig. 5. Environmental impact of main indicators and economic impact for 1t PG resource-utilization under varying scenarios.

representing 37.8 % of the national total, followed by Yunnan Province at 26.8 %. Due to transportation cost constraints, the sales radius for PG-based building products (such as cement retarders and β-building gypsum) is generally limited to within 100 km and is primarily confined within each province. According to Table S14, based on the cement retarder market capacity of each province, Sichuan and Anhui can essentially absorb their own PG production annually, while Hubei, Yunnan, and Guizhou can absorb only 15.2 % to 34.3 % of their annual production. However, if the PSC technology is adopted, the sulfuric acid product will be entirely reused in the original phosphoric acid unit. Meanwhile, the output of co-produced cement will only account for 1.3 % to 13.6 % of each province's cement production, thereby not affecting the existing cement production lines. This enables the complete recycling of PG, achieving 100 % circular utilization. Furthermore, comprehensive PG utilization must address not only current production volumes but also historical stockpiles to achieve reduction. Therefore, the PSC technology is an important development direction for the large-scale application of PG. Considering the environmental and economic benefits and feasibility of large-scale implementation, we recommend vigorously developing the PSC technology to eliminate the environmental impacts of PG stockpiling. This advancement would promote the sustainable development of the phosphorus chemical industry and contribute to achieving carbon peak and neutrality targets.

According to statistics from the China Phosphate Compound Fertilizer Industry Association, PG production in China was approximately 80 million tons in 2021. Fig. 6(a) illustrates the overall environmental impact and economic outcomes of PG disposal. The environmental impact mainly stems from stockpiling the majority of PG. In contrast, resource utilization technologies have provided positive environmental benefits across 16 indicators. The PCR and the PBG technologies have similar proportions, with environmental benefits primarily in resource and energy consumption, FEP, GWP, EOFP, and ODP indicators. Together, these two technologies have saved 308.9 kt oil-Eq of fossil resource, 55.8 kt Cu-Eq of metal resource, and 2.3×10^6 m^3 of water resource. They have also reduced emissions by 6.4 t of P-Eq nutrients, 301.2 kt CO_2-Eq of greenhouse gases, 12.3 kt NO_x-Eq of photochemical oxidants, and 378.0 kt CFC-11-Eq substances. Despite its small proportion (1.4 %), the PSC technology has not fully demonstrated its advantages in environmental benefits in terms of TAP, GWP, PMFP, ODP, etc. However, it still positively impacts reducing the consumption of metal resource and water resource, contributing 26 % and 49 % of reduction, respectively. Economically, PG storage disposal incurred a cost of 2.43 billion CNY, while the three utilization technologies generated 3.33 billion CNY in economic revenue, with the PBG technology accounting for the largest share at 80 %. Overall, the current PG disposal mode in China has resulted in certain environmental benefits in terms of MDP, FWC, EOFP, and ODP, and generated a net economic benefit of 900 million CNY.

To explore the potential of PG treatment in improving environmental and economic outcomes, we have established three optimization

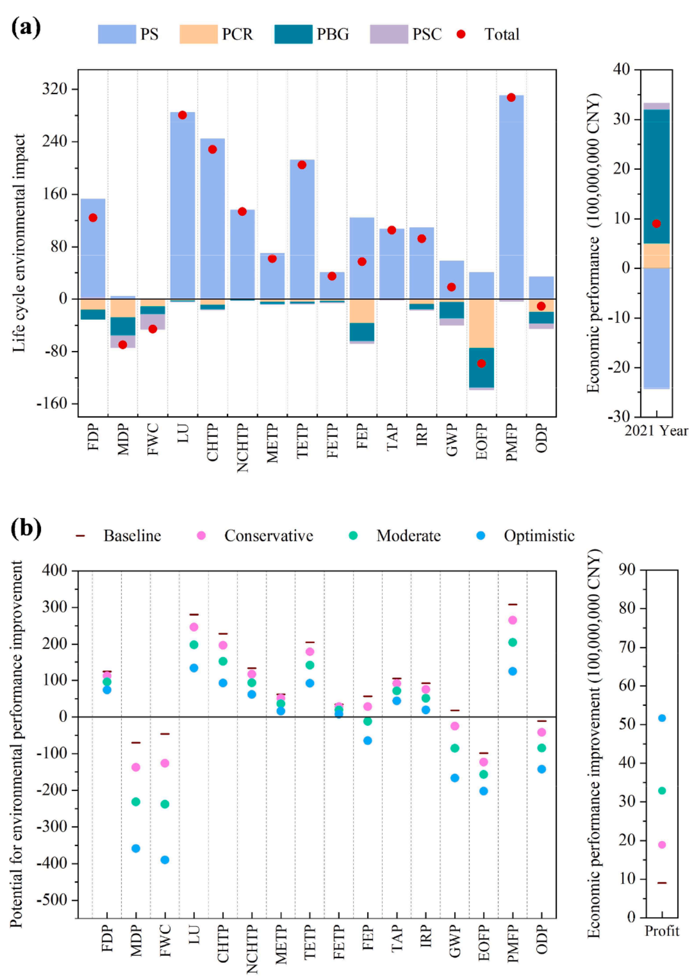

Fig. 6. Environmental and economic impact of China's PG disposal in 2021 and potential changes under different scenarios. (a) Environmental and economic impact in 2021; (b) Potential changes under different optimized scenarios. Notes: The units used in this figure are as follows: FDP 10 kt oil-Eq, MDP kt Cu-Eq, FWC 10^5 m^3, LU 10^5 m^2·yr annual crop land-Eq, CHTP 10^2 t 1,4-DCB-Eq, NCHTP 10 kt 1,4-DCB-Eq, METP 10^2 t 1,4-DCB Eq, TETP 10 kt 1,4-DCB-Eq, FETP 10^2 t 1,4-DCB-Eq, FEP 10^2 kg P-Eq, TAP 10 kt SO_2-Eq, IRP MBq Co-60-Eq, GWP 10 kt CO_2-Eq, EOFP 10^2 t NO_x-Eq, PMFP kt $PM_{2.5}$-Eq, and ODP 10 kg CFC-11-Eq.

scenarios and evaluated their overall impact. As depicted in Fig. 6(b), increasing the proportion of PG resource utilization significantly enhances both environmental and economic impacts. In the conservative, moderate, and optimistic scenarios, it can save 117.4, 283.5, and 500.0 kt oil-Eq of fossil resource, 67.2, 161.5, and 289.0 kt Cu-Eq of metal resources, as well as 8.0×10^6, 1.9×10^7, and 3.4×10^7 m^3 of water resources, respectively. In the optimistic scenario, compared to the current disposal plan, environmental impacts in categories such as human health and ecological toxicity potential can be reduced by 54 % to 78 %. Additionally, the environmental impacts of FEP, TAP, GWP, and PMFP can be reduced by 12.1 t P-Eq, 611.9 kt SO_2-Eq, 1845.3 kt CO_2-Eq, and 183.0 kt $PM_{2.5}$-Eq, respectively. Moreover, the environmental benefits of EOFP and ODP can be increased by 10.4 kt NO_x-Eq and 1.3 t CFC-11-Eq. In the conservative, moderate, and optimistic scenarios, the economic benefits for PG treatment may increase by 9.8, 23.9, and 42.6 billion CNY, respectively. Therefore, increasing the resource utilization and recycling of PG, especially through the application of PSC technology, will be a crucial step towards achieving efficient and large-scale utilization.

3.6. Implications for the future management of PG in China

Based on the findings of this study, we propose the following recommendations for the future management of PG in China.

Promote technological innovation and application. Encourage collaboration between research institutions and enterprises to jointly optimize existing technologies, enhancing both environmental and economic benefits. According to the findings in Sections 3.1 and 3.2 of this study, the use of quicklime as a neutralizing and modifying agent in PCR technology imposes a significant environmental burden. Therefore, it is recommended to use industrial alkaline waste, such as calcium carbide slag, for neutralization and modification to reduce environmental impacts. For PBG and PSC technologies, reducing energy consumption and transitioning to cleaner energy sources are crucial for minimizing environmental impacts. Additionally, PSC technology faces challenges related to cost investment and economic efficiency. Thus, there is an urgent need for breakthroughs in technological innovation, particularly in key areas such as kiln-external PG decomposition and non-thermal dewatering and drying process. Furthermore, promote enterprises and research institutions to develop more advanced and higher-value resource utilization technologies.

Accelerate the popularization and application of large-scale and high-value comprehensive utilization technologies. Currently, PG utilization in China is dominated by primary pathways, with low value-added products and challenges in industrial scale expansion. In order to maximize the economic and environmental benefits, regions should give priority to the development of PSC technology and other utilization pathways that have the best environmental benefits and can consume PG on a large scale. It is suggested that these technologies should be included in the catalog of the national key promotion technologies, guiding enterprises to vigorously develop related industries, thus accelerating the transformation of PG into high-value and large-scale utilization. In addition, it is recommended that the state and local relevant departments formulate, introduce and implement preferential tax policies, financial subsidies and incentives for the PG application market to boost enterprises enthusiasm.

Formulate comprehensive utilization products and engineering application standards. Due to the lack of universal standards for PG recycling products, market acceptance is low and the products are difficult to promote. Therefore, it is recommended to establish and introduce provincial and even national universal standards for harmless treatment and recycling products as soon as possible, along with supporting engineering standards. This will ensure that enterprises have guidelines to follow and operational methods to adhere to, improving the market acceptance of recycled products.

4. Conclusion

We assessed the environmental and economic impacts of PG stockpiling and three typical resource utilization technologies from a lifecycle perspective. By analyzing the overall environmental and economic benefits across different resource utilization scenarios on a national scale in China, we proposed optimization strategies for PG management. The main findings are as follows: all three PG resource utilization technologies demonstrate clear environmental benefits. Among them, the PSC technology exhibits the most significant environmental advantages in impact categories such as MDP, FWC, TAP, GWP, PMFP, and ODP. Economically, all three resource utilization technologies yield positive net benefits, with PBG technology providing the highest net economic gain. Notably, PSC technology could achieve 100 % circular utilization of PG, thereby mitigating the environmental burden of long-term stockpiling. This positions PSC technology as a highly promising solution for sustainable and large-scale PG management. To achieve optimal dual environmental and economic benefits, we strongly recommend the widespread adoption of PSC technology. This study provides valuable methodological and technical insights for the future management of PG in China.

CRediT authorship contribution statement

Lu Chen: Writing – original draft, Methodology, Investigation, Conceptualization. **Xiaoyu Luan:** Formal analysis. **Feng Han:** Formal analysis. **Yuwei Zhao:** Investigation. **Huiying Yang:** Investigation. **Long Zhang:** Data curation. **Yongquan Yin:** Data curation. **Wei Liu:** Writing – review & editing, Supervision. **Zhaojie Cui:** Writing – review & editing, Project administration.

Declaration of competing interest

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

Data availability

I have shared the link to my data at the Attach File step

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Supplementary materials

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