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Life Cycle Assessment of Phosphogypsum as Filler Material for Coal Fly Ash-Based Geopolymer

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Abstract

The global accumulation of phosphogypsum (PG), with annual generation exceeding 175 Mt/year, presents significant environmental challenges. While studies have demonstrated PG's potential as a filler material in geopolymer composite, comprehensive environmental impact assessments of such valorization approaches remain limited. This study presents the first comparative life cycle assessment (LCA) of acid- and alkali-activated PG-CFA geopolymers in the context of sustainable industrial waste management. Geopolymer technology can eliminate the need for traditional landfilling of PG in coastal areas and, therefore, reduce their negative environmental impacts. LCA was conducted to assess the impacts of repurposing 1kg functional unit of PG as geopolymer precursors coupled with acid- and alkali-based activators compared to the current disposal practices of these solid wastes. The inventory was modeled after a phosphoric acid plant using the wet dihydrate process, a coal-fired power plant, and a laboratory-scale coal fly ash-phosphogypsum geopolymer (FAPG) synthesis upscaled for industrial application. The most number of environmental benefits was observed for acid FAPG particularly via reductions in CO_2-eq emissions by 40%, 90% in energy consumption, and 36% in mineral resource extraction. Alkali FAPG excelled in water acidification and scarcity by 60% and it could outperform acid FAPG environmentally via sensitivity analysis under a similar formulation blend. Further research can focus on optimizing FAPG formulation, finding alternatives for the acid and alkali activators, and reviewing industrial standards for widespread FAPG applications. These results imply the potential of integrating FAPG manufacturing in PG- and CFA-generating industries to emulate a circular economy.

Keywords: Geopolymer; Waste Valorization; Impact Assessment; Construction Material; Phosphogypsum.

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1. Introduction

Phosphoric acid (PA) is a basic chemical raw material critical for the production of phosphorus-rich fertilizers [1, 2]. It is also essential in the food industry, so there exists a widespread global activity of producing PA. Production of PA from phosphate-bearing rocks via wet processing accounted for about 90% of production globally [3-5]. However, PA manufacturing generates large amounts of waste such as phosphogypsum (PG), which often contain heavy metals and radioactive materials because phosphate-bearing ores are naturally occurring radioactive materials (NORM) [6]. For example, a ton of raw PA generated via wet processing produces approximately 4−5 tons of PG [7]. In 2024, 175 million tons (Mt) of PG was generated, and it has been estimated that more than 3 billion tons (Bt) were dumped/stored near coastal areas found in 65 countries, including Morocco, Tunisia, US, China, and Spain [8]. Unfortunately, only 15% of this waste is repurposed globally for cement production [9]. A local PA plant has been engaged in waste reduction efforts increasing the percentage recycled by 30-40% [10]. The huge volume generation worldwide continues to pose major socio-environmental issues due to the potential leaching of hazardous elements like arsenic, selenium, boron and heavy metals like lead and zinc in coastal regions [11]. To reduce its impact, several studies have been published to repurpose and reprocess PG, including soil amendment [12], rare earth element (REE) extraction, carbon dioxide (CO_2) sequestration [8], road-base materials [13], and construction materials [11].

One particularly promising application for PG that could address the issues highlighted previously is geopolymer technology. Geopolymers, as introduced by Davidovits in the late 1970s, are inorganic binders formed via a process called geopolymerization [14, 15]. Geopolymers have several applications, especially in the construction industry, particularly as a cement alternative to address the unsustainable impact of clinker calcination for cement production [16]. During geopolymerization, aluminosilicate-rich precursors are dissolved by an activator, typically a strong alkaline compound, polymerizing into Al-Si-O chains with the help of polysilicate-bearing compounds [15]. Municipal and industrial wastes such municipal sewage sludge incineration (MWSI) ash [17, 18], rice husk ash [19, 20], blast furnace slag [21], bottom ash [22], bypass cement waste dust [23], and coal fly ash (CFA) [24] have been shown as suitable precursors due to their relatively high amorphous aluminosilicate content . CFA has a similar management profile to PG in the Philippines where large amounts of this waste are generated, but the bulk is disposed of in special landfills [25, 26]. Both are considered hazardous wastes because they contain easily soluble heavy metals, toxic metalloids, and radioactive elements [27, 28].

Several studies have explored the synergy of CFA and PG in geopolymer synthesis [29]. Compressive strength (CS) of the geopolymer product is a measure of its performance. Bebana et al. [30] optimized and predicted the CS of an alkali-activated FAPG at different CFA/PG compositions, activator concentration, temperature, and curing time using artificial neural networks (ANN). The optimal CS determined experimentally and by the ANN equals roughly 8.9 MPa at 10% PG, 28-day curing time, and 60 °C curing temperature. In the study of Pratap et al. [31], a maximum strength of 47.97 MPa was observed for FAPG concrete using 30% PG and 12 M NaOH. It has been noted in this study that partial substitution of PG instead of pure CFA decreased the leachability of heavy metals and its pH to almost neutral. The observation supports the heavy metal immobilization effect of FAPG. The addition of PG also induced a stabilizing and reinforcing effect to FAPG by lowering its water absorption, porosity, and sulfate attack [32]. Other studies attempted to synthesize ternary blends of CFA/PG geopolymer composites with other industrial wastes. Red mud and ground blast furnace slag had been used in the study of Liu et al. [33] and Zhang et al. [34], respectively. These studies demonstrated that a replacement rate of 10-20% PG for the third component is the optimal range. Within the range, PG reduced efflorescence and increased CS.

Research dealing with FAPG so far has dealt with the alkali-based activator with minimal attention to acid-based activator. CFA has been used extensively in acid activated systems using PA. As for PG itself, it has only been found to be studied in two articles using acid geopolymer technology. Majdoubi et al. [35] explored the synergy of PG with metakaolin (MK) via PA activation. The CS could reach as high as 48.43 MPa at 70% PG of the matrix. This observation was attributed to the limiting effect of PG to microcrack propagation and to the increased density of the material. PG was not observed to induce geopolymerization, but it demonstrated firm adhesion to the matrix. However, PG tends to agglomerate at higher weight fractions. The other study investigated PG with red clay instead [36]. The same observations from the previous study were noted for this study except that the CS is lower (21.46 MPa) and the optimal PG replacement is 28.83%. The difference could be attributed to the higher Al content of MK thus leading to a more established geopolymer networks. These studies demonstrated that acid-based activators such as PA have been emerging for use in the geopolymer landscape [37, 38]. This could be beneficial for a sustainable economy as PG and PA are simultaneously produced in phosphoric acid plants.

The abovementioned literature expressed the positive effects of PG in geopolymer performance. However, the environmental effects of employing these processes are limited. Leaching tests and theoretical carbon accounting had loosely been made to mean the same thing as environmental impact. Apart from the current practice of handling and disposing of CFA and PG, the production of their geopolymer composite entails its own environmental impacts. To address the issue, the life cycle assessment (LCA) is a methodology encompassing life cycle thinking capable of quantifying these effects. LCA is an analytical method defined by the International Organization for Standardization

(ISO). It aims to assess the environmental effects brought about by a process or a product from the acquisition of raw materials, processing, and production until end-of-life use. An LCA approach is beneficial since it can dissect the environmental effects of a certain production line and propose solutions depending on the resulting adversity or benefit in various criteria [39, 40]. Due to the lack of relevant information regarding commercial applications of FAPG, the usage of ex-ante LCA can prove advantageous because of the economic and environmental impacts that might entail from the application of geopolymer technology [41]. To the best of the knowledge of the authors, this is the first work about the consequential life cycle assessment of PG-CFA geopolymer composite synthesis routes. While several studies have explored geopolymer synthesis using PG and CFA, systematic evaluation of environmental impacts across different activation routes remains limited.

Assessment of the impacts thereof mainly focus on the reduction in carbon dioxide equivalent (CO_2-eq) emissions as in the compilation of LCA studies elaborated in Sbahieh et al. [42]. The preferential scoping in this specific environmental criterion is largely due to the perception of geopolymer products being direct substitute to traditional concrete. Most studies only evaluated single synthesis routes that highlights the property of the final material product. Previous life cycle assessments have primarily focused on alkali-activated geopolymers, with minimal attention to emerging acid-activation approaches [43, 44]. Thus, the research gap is the lack of the comparison of environmental impacts of different activation routes of FAPG.

This study evaluates the life cycle environmental impacts of utilizing PG as a precursor material for the synthesis of FAPG. The current and baseline disposal method of PG and CFA poses threats to the population and ecosystem. The paper intends to focus on the environmental impact of PG as the waste byproduct, which has been studied less intensively than CFA. The integration of LCA in scrutinizing the sustainability of the proposed geopolymer synthesis method anticipates the ex-ante impact of FAPG which can pave the way for setting local industrial standards for geopolymers. Repurposing of these wastes could potentially address and provide long-term relief via FAPG synthesis. The study proposes a process integrating FAPG synthesis using acid- and alkali-based activators. The main objective is to assess the life cycle of PG as a filler material for coal fly-ash based geopolymer evaluated using IMPACT World+ midpoint impact assessment method using 1 kg of PG as the functional unit. The study aims to determine whether upcycling PG in geopolymer composites provides environmental benefit compared to traditional landfilling. The technical and environmental performance of FAPG was related in this study. Through LCA, the consequential environmental impacts of the baseline scenario and the proposed process can be quantified. The results of the LCA determine whether the integration of geopolymer synthesis process can alleviate the environmental burdens brought upon by landfilling PG and CFA.

In this paper, Section 1 discussed the existing literature and the research gap on FAPG. Section 2 elaborated on the methodology of synthesizing acid- and alkali-based FAPG to be evaluated by LCA and sensitivity analysis. Section 3 displayed the characteristics and properties of the geopolymer product; interpretation of its environmental impacts was conducted via midpoint impact assessment. Section 4 discussed the concluding remarks and future direction for this research.

2. Materials and Methods

This section elaborates the method of synthesis of the acid- and alkali-based FAPG specimens. The technical-environmental performance of FAPG was evaluated by varying the ratio of FAPG components. Then, LCA was conducted using representative specimens from both routes which yielded the highest unconfined compressive strength (UCS).

2.1. Synthesis of Coal Fly Ash-phosphogypsum Geopolymer

Raw PG and raw CFA were combined at different precursor (P) proportions mixed with an acid or alkali (A) activator for the synthesis of FAPG. The methodology of Kalaw et al. [45] was implemented from raw material handling up to the ambient curing of the FAPG. Ranges for the CFA/PG proportion and A/P ratio utilized in the study were adopted Majdoubi et al. [35] and Longos et al. [46] for the acid and alkali-based FAPG, respectively. The former study explored the synergistic effects of PG-metakaolin (MK) precursor mix with PA as an acid activator while the latter investigated CFA-nickel laterite mine waste (NMW) with alkali activator. The other raw materials in these referenced studies were substituted with appropriate material that is of interest in this paper. Tables 1 and 2 summarize the formulation blends conducted on a laboratory scale using central composite design (CCD) of experiment.

Table 1. Values of factors and levels for alkali-based geopolymer synthesis

Alkali FAPG Factors	Levels				
Precursor proportion (PG/CFA ratio)	4/96	8/92	12/88	16/84	20/80
Activator to precursor (A/P ratio)	0.8	0.9	1.0	1.1	1.2

Table 2. Values of factors and levels for acid-based geopolymer synthesis

Acid FAPG Factors	Levels				
Precursor proportion (PG/CFA ratio)	10/90	20/80	30/70	40/60	50/50
Activator to precursor (A/P ratio)	0.8	0.9	1.0	1.1	1.2

2.2. Product Characterization and Property Evaluation

The cured FAPG specimens were prepared and subjected to UCS tests in accordance with ASTM C109M using 50 mm cubes [47]. Consequently, the raw materials, PG and CFA, were analyzed via X-ray fluorescence spectroscopy (XRF) using HORIBA Scientific (China) and scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS) via JSM-IT500HR InTouchScope (Germany) to confirm the applicability of the raw materials for geopolymer synthesis. The synthesized geopolymer specimens with highest UCS were characterized via Fourier transform infrared (FTIR) spectroscopy using Shimadzu IRSpirit FTIR (Japan) and scanning electron microscopy (SEM) to evaluate whether geopolymerization took place [35, 45, 46].

2.3. Life Cycle Assessment

Life cycle assessment was conducted following the ISO 14040:2006 and 14044:2006, which includes four integral phases: (1) goal and scope definition, (2) life cycle inventory analysis, (3) impact assessment, and (4) interpretation [48, 49]. The approach enables a holistic overview of the environmental sustainability of a certain process.

Goal and scope definition allows the user to outline the system to be evaluated. In accordance with ISO 14040/44, goal definition establishes the intended application, target audience, and reasons for conducting the study, while scope definition is a process of selecting relevant input or output flows and impact categories to ensure methodological consistency. Through this, the system boundary is defined. A functional unit is also determined at this stage. It will be utilized as the basis of calculations of environmental impacts in LCA.

Inventory analysis involves data gathering for input and output flows within the system boundary. The equation governing the transformation of system inputs (raw materials) to its corresponding output (final product) is defined as follows:

$$As = f \quad (1)$$

where A is the technology matrix, s is the scaling vector, and f is the final product matrix. The technology matrix corresponds to the amount of material or number of services in the system. The scaling vector is a multiplier to the technology matrix to obtain the amount of the final product required. The f matrix represents the economic flows which are streams that have economic equivalent or value.

The environmental flows are determined based on the final product matrix. These are classified as the emissions and wastes produced by the system. Using the same scaling vector, this can be defined as:

$$Bs = g \quad (2)$$

where B is the intervention matrix and g is the inventory matrix. The intervention matrix comprises the environmental flows per technology in the system. The inventory matrix provides the final set of environmental flows in relation to the functional unit; it is subjected for life cycle impact assessment.

In life cycle impact assessment, the environmental flows are characterized under different impact categories. Common environmental impact categories such as global warming potential, eutrophication, mineral resources depletion, water scarcity, ozone layer depletion, etc. are quantified as in:

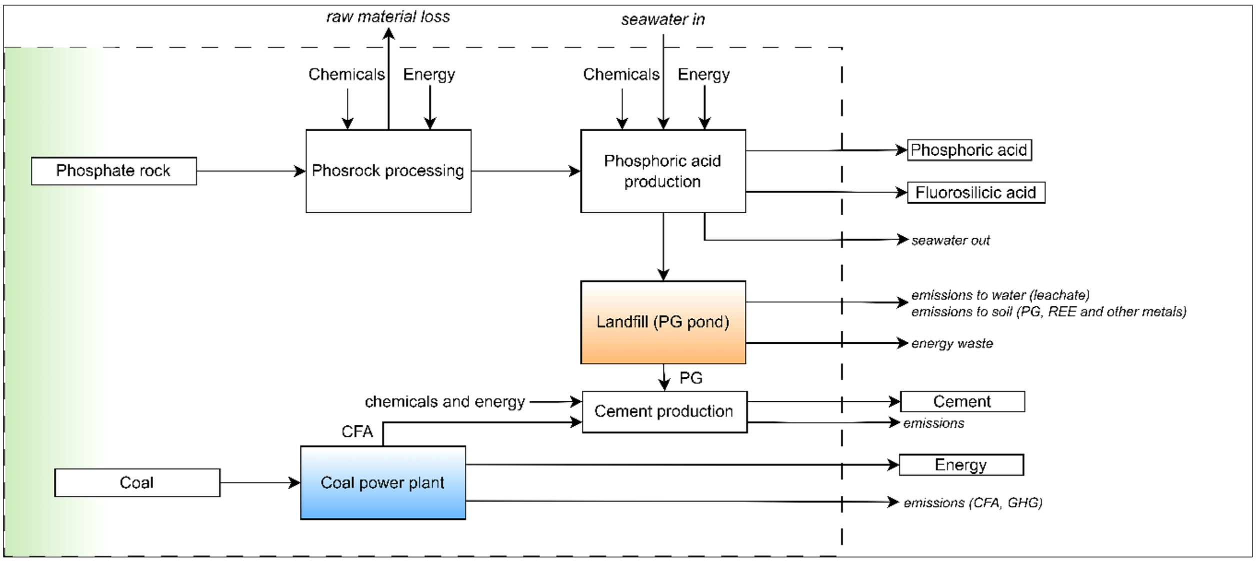
$$Qg = h \quad (3)$$

where Q is the characterization matrix and h is the impact vector. The characterization matrix is unique per impact categories. The impact vector describes the environmental impact of the system. The transformation of environmental flows to environmental impact allows the aggregation of emissions with similar environmental effects to be normalized. For instance, global warming potential is based on the impact of 1 unit mass of CO_2. Methane has a global warming potential equal to 28 times that of CO_2. Thus, 1 kg methane is equivalent to 28 kg CO_2 equivalent.

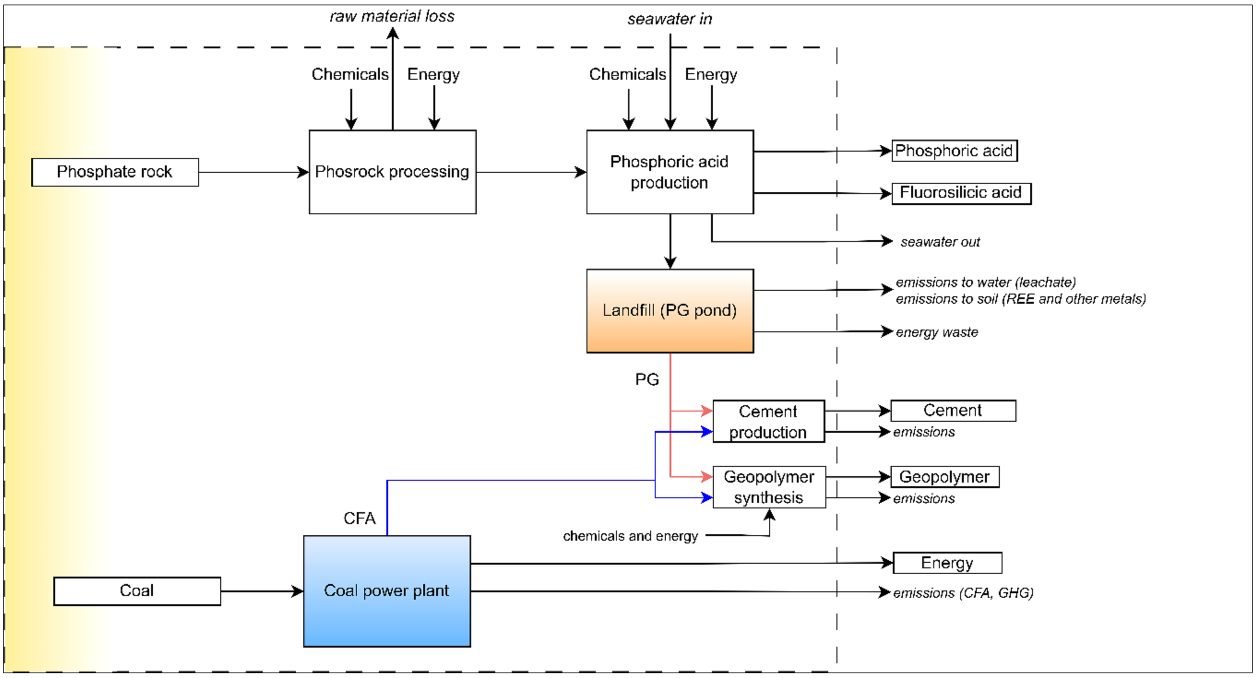
2.3.1. Goal and Scope Definition

The goal and scope of study commences at the production of CFA and PG from a coal power plant and phosphoric acid production plant, respectively. The study aims to comparatively evaluate the environmental impacts of applying geopolymer technology to repurpose PG. The goal of conducting LCA is to determine if upcycling PG as a geopolymer precursor contributes to less environmental impact than landfilling. Hence, 1 kg of PG was taken as the functional unit of LCA. The system boundary starts with the extraction of raw materials—coal and phosphate ore. Other reagents necessary for the entire production line were also considered.

The study involved the comparison of a baseline scenario or the "business as usual" case against two dynamic scenarios where FAPG was synthesized via an acid- and an alkali-based activator. Boundaries for all the scenarios include an PA plant (PAP), a coal-fired power plant (CFPP), and a cement production plant (CPP). Figure 1-a demonstrates the baseline scenario where in the PA plant, phosphate rock is processed via the wet dihydrate process using concentrated sulfuric acid producing PA and fluorosilicic acid (H_2SiF_6). PG is generated as a byproduct generated in large quantities, 30% of which is recycled as a cement additive, then the rest is dumped in stacks called ponds. Additionally, in another process, sub-bituminous coal is consumed in the CFPP generating electricity alongside CFA as the byproduct and other gaseous emissions typical to that of burning coal. As with PG, coal is also being recycled for cement production while the unutilized CFA is dumped in an open landfilling. Figure 1-b shows the alternative scenario where CFA and PG were further repurposed to FAPG. It is assumed that the unutilized solid waste dumped in the previous scenario were utilized for the FAPG synthesis. Two synthesis scenarios were analyzed: acid-based as Scenario 2, and alkali-based geopolymerization as Scenario 3.



(a) Scenario 1 (baseline)



(b) Scenarios 2 and 3 (FAPG synthesis)

Figure 1. Simplified system boundary for the LCA

2.3.2. Life Cycle Inventory Analysis

Life cycle inventory (LCI) of the production line of the scenarios was performed using data from laboratory-scale experiments, literature, government reports, and databases. Typical and background environmental and economic flows were referenced from ecoinvent 3 and Agri-Footprint as available in SimaPro v9.6 [50, 51]. The summarized LCI is shown in Table 3. Negative values indicate process inputs while positive values are process outputs. The inventory for the PAP was referenced from common industrial practice via the wet dihydrate process [52]. Raw material phosphate-bearing rock corresponds to 0.267 kg per 1 kg PG produced. Concentration values used were 98.5% H_2SO_4, 79% merchant-grade PA, and 20% H_2SiF_6. Per 1 kg PG functional unit, 0.086 kg 79% PA and 0.01 kg 20% H_2SiF_6 are produced while consuming 0.2665 kg phosphate rock. Electricity was utilized at a rate of 0.1638 kWh/kg PA produced in which the concentration of the acid accounts for most of the energy use. 11.38 kg seawater was also used as a coolant and used to form slurry with phosphogypsum as it was discharged to the ponds.

Table 3. Summary of life cycle inventory for three scenarios

Functional unit (1 kg PG)		S1*			S2*	S3*
		PAP	CFPP	CPP	FAPG	FAPG
Materials/fuels						
Phosphate rock	kg	-0.27	-	-		-
Coal	kg	-	-52.20	-		-
Sulfuric acid (98.5% w)	kg	-0.15	-	-		-
Phosphoric acid (56% P2O5)	kg	0.09	-	-	-3.10	-
Fluorosilicic acid (20% w)	kg	0.01	-	-		-
Cement	kg	-	-	7.50		-
Sodium hydroxide (10 M)	kg	-	-	-		-2.81
Sodium silicate (48% w)	kg	-	-	-		-6.99
FAPG	kg	-	-	-	6.65	31.5
Water	kg	-	-	-	-0.75	-4.91
Avoided products						
Coal fly ash	kg	-	7.31	-1.69	-2.8	-16.8
Electricity						
Electricity mix, PH	kWh	-0.17	365.03	-3.67	0.00	0.00
Emissions to air						
Carbon dioxide	kg	0.0809	106.8	12.7	n/a	n/a
Carbon monoxide	kg	n/a	13.59	n/a	n/a	n/a
Sulfur dioxide	kg	n/a	5.12	n/a	n/a	n/a
Waste to treatment						
CFA to landfilling	kg	-	5.63	-	avoided	
PG to landfilling	kg	0.7	-	-		

* *Note: SX refers to Scenario X where X = 1, 2, and 3.*

Localization of the electricity mix was based on the official report of the Philippine Department of Energy (DOE). The flows and emissions for CFPP in all scenarios were taken from the DOE data representative of the coal-based power plants in Region X of the Philippines [53]. The Philippine sub-bituminous coal used in LCA was represented by a coal with a calorific value of 24.8 MJ/kg [50]. Proximate analysis of coal was assumed to be 51% fixed carbon (FC), 26% volatile combustible matter (VCM), 5% moisture, and importantly, 14% ash which became the CFA in the system. There was a lack of local data availability for the process of cement production thus, global-based inventory was applied. Synthesis of FAPG in Scenarios 2 and 3 was conducted on a laboratory-scale using the formulation as reported in the studies of Majdoubi et al. [35] using PA (Scenario 2) and Longos et al. [46] using NaOH and Na_2SiO_3 (Scenario 3). Values for CFA/PG proportion in the precursor and activator to precursor (A/P) ratio employed for the scenarios are based on the equal highest UCS yielded on a 28-day curing time basis for fair comparison of their performance. Summary of the pertinent ratios and values used for these parameters and the corresponding reagents for the scenario as modelled in SimaPro is in Table 4. Current local practices for the disposal strategies for the PG and CFA were implemented in the study for Scenario 1. In the alternative scenarios, PG and CFA were repurposed, thus included as avoided products.

Table 4. FAPG parameters modelled for Scenario 2 and Scenario 3

Parameter	S2 (acid-based FAPG)	S3 (alkali-based FAPG)
Activator (concentration)	H_3PO_4 (aq., 10M)	NaOH (aq., 10M) Na_2SiO_3 (aq., 2.5:1 wt. with NaOH)
CFA/PG proportion	80/20	96/4
A/P ratio	0.9	0.8
Compressive strength	7.9±0.18 MPa	

2.3.3. Life Cycle Impact Assessment

Impact assessment method used was the IMPACT World+ Midpoint v1.04 in SimaPro v.9.6. Bulle et al. [54] developed a midpoint-damage framework that comprehensively and efficiently characterizes the consequences of the environmental flows in the system. These indicators are climate change (short-term and long-term), fossil and nuclear energy use, mineral and resources use, photochemical oxidant formation, ozone layer depletion, freshwater ecotoxicity, human toxicity cancer, human toxicity non-cancer, freshwater acidification, terrestrial acidification, freshwater eutrophication, marine eutrophication, particulate matter formation, ionizing radiation, land transformation, land occupation, and water scarcity. Midpoint impact analyses allow the contributions to be highlighted of the environmental and economic flows in a certain impact category [39]. Through this, the scenarios can be optimized as the key contributors were specifically identified supported further by process contribution analysis.

2.3.4. Sensitivity Analysis

Effects of the LCA input parameters on midpoint impacts were evaluated using sensitivity analysis. Design of experiments (DoE) was utilized to describe the response of the midpoint impact characterization from the formulated regression equations relating to the factors [55]. Three factors: the percentage allocation of PG to geopolymer synthesis (A), CFA/PG ratio in FAPG precursor (B), and A/P ratio in FAPG (C), with three levels, were analyzed. In the current scenario, 30% of the waste PG is transported as a substitute to gypsum in cement manufacturing. CFA/PG ratio in FAPG and A/P ratio were also varied as these are the experimental parameters varied; industrial standards for geopolymer blend are still unavailable to date. The response variables were the environmental indicators as discussed in the previous section. Table 5 shows the Box-Behnken DoE factors, levels, and responses for sensitivity analysis.

Table 5. Summary of life cycle inventory for three scenarios

Factors	Levels			Responses
PG allocation (**A**)	0.3	0.4	0.5	IMPACT World+ Midpoint Impact Indicators for Scenarios 2 and 3
Precursor proportion (as CFA fraction, **B**)	0.80	0.85	0.90	
Activator to precursor (A/P ratio, **C**)	0.8	0.9	1.0	

3. Results and Discussion

3.1. Characteristics and Properties of PG and CFA

PG and CFA were analyzed by XRF, FTIR, XRD, and SEM-EDS to determine their elemental and mineralogical composition, and morphology. Tables 6 and 7 show the XRF and SEM-EDS results reported in percentage weight composition, respectively for both PG and CFA. PG is mainly composed of calcium (Ca) (as CaO) and sulfur (S) (as SO3) with small quantities of silicon Si (as SiO2) via XRF. These results agreed with the observation of different studies compiled by Rashad [56]. Elemental analysis using SEM-EDS also confirmed the elemental composition of PG where Ca and S dominated the PG composition. CFA is composed mainly of SiO_2, Al_2O_3, CaO, and Fe_2O_3. The ratio in which these oxides are present classifies the CFA used in the study as Class C according to ASTM C618 [57]. Pratap et al. [29] employed CFA with the same constituents for alkali geopolymerization. High silicon and aluminum content is crucial in the precursor to facilitate the formation of Si-Al-O polymer chains which are exhibited in the XRF and EDS analyses of CFA.

Table 6. XRF analyses of PG and CFA

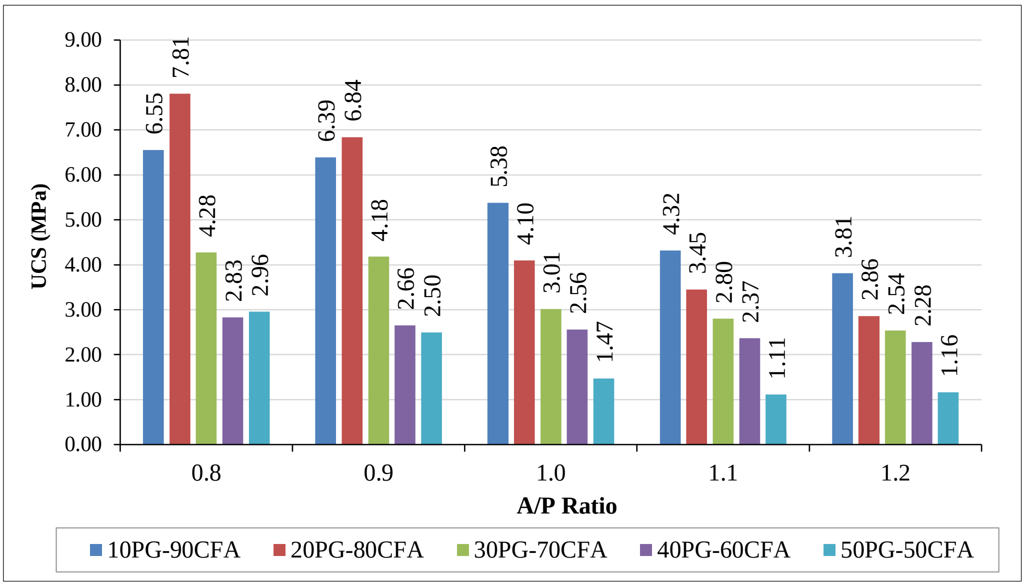
Composition	SiO_2	Al_2O_3	CaO	SO_3	Fe_2O_3	K_2O	SrO	Rh_2O_3	Rb_2O
PG	1.44	-	62.02	35.51	0.4	-	0.29	0.3	-
CFA	27.03	12.07	25.38	7.7	24	1.29	0.19	0.29	0.01
Composition	Y_2O_3	ZrO_2	TiO_2	MnO	BaO	As_2O_3	CuO	NiO	
PG	0.01	0.003	-	-	-	-	0.02	-	
CFA	0.02	0.07	1.08	0.37	0.34	0.01	0.03	0.03	

Table 7. SEM-EDS analyses of PG and CFA

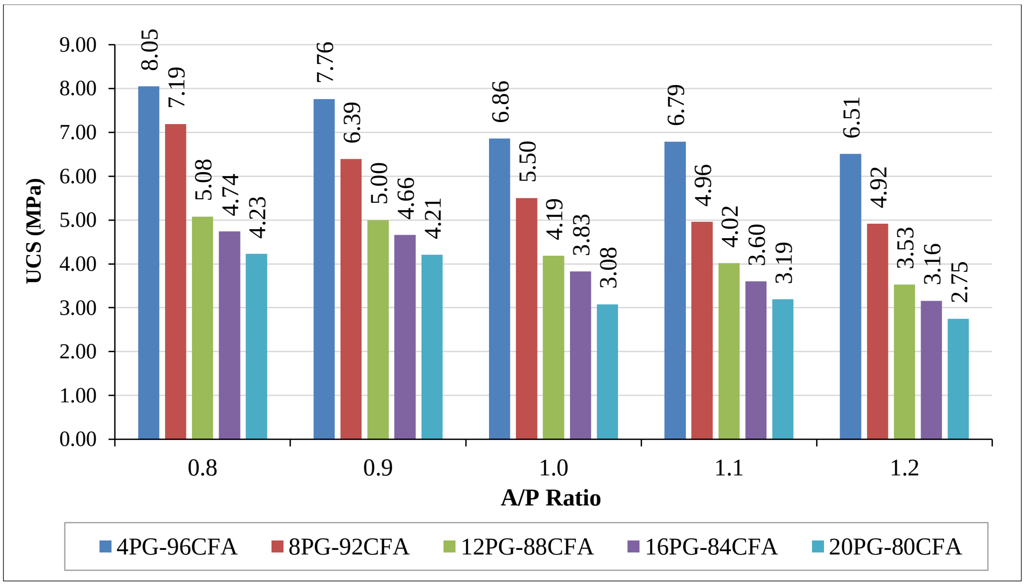
Element	O	Si	Al	Ca	S	Fe	C	Zr	Mg	Ti	K
PG	48.41	0.52	-	20.25	15.83	-	11.22	3.78	-	-	-
CFA	45.3	14.38	6.77	6.77	2.06	5.84	17.24	0.83	1.13	0.32	0.59

3.2. Characteristics and Properties of FAPG

UCS tests were conducted for the acid- and alkali-based FAPG. Figures 2-a and 2-b summarize the CS of the specimens in MPa. The UCS results ranged from 1.11 to 7.81MPa for acid FAPG and from 2.75 to 8.05MPa for alkali FAPG. A precursor proportion of 10PG-90CFA and an A/P ratio of 0.8 yielded the highest UCS (7.81 MPa) for the acid FAPG, while 4PG-96CFA and A/P ratio of 0.8 resulted in the highest UCS (8.05 MPa) for alkali FAPG. PG was discovered to demonstrate a reinforcing effect that limited the propagation of microcracks hence, increasing the UCS of the geopolymer [35]. The increase in the A/P ratio in both synthesis routes negatively affected the UCS. The decline in performance may be attributed to the increase in unreacted water as A/P ratio increases, thus leading to a more porous structure [58]. Interestingly, a decrease in UCS was observed starting from 20% PG onwards for A/P ratios of 0.8 and 0.9 due to PG agglomeration [59]. The trend is consistent for all A/P ratios on the alkali-synthesized FAPG. The formation of Si-Al-O chains was prominent based on the wavelength bands and the surface morphology as analyzed via FTIR and SEM.



(a) Acid FAPG



(b) Alkali FAPG

Figure 2. UCS tests results of FAPG samples after 28 days of curing

3.3. Life Cycle Impact Assessment

The evaluated midpoint damage indicators are demonstrated in Table 8. Scenario 2 had the highest environmental benefits in climate change, fossil and nuclear energy use, photochemical oxidant formation, freshwater ecotoxicity, and human toxicity non-cancer. For mineral resources use and particulate matter formation, Scenarios 2 and 3 performed almost similarly, having the least burden. Scenario 3 was only beneficial in the other three midpoint levels: water scarcity, freshwater, and terrestrial acidification. Scenario 1 performed better than the alternatives in terms of ozone layer depletion, human toxicity cancer, eutrophication of water bodies, ionizing radiation, and land use.

Table 8. Characterization of environmental impact assessment at midpoint level

Midpoint level impact indicators	Unit	Scenario 1	Scenario 2	Scenario 3
Climate change, short term	kg CO_2 eq	20.01	12.12	19.72
Climate change, long term	kg CO_2 eq	19.70	11.63	18.74
Fossil and nuclear energy use	10^{-3} MJ deprived	1.12	10.5	17.9
Mineral resources use	kg deprived	1.92	1.24	1.42
Photochemical oxidant formation	kg NMVOC eq	4.11	0.05	0.06
Ozone layer depletion	10^{8} kg CFC-11 eq	2.88	5.60	45.7
Freshwater ecotoxicity	10^{-4} CTUe	103	9.43	28.2
Human toxicity cancer	10^{7} CTUh	5.61	13.2	32.7
Human toxicity non-cancer	10^{6} CTUh	9.65	1.42	3.23
Freshwater acidification	10^{7} kg SO_2 eq	23.1	3.84	1.96
Terrestrial acidification	10^{3} kg SO_2 eq	3.16	0.30	0.16
Freshwater eutrophication	10^{5} kg PO_4 eq	1.06	20.5	15.4
Marine eutrophication	10^{4} kg N eq	4.41	7.90	15.4
Particulate matter formation	kg $PM_{2.5}$ eq	0.23	0.01	0.01
Ionizing radiation	Bq C-14 eq	4.12	23.87	53.65
Land transformation, biodiversity	10^{4} m^2yr arable	2.78	7.43	14.3
Land occupation, biodiversity	m^2yr arable	0.05	0.16	0.34
Water scarcity	m^3 world eq	337.76	11.21	5.68

Meanwhile, Scenario 1 dominated as the most detrimental to the environment, being the worst scenario in 11 categories: climate change (short- and long-term), fossil and nuclear energy use, mineral resources use, photochemical oxidant formation, freshwater ecotoxicity, human toxicity non-cancer, freshwater and terrestrial acidification, particulate matter formation, and water scarcity. These offsets the benefits brought upon by the baseline in other damage characterization. The rest of the midpoint-level impacts resulted in Scenario 3 being the worst alternative, except for freshwater eutrophication, where Scenario 2 had the most environmental burden. These environmental burdens can be attributed to the additional chemicals employed and the energy required for FAPG synthesis. Improvements in the process and formulation of FAPG are imperative to alleviate the aforementioned effects.

Climate change (long term), fossil and nuclear energy use, mineral and resources use, ionizing radiation, and water scarcity are the criteria of concern (CoC) which are categories directly affected by the alternative scenarios especially. Figure 3 demonstrates the impact characterization values per 1 kg of PG for each scenario in these categories. Discussion around the long-term effects of climate change is crucial as the other impact categories such as acidification, photochemical oxidant formation, ozone layer depletion, and ecotoxicity contribute to the overall planet health wherein climate change (long-term) as a midpoint level impact indicator aims to measure. Energy consumption, resource extraction, and water availability are directly tied to process sustainability. These criteria hold relevance in the discussion due to the heavy involvement of mining operations in PA production and CFPP. The ionizing radiation category is also of key importance since PG is a NORM solid waste.

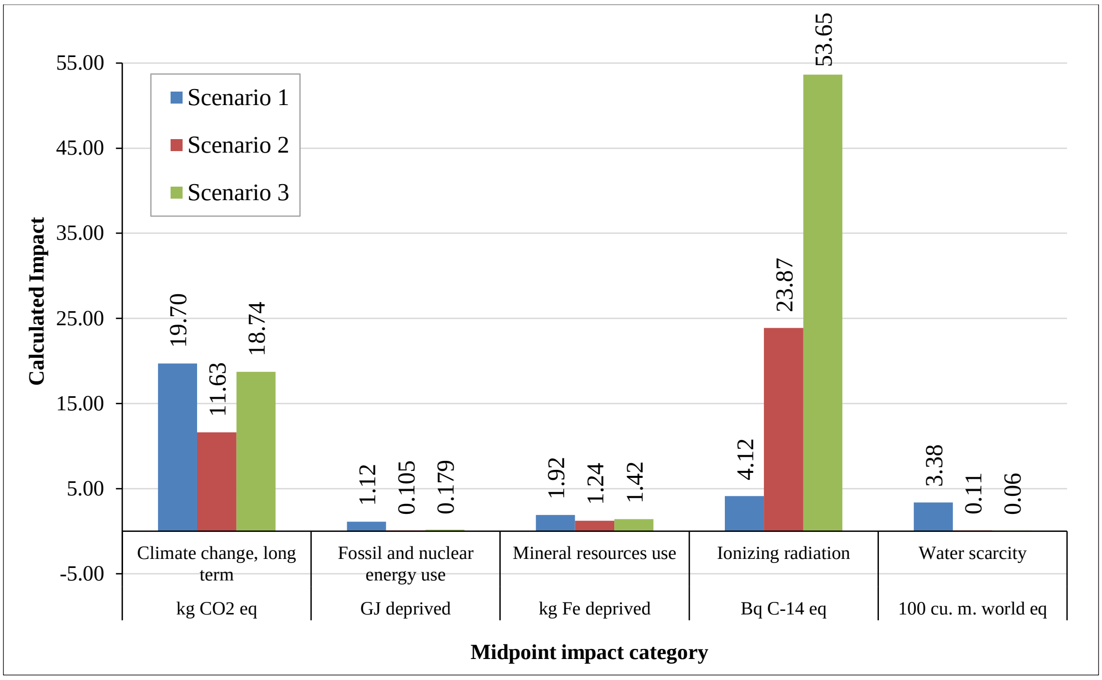


Figure 3. Summary of impact characterization per scenario for criteria of concern

The normalization of midpoint-level impacts is shown in Figure 4, wherein the relative impacts of the scenarios are demonstrated. The highest positive percentage (100%) corresponds to the highest relative environmental burden, while the least positive percentage (-100%) translates to the highest relative environmental benefit. For every 1 kg of PG within the system as the functional unit, Scenario 2 offered improvements compared to Scenario 1 in terms of less CO_2-eq emissions by 40%, less than 35% of mineral resources deprived, and energy resources savings of around 91%. Moreover, the acid FAPG route proved to have minimal impact on photochemical oxidant formation, freshwater ecotoxicity, and non-cancer toxicity relatively by around 90%. Both Scenarios 2 and 3 fared better against the baseline in PM2.5 emissions due to the reduction in PG and CFA landfilled. Due to the use of acid in Scenario 2, Scenario 3 is the more environmentally friendly option in the acidification criteria. The midpoint impacts were lessened as these were highly attributed to the replacement of gypsum and sand with PG and CFA, which are otherwise obtained via quarrying whose operations have corresponding global warming contributions and resource extraction [60].

Despite the beneficial effects of Scenarios 2 and 3, the baseline scenario proved to have less environmental burden under land use, eutrophication, ozone layer depletion, human toxicity cancer, and ionizing radiation. Additional land would be required for the industrial-scale production of FAPG. Using a phosphate-based acid directly contributes to the eutrophication of freshwater and marine water bodies of Scenario 2 [61]. Scenario 3 is the least desirable alternative in most of these aspects. Moreover, uranium and its decay products, which is found to be present in phosphate-bearing ore and PG, is leachable in acidic and basic conditions. Weiksnar et al. [62] observed increased leachability of Ra-226 as the pH increases due to lime rock from a minimum of 6-8. Fu et al. [63] demonstrated the increased dissolution of uranium from iron oxide-copper-gold, another type of NORM, in H_2SO_4 solution. The relative Bq C-14 of Scenario 2 and 3 is particularly attributed to the fact that some radioactive components were carried over to the FAPG synthesis [64]. Despite this, the reported average range of Bq Ra-226 per kg PG in the Philippines 91.5-935. The midpoint level LCIA results suggest that though Scenario 2 had the most environmental with the least number of burdens, some aspects of the process need to be improved, such as the use of PA being the cause of the acidification and eutrophication of water bodies.

The contribution of the major processes per scenarios are shown in Figures 5-a to 5-c each corresponding to the baseline, acid-based FAPG, and alkali-based FAPG, respectively. It is evident from the baseline scenario that no environmental credits could be gained from landfilling of the industrial wastes. CPP and CFPP dominate the impact contribution due to their high CO2-eq emitting activity, energy and raw material intensive processes. As for S2 and S3, FAPG synthesis for both scenarios contributed to having adverse effects. The negative impacts of the baseline via PG and CFA reuse were diminished but insufficient in achieving a net zero circular process. Similarly, CPP and CFPP contribute a large chunk to the cradle-to-grave life cycle of CFA and PG. Such outcome is expected since variations were not employed for these operations.

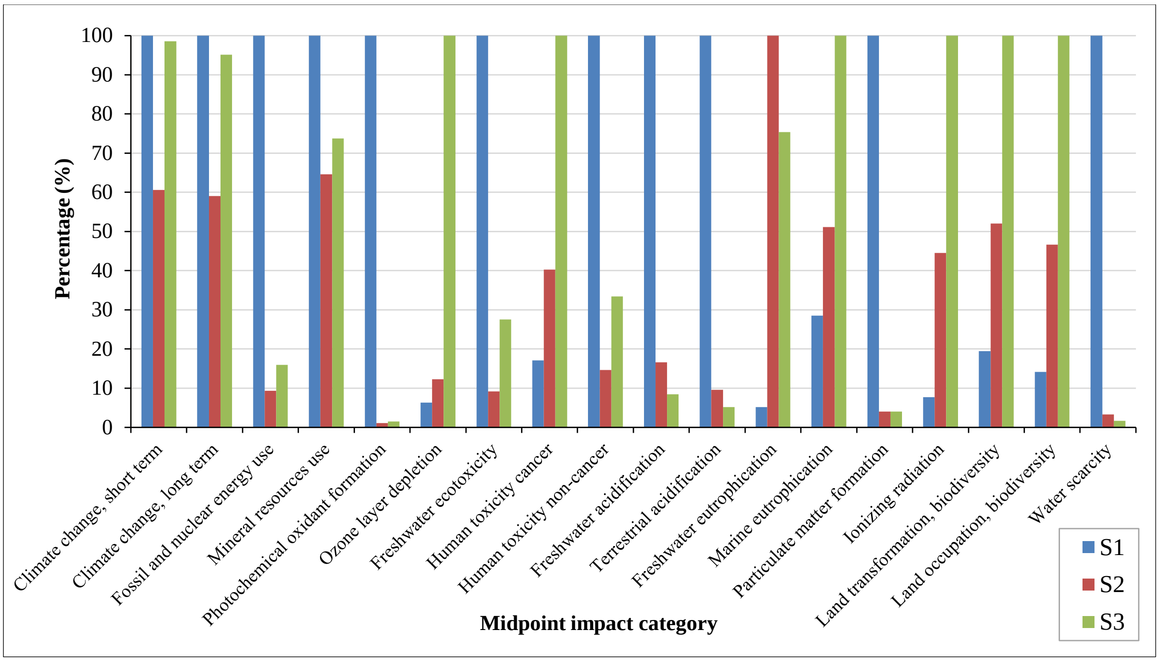
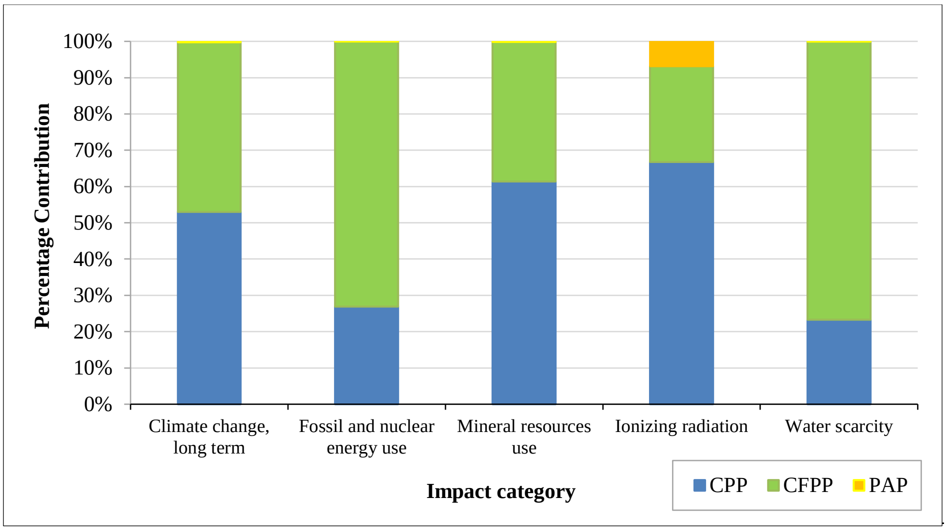
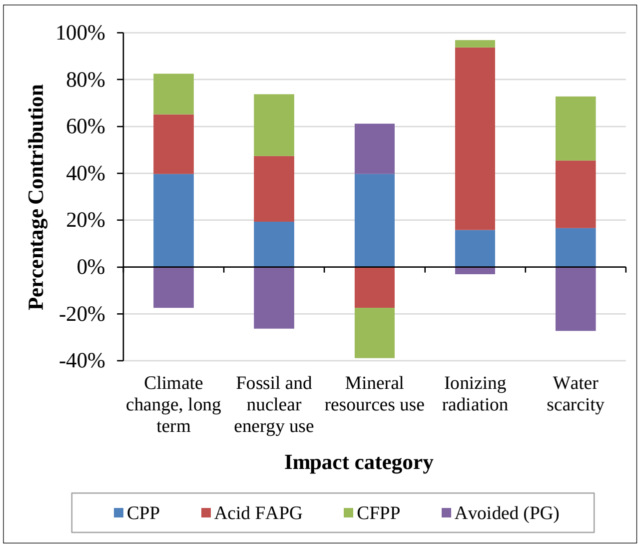
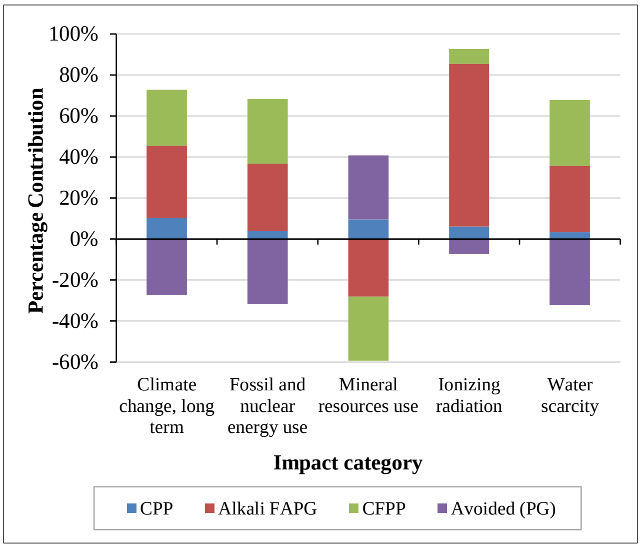


Figure 4. Normalized midpoint impact characterization for Scenarios 1-3



(a) S1





(b) S2 **(c) S3**

Figure 5. Contribution analysis of the LCA scenarios

3.4. Sensitivity Analysis

Sensitivity analysis allows a more robust and comprehensive LCA wherein the input parameters impact the response of the midpoint level impacts using a mathematical model [65]. Particularly, the sensitivity analysis is conducted to determine the effects of variation in the percentage allocation of PG (A) which correlates directly to the distribution of PG to landfilling, CFA/PG precursor ratio (B), and A/P ratio (C) to the impact characterization of Scenarios 2 and 3. The summary of the response coefficients are summarized in Tables 9 and 10. The discussion will focus on the CoCs elaborated in Section 3.3. ANOVA was used to study the relationship between the factors and midpoint impact responses where quadratic relationships were exhibited. The resulting quadratic regression equations are in the form shown in Equation 4:

$$\text{Midpoint impact} = k + A + B + C + AB + BC + B^2 \quad (4)$$

Table 9. Results of sensitivity analysis for Scenario 2

Midpoint level impact characterization	Unit	*k*	A	B	C	AB	BC	B^2
Climate change, long term	kg CO_2 eq	15.0	2.58	1.04	0.33	-0.17	0.12	0.35
Fossil and nuclear energy use	MJ deprived	133	16.9	14.5	4.55	-2.41	1.61	4.82
Mineral resources use	kg deprived	1.64	0.37	0.04	0.01	-0.01	0.00	0.01
Ionizing radiation	Bq C-14 eq	28.0	-2.86	8.65	2.72	-1.44	0.96	2.88
Water scarcity	m^3 world eq	13.0	-1.74	4.38	1.38	-0.73	0.49	1.46

Table 10. Results of sensitivity analysis for Scenario 3

Midpoint level impact characterization	Unit	*k*	A	B	C	AB	BC	B^2
Climate change, long term	kg CO_2 eq	14.7	2.63	0.92	0.29	-0.15	0.10	0.31
Fossil and nuclear energy use	MJ deprived	123	18.8	10.4	3.27	-1.73	1.15	3.46
Mineral resources use	kg deprived	1.61	0.37	0.03	0.01	0.00	0.00	0.01
Ionizing radiation	Bq C-14 eq	17.8	-1.05	4.82	1.52	-0.80	0.54	1.61
Water scarcity	m^3 world eq	2.52	0.12	0.45	0.14	-0.08	0.05	0.15

For both Scenarios 2 and 3, the interaction between A and C was statistically insignificant ($p > 0.05$). CC, FNEU, and MRU have positive coefficients for the linear variables A, B, and C and the quadratic variables BC and B2. A negative relationship was established for the interaction between A and B. In IR and WS, similar observations hold true except for A, where a negative relationship was determined. Sensitivity of Scenario 2 is much greater than Scenario 3 as seen in the numerical coefficients.

Based on the sensitivity analysis, CC, FNEU, and MRU environmental indicators correspond to more adverse effects by the increased landfilling of PG represented by A while the opposite holds true for IR and WS criteria. Notably, the interaction between A and B variables result in the reduction of the negative effects. The AB variable does not offset the variations of A especially in CC, MRU, and FNEU. Furthermore, the single parameter C which denotes the A/P ratio has the least impact on the midpoint impacts unlike A and B. Generally, the sensitivity analysis suggests the imperativeness of avoiding the landfilling of PG but not upcycling a large portion of PG stockpile for geopolymer synthesis. While the proposed process has reduced the environmental effects of traditional PG management based on the synthesis experiments, discretion must still be practiced in the allocation of PG before it can offset the environmental credits gained.

Uncertainty is present in the data set due to limited primary source for the energy mix. The selected factors are subject to uncertainty, especially with the allocation of PG. Sensitivity analysis should suffice for this study in quantification of uncertainty in industrial inventory which is the case for the CPP, PA, CFPP, and FAPG synthesis [66]. Regional and temporal variability of the background data causes uncertainty to arise from the results of midpoint impact. Ecoinvent is one of the databases used in the LCA study which had already accounted for data inaccuracy via Pedigree matrix approach. A separate uncertainty analysis for the systems presented might only lead to redundancy.

3.5. Interpretation

The environmental impacts of the current and proposed alternative for handling PG and CFA in the context of the local Philippine scenario were quantified. Finding solutions to alleviate the continuous disposal of these solid wastes is imperative. An efficient approach to minimize the potential damage to human health, ecosystem, and resources posed by hazardous waste is through policy improvement and process integration led by an effective leadership [39]. However, these policies catering to improved solid waste management practices, and geopolymer synthesis industrial standards would not be possible within the incoming years due to economic and spatial limitations [67]. Policies may focus more on reducing the process' global warming capabilities as it is a direct measure of the planet's health [68]. Widespread engineering applications of geopolymer material could not find their place yet in the industry as it is still an emerging technology [69]. LCA is a tool that can shift the paradigm by providing insights into the potential of the proposed solution towards circular economy [70].

Geopolymer technology has been around for decades; however, an intensive life cycle approach has not been applied yet to the use of PG and CFA for this technology. Philippines is still a coal-dependent economy and was the largest exporter of phosphoric acid from the SEA region thus, this study finds relevance in the local climate [10, 53]. The LCA may benefit those within the geographical boundary, improving the quality of life in the long run. The midpoint impact damages of utilizing PG in an entirely new process should be studied for a sustainable management of solid waste [71].

The current scenario (Scenario 1) exhibited beneficial effects in seven midpoint impact level over the alternatives mainly due to less involved chemicals and energy consumption. In terms of land use, landfilling appeared to be the most logical choice as manufacturing of the additional chemicals warrant more land area. Scenario 3 is more detrimental than Scenario 2 in this sense because of the inclusion of Na_2SiO_3 aside from NaOH whereas FAPG synthesis via the former can be facilitated with PA alone in the activator. Furthermore, NaOH is commonly produced via the chlor-alkali process which emits free radical chlorine in its production line [72]. Cl-containing compounds in the atmosphere are known for their ozone layer-destroying capabilities [73]. Scenario 2 emulates circular economy and thus reduces the life cycle impact of PG because of the main product being employed in geopolymerization. Eutrophication of freshwater was minimized in Scenario 3 compared to Scenario 2 evident by the use of phosphates.

The benefits credited to Scenario 2 may be attributed to the decrease in need for the extraction of raw materials. The partial substitution of gypsum with PG and sand with CFA proved to be beneficial in lessening the carbon emissions, boosting the energy economy, and reduce ecosystem and human toxicity impact. The same reason is applicable with Scenario 3, as well, but it is more advantageous in characterization where acidic discharges directly impact the criteria: freshwater and terrestrial acidification. Ideally, the landfilling of PG and CFA will be minimized as they are being valorized as cementitious material and geopolymers. FAPG synthesized in Scenarios 2 and 3 finds use conforming to several American Society for Testing and Materials (ASTM) International standard namely in: structural clay load-bearing tile, low-end loadbearing concrete masonry units, facing brick, non-loadbearing concrete masonry units, and as mortar for repair [47,74-77]. These signify the compatibility of the synthesized FAPG of both scenarios for non-loadbearing use. Scenario 3 was surpassed by Scenario 2 because the quantity of PG recycled in alkali FAPG is only 20% of what was utilized in acid FAPG. Midpoint impact assessment may vary depending on the potential engineering and construction applications to be examined in LCA.

Sensitivity analysis demonstrated that Scenario 3 could potentially outperform Scenario 2 in terms of environmental impact by employing similar CFA/PG proportion and A/P ratio in FAPG synthesis. However, the construction applications may be limited should that be the case, as the UCS of Scenario 3 using that formulation is lower. This shows a negative relation between the technical and environmental performance of FAPG for the alternative scenarios. Additional material in the precursor mixture has also been considered in previous studies. Rashad [56] elaborated studies that probed the suitability of PG in cement, concrete, mortars, and pastes including those synthesized via geopolymer technology as well. Substitution of ordinary Portland cement (OPC) is usually addressed when PG is added in construction material mixture. Shen et al. [78] tested the CS of cement with 5% raw PG against washed PG. A decrease in CS relative to using natural gypsum was observed for PG as received on a 28-day curing; mechanical strength increased with the washed PG. Other studies showed declination in workability with increasing PG concentration especially in concrete formulations [79, 80]. This phenomenon can be attributed to the water-soluble impurities (H2SO4, HF) which hinder the capability of PG for reuse [8]. Pre-treatment strategies had been suggested to circumvent this issue. Acidic components present in raw PG were neutralized via washing with water or with alkaline agents [81].

Wang et al. [82] developed a hemihydrate PG-based (HPG) geopolymer cement with fly ash (FA) and slag using an alkali activator. Highest UCS was observed for 50 HPG:44 slag:6 FA with a 28-day strength exceeding 60 MPa. These studies prove that the alkali-route synthesis exhibits room for improvement to achieve the performance akin to acid FAPG while increasing the amount of repurposed PG. The use of supplementary materials like PG in building materials reduces the environmental risk and minimizes material and transportation costs [83]. However, this poses a question on the possible implications of the hazardous components of PG and CFA and their intended purpose. Jaiswal et al. [84] emphasized the need for pre-processing steps for PG to deal with residual phosphates, fluorides, and heavy metals. The leachability of these components were addressed by Tomašević Pilipović et al. [85] in their study of applying PG in pavements in 3-5% mixtures. Minimal leaching was observed in their reported mixtures. The radioactivity of FAPG (46-105 µBq Rn-222) conforms to the standard value of 200 Bqm-3 for Rn-222 set by the International Commission on Radiological Protection (ICRP) for indoor dwellings [86].

This study evaluated the life cycle impact of the traditional management of PG and CFA against the two FAPG synthesis routes. Past studies conducted LCA involving geopolymers, but it was mostly in reference to OPC-based materials as the baseline. This study included OPC production within the baseline with partial substitution of PG and CFA to some of its components. OPC, however, is not utilized in the final geopolymer product mix. Rabie et al. [87] quantified the environmental impact of alkali-activated CFA geopolymer using the Centrum voor Mi lieukunde Leiden (CML) 2016 method. It demonstrated a decrease in CO_2-eq emissions, acidification, eutrophication, and energy resource use by 72%, 65%, 41%, and 29%, respectively. CPP is the main process contributing to the said impact categories for the baseline, while the major contributor in the CFA geopolymer process is attributed to alkali activator production. Similar observations apply for this study in the case of alkali FAPG.

LCA for FAPG synthesis remains limited. Previous studies that quantified the environmental impacts of FAPG evaluated mainly its CO_2-eq emissions avoided and leaching characteristics by employing this technology. Pratap [88] observed a positive correlation between CO_2-eq and the mechanical strength of FAPG. The observation of the current study for both scenarios agrees with the tendency of CS to decrease with the increase in PG replacement for CFA. Furthermore, acid- and alkali-activated FAPG displayed remarkable ability encapsulates the toxic elements found in PG. This makes the proposed technology a viable option to mitigate the environmental hazards brought by unmanaged industrial waste. By conducting a comparative LCA of the FAPG routes, the research gap is bridged through this study.

Despite the favorable benefits of FAPG, research remains limited; alternative activators, in situ experimentation, harmonized design standards, and low energy methods directs future research for its improvement [89]. Outside the regional context of the Philippines, other countries such as Spain, Morocco, and China deal with the similar problem of accelerating PG accretion. The sensitivity analysis could reflect how the life cycle impact varies considering market penetration or a larger scale of production volume. It can also provide baseline information should industrial standards are to be developed for FAPG materials for infrastructure development. Policies oriented towards decreasing the accumulation of PG in the environment can also be inferred from the sensitivity analysis.

4. Conclusion

The environmental effects of the acid- and alkali-based FAPG synthesis were quantified via LCA. The repurposing of PG as filler material for geopolymer application with alkali-based and acid-based activators were assessed against the use of PG for cement production. According to the midpoint level impact assessment via IMPACT World+ Midpoint v1.04, the acid-based geopolymerization process of CFA and PG holds the most advantage having the least environmental burden in climate change, fossil and nuclear energy use, mineral resources use, photochemical oxidant formation, freshwater ecotoxicity, human toxicity non-cancer, and particular matter formation. It reduced the CO_2-eq emissions PG and CFA production by 40%, decreased the energy consumption of extracting 36% less mineral resources by as high as 90%. Freshwater and human toxicity were also considerably reduced due to the immobilizing feature of heavy metals of the geopolymerization process. A large gap in environmental benefits was observed between acid-based and alkali-based FAPG in the characterization, where the alkali route is the most desirable.

On the other hand, alkali FAPG outperforms acid FAPG in terrestrial and freshwater acidification and water scarcity impact categories by 60%. The use of acid reagent is mainly responsible for this impact characterization. Both the alternative scenarios exhibited environmental burdens unlike the baseline due to increased land use geopolymer production plant, freshwater and marine eutrophication attributed to the use of phosphates, and ionizing radiation due to the inherent solubility of radioactive elements in acidic and basic conditions. Sensitivity analysis showed acid FAPG being more sensitive to changes in PG allocation, PG/CFA precursor ratio and A/P ratio than alkali FAPG. While the addition of chemicals usually entails more emissions and thus adverse environmental effects, process optimization and intensification can improve the current performance of the FAPG synthesis process. These insights are valuable references in the future direction of valorization of PG and CFA using geopolymer technology. Further research will focus on assessing the substitution of conventional chemicals with more sustainable ones and expanding the application of geopolymers as construction materials to emulate circular economy. Nonetheless, employing PG as a geopolymer material has an overall environmental benefit resulting from the reduction of solid waste discarded to the environment

5. Declarations

5.1. Author Contributions

Conceptualization, A.H.O., B.L.P., M.A.B.P., A.B.B., and J.F.D.T.; methodology, A.H.O., J.L.R.W.P., C.M.J.P., J.F.D.T., and V.J.T.R.; investigation, J.L.R.W.P., C.M.J.P., J.F.D.T., C.B.T., and V.J.T.R.; resources, A.H.O., A.B.B., M.A.B.P., J.F.D.T., C.M.J.P., B.L.P., and J.D.R.; data curation, J.L.R.W.P., M.A.N.P., J.P.P., A.H.O., A.B.B., and V.J.T.R.; formal analysis, J.L.R.W.P., C.M.J.P., and J.F.D.T.; validation, J.F.D.T., C.M.J.P., C.B.T., and V.J.T.R.; visualization, J.L.R.W.P., C.M.J.P., and J.F.D.T.; writing—original draft preparation, J.L.R.W.P.; writing—review and editing, A.H.O., A.B.B., M.A.B.P., J.F.D.T., C.J.M.P., M.S.M., C.B.T., V.J.T.R., B.L.P., J.D.R., J.L.R.W.P., M.A.N.P., and J.P.P.; supervision, A.H.O., A.B.B., M.A.B.P., J.F.D.T., and C.J.M.P.; project administration, A.H.O.; funding acquisition, A.H.O. and B.L.P. All authors have read and agreed to the published version of the manuscript.

5.2. Data Availability Statement

The data presented in this study are available in the article.

5.3. Funding

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5.5. Conflicts of Interest

The authors declare no conflict of interest.

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Appendix I

Table S1. Results of sensitivity analysis for Scenario 2

Midpoint level impact characterization	Unit	*k*	A	B	C	AB	BC	B^2
Climate change, short term	kg CO_2 eq	15.7	2.64	1.12	0.35	-0.19	0.12	0.37
Climate change, long term	kg CO_2 eq	15.0	2.58	1.04	0.33	-0.17	0.12	0.35
Fossil and nuclear energy use	MJ deprived	133	16.9	14.5	4.55	-2.41	1.61	4.82
Mineral resources use	kg deprived	1.64	0.37	0.04	0.01	-0.01	0.00	0.01
Photochemical oxidant formation	10^2 kg NMVOC eq	5.76	0.55	0.78	0.25	-0.13	0.09	0.26
Ozone layer depletion	10^8 kg CFC-11 eq	69.4	3.8	11.9	3.7	-1.98	1.32	4.0
Freshwater ecotoxicity	10^4 CTUe	1.12	-0.07	0.30	0.10	-0.05	0.03	0.10
Human toxicity cancer	10^7 CTUh	158	-6.91	40.7	12.8	-6.79	4.52	13.6
Human toxicity non-cancer	10^7 CTUh	177	13.1	27.4	8.61	-4.56	3.04	9.12
Freshwater acidification	10^8 kg SO_2 eq	454	-35.8	131	41.3	-21.8	14.6	43.7
Terrestrial acidification	10^3 kg SO_2 eq	4.00	-	1.00	-	-	-	-
Freshwater eutrophication	10^3 kg PO_4 eq	2.00	-	1.00	-	-	0.09	-
Marine eutrophication	10^3 kg N eq	10.0	1.00	2.00	-	-	-	1.00
Particulate matter formation	10^2 kg $PM_{2.5}$ eq	11.4	-0.30	2.70	0.90	-0.50	0.30	0.90
Ionizing radiation	Bq C-14 eq	28.0	-2.86	8.65	2.72	-1.44	0.96	2.88
Land transformation, biodiversity	10^4 m^2yr arable	90.0	-0.67	20.0	10	-	-	10
Land occupation, biodiversity	10^2 m^2yr arable	188	-7.60	47.8	15.1	-8.00	5.30	15.9
Water scarcity	m^3 world eq	13.0	-1.74	4.38	1.38	-0.73	0.49	1.46

Table S2. Results of sensitivity analysis for Scenario 3

Midpoint level impact characterization	Unit	*k*	A	B	C	AB	BC	B^2
Climate change, short term	kg CO_2 eq	15.3	2.70	0.99	0.31	-0.16	0.11	0.33
Climate change, long term	kg CO_2 eq	14.7	2.63	0.92	0.29	-0.15	0.10	0.31
Fossil and nuclear energy use	MJ deprived	123	18.8	10.4	3.27	-1.73	1.15	3.46
Mineral resources use	kg deprived	1.61	0.37	0.03	0.01	0.00	0.00	0.01
Photochemical oxidant formation	10^2 kg NMVOC eq	4.54	0.77	0.33	0.10	-0.05	0.04	0.11
Ozone layer depletion	10^8 kg CFC-11 eq	147.9	-10.1	41.3	13.0	-6.89	4.59	13.8
Freshwater ecotoxicity	10^4 CTUe	0.98	-0.04	0.25	0.08	-0.04	0.03	0.08
Human toxicity cancer	10^7 CTUh	124	-0.92	28.0	8.82	-4.67	3.11	9.34
Human toxicity non-cancer	10^7 CTUh	167	14.8	23.6	7.43	-3.93	2.62	7.86
Freshwater acidification	10^8 kg SO2 eq	135	20.8	11.3	3.57	-1.89	1.26	3.78
Terrestrial acidification	10^3 kg SO2 eq	1.00	-	0.10	0.03	-0.02	0.01	0.03
Freshwater eutrophication	10^3 kg PO4 eq	-	-0.05	-	0.04	-0.02	0.02	0.05
Marine eutrophication	10^3 kg N eq	9.00	1.00	1.00	-	-	-	-
Particulate matter formation	10^2 kg $PM_{2.5}$ eq	5.70	0.70	0.60	0.20	-0.10	0.10	0.20
Ionizing radiation	Bq C-14 eq	17.8	-1.05	4.82	1.52	-0.80	0.54	1.61
Land transformation, biodiversity	10^4 m^2yr arable	70.0	-	10.0	-	-	-	-
Land occupation, biodiversity	10^2 m^2yr arable	135	1.80	28	8.80	-4.70	3.10	9.30
Water scarcity	m^3 world eq	2.52	0.12	0.45	0.14	-0.08	0.05	0.15