



Review article

A phytoremediation approach for the restoration of coal fly ash polluted sites: A review

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ARTICLE INFO

Keywords:

Coal fly ash
Environmental pollution
Phytoremediation
Ecological restoration
Aromatic plants
Legumes
Grasses

ABSTRACT

Coal fly ash (CFA) is a predominant waste by-product of coal combustion which is disposed of in open ash dams that utilize large pieces of land. This waste material is classified as a hazardous substance in South Africa as well as in other countries due to its fine particles that are easily blown to the atmosphere and the unacceptable levels of heavy metals and persistent organic pollutants. Contaminants in CFA can pollute surface and ground water, agricultural sites, soil and therefore pose risks to the health of humans and the environment. More than 500 million tons of CFA is produced yearly and over 200 million tons remain unused globally. The production will continue due to high consumer energy demands, especially in countries with heavy reliance on coal for power generation. Despite a significant progress made on the application of phytoremediation approach for decontamination of polluted sites, there is very limited evidence for its potential in the rehabilitation of CFA dumps. Low organic carbon, microbial activities and availability of nutrients including nitrogen contribute to restricted plant growth in CFA, and therefore converting ash dumps to barren lands devoid of vegetation. Leguminous plant species can fix atmospheric nitrogen through symbiotic association with bacteria. Therefore, their intercropping mixture development can improve the chemistry of the substrate and facilitate nutrients availability to the companion plants. This approach can enhance the performance of phytoremediation and promote sustainable practices. The paper provides an overview of the ongoing burden of CFA disposal and discusses the ecological and economic benefits of using legumes, aromatic and bioenergy plants. We identify knowledge gaps to establishing vegetation in ash dumping sites, and provide insights to encourage continued research that will enhance the applicability of phytoremediation in restoration programs.

1. Introduction

Globally, South Africa has been classified as the sixth largest coal producer and exporter, and the mining industry contributes almost 27 % of the mineral resource sales, and therefore contributes significantly to the economy of this country [1,2]. Due to the significant economic contributions made by coal mining over the past few decades, it has gained prominence among the world economies. Coal-fired power plants (C-FPPs) produce approximately 40 % of the world's electricity in comparison to the 27 % generated in 2007. This supported the prediction that between 2007 and 2020, the world's coal consumption would increase by an average of 1.1 % per year and by an average of 2.2 % annually for the period 2020–2035 [3]. Many countries including South Africa

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<https://doi.org/10.1016/j.heliyon.2024.e40741>

Received 7 March 2024; Received in revised form 31 October 2024; Accepted 26 November 2024

Available online 30 November 2024

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depend on coal for power generation, largely due to the fact that it is the most available fossil fuel and affordable means of supplying electricity to consumers [4,5]. Eskom (national electricity supply utility) generates almost 90 per cent of useable electricity in the country from coal [6,7]. Approximately 600 million tons of CFA is produced per year, dominated by China, India, USA, Germany and South Africa contributing more than 300 million tons [8–10]. Of the CFA produced globally, more than 200 million tons remains unused and is stored in ash dumps thereby presenting serious environmental pollution problems. In South Africa, approximately 28 million tons of CFA is produced annually with utilization rate of less than 10 % [11–13]. Therefore, approximately 26 million tons of CFA are stored in disposal facilities requiring an area of land in excess of 600 ha [11]. Many power stations are currently running out of ash disposal sites and there is a need to expand the disposal facilities [12], this suggesting that more land is needed for storage. CFA storage in disposal facilities results in air pollution during windy seasons, groundwater and surface water contamination, and it affects soil quality and human health [14]. The tiny spherical particles of CFA ranging between 1 and 100 μm in size can suspend in air for longer periods and sometimes reduce atmospheric visibility [15,16]. Many ailments including respiratory distress, hypertension, kidney disease, gastrointestinal illness, heart damage, reproductive problems, cognitive deficits, impaired bone growth, birth defects in children, cardiopulmonary disease and cancers were reported in people living in the vicinity of C-FPPs [17–20]. All these health problems are linked to the inhalation of CFA particles. Because of all the health issues caused by CFA, this waste material is classified as a health hazard in many countries including South Africa. The CFA is disposed of as slurry which is stored in dumping areas and can be released directly into nearby surface water body [21]. Long-term storage can result in the leaching of toxic heavy metals and organic contaminants from CFA, ultimately contaminating groundwater and the underlying soils [13,21]. Although CFA is predominantly composed of carbon, it also contains iron, aluminium, Sulphur, organometallic compounds, mercury, chromium, cadmium, arsenic, lead and organic pollutants that are harmful to the biosphere [19]. The management of CFA is an important environmental challenge, which requires sustainable strategies to restore degraded lands [15,16].

Phytoremediation technology applies various plant species to transfer, stabilize, remove and lower availability of pollutants in the soil, groundwater and other waste materials [22–24]. It is the most eco-friendly and cost-effective strategy as the plants can colonize the ash dumps, by establishing a self-sustaining vegetative layer for recovery of the ecosystem health [25]. The polluted area is restored gradually over time, and this results to reduction of pollutants in the environment. Although, there are many reports on phytoremediation of polluted sites, investigations regarding the potential use of phytoremediation strategies for restoration of ash dams are rather sparse. This review aims to discuss the pollution problems related to the disposal of CFA, physicochemical properties of CFA, conventional clean-up methods, various means of CFA utilization and their limitations and explores the potential use of plants for restoration of degraded lands and economic opportunities. Commercialization is relevant for developing countries, as this will not only solve pollution problems, but will also address societal challenges. In an attempt to suggest suitable species, many studies focus on species that grow in the vicinity of ash disposal sites. However, these plants grow in soils polluted with CFA and therefore may not adapt to 100 % CFA media, hence there is sparse or no vegetation on the CFA disposal sites. Optimum pH and adequate supply of nutrients are among the important factors that support plants growth and therefore allowing vegetation and colonization of sites. However, the unfavorable conditions of CFA prevent plants from colonizing barren ash dams and for that to occur, the physicochemical properties of CFA must be improved. Leguminous species are known to improve soil fertility, enhancing carbon and nitrogen levels through increasing biomass input. These plants can provide nitrogen to the non-nitrogen fixing species, which is significant for plant growth therefore assimilation of carbon and consequently leading to natural vegetation. This paper examines the available studies on restoration of CFA deposits and suggesting unexplored leguminous species that may be suitable due to their capabilities in altering the chemistry of the rhizosphere. This approach encourages natural colonization of barren ash dumps using suitable species without the need for supplementation, which is often linked to secondary pollution phenomenon. Intensive nitrogen fertilization is linked to reduced plant species diversity and build-up over time that results to secondary pollution. In this regard, safer remediations approaches are recommended and the inclusion of legumes in planting programmes is sustainable due to their capabilities in improving properties of the substrate. Legumes in combination with other species including aromatic and bioenergy plants present opportunities for natural reclamation of barren ash dams with multiple benefits.

2. Production of CFA

Generating electricity through coal combustion results to the production of solid residues with environmental problems. Large quantities of solid wastes generated depend on the type of coal, combustion methods and emission controls [26,27]. Furthermore, ash forms and properties depend on temperatures used, fineness of the coal and the length of time the minerals are retained in the furnace as well as post-combustion conditions [28]. Ash produced from burning coal in C-FPPs consist of bottom ash, coal fly ash, boiler slag and flue gas desulphurization material [20,29]. Lower quality coal has very high ash content (35–45 %), resulting to high rate of CFA production [30]. Coal combustion waste accounts for 5–20 wt% of unburnt coal and includes coarse bottom ash (BA) (5–20 wt%) and fine CFA representing 80–95 wt% of the ash produced [9,13,31]. The bottom ash and boiler slag are the coarse and granular fractions, that are collected from the bottom of furnaces that burn coal for the generation of steam. The CFA is composed of very fine spherical glassy nanoparticles that is captured before reaching the atmosphere by highly efficient electrostatic precipitators or by a variety of other devices such as baghouses or wet scrubbers [13,32,33].

Coal is the dominant source of energy for South Africa, comprising more than 80 percent of the country's system load. The ash yields of coal samples from different coal mines in South Africa range between 20.0% and 49.6 % depending on the geographical origin of the coal used, emission control measures, ash collection methods, and other factors [6,34,35]. According to Yang [36], a reasonably clean coal should contain an ash content less than 12.5 %, indicating that much of the coal used is of poor quality, hence high production of ash. The coal reserves remaining are estimated to reach 116 years and due to consumer demands, it will remain a

cost-effective resource for energy production. More than 119 million tons of coal has been consumed between 2014 and 2015, resulting to approximately 35 million tons of CFA [12]. Though several alternative energy sources have been proposed and implemented, the rate at which the coal is being used as the main source of energy cannot be counterbalanced and therefore CFA production will continue to increase as shown in Fig. 1 and Table 2 [37,38].

3. Characterisation of CFA

The chemical and physical properties of CFA depend on the type of coal (bituminous, subbituminous, lignite and pozzolanic) used, geographical origin of coal, combustion and cooling procedures, emission control measures, ash collection methods which may impact its suitability of applications [34,40]. CFA consists of fine, powdery particles that are principally sphere shaped either solid or hollow and are predominantly glassy in nature [41]. The carbonaceous material in fly ash is composed of angular particles [42]. Mineralogical analysis of CFA has shown that 70–90 % of particles from pulverized coal consist of amorphous ferro-aluminosilicate glassy spheres with a smooth outer surface [43]. The spherical particles result from the burning of pulverized coal at high temperatures that range between 1400 and 1700 °C [44,45]. The hollow spheres in CFA, commonly known as cenospheres range between 45 and 150 μm [45–47]. CFA has a high specific surface area and low bulk density [9]. The optimal moisture content value for CFA varies between 11 % and 53 %, with maximum dry density values between 1 and 1.78 g/cm^3 . Physical properties of CFA including particle size, specific surface area, shape, permeability, angle of friction, thermal stability, colour, optimum moisture, specific gravity, and bulk density impact its suitability in various applications [9,48,49].

The most abundant materials in CFA are inorganic compounds, formerly present in the raw coal, which remain after the coal burning process with a small amount of unburned carbon that remains from incomplete combustion [44,45,50]. CFA is mainly composed of silica (SiO_2), alumina oxides (Al_2O_3), iron (Fe_2O_3), and calcium oxide (Cao) and with low levels of magnesium oxide, sulphur oxides, trace elements, base metals, and rare earth metals [49,51–54]. The most abundant elements in CFA include Si, O₂, Ca, Fe, Al, C, K, Na, Ti, P, Mg, and Ba [55–58]. CFA is classified by the American Society for Testing and Materials C618 (ASTM C618) standards as either class C or F [59,60]. Class C is mainly composed of calcium, alumina and silica and its calcium oxide content is 20 % or more [61,39]. This type is derived from subbituminous and lignite coals and is used as the replacement material in cement. Class F has lesser content of calcium oxide as compared to Class C and is mainly pozzolanic in nature. Pozzolanas are materials with reactive SiO_2 and Al_2O_3 , and low proportion of reactive CaO [62]. The Pozzolanic Activity Index is 93 % for Class F, while Class C is 79 %. The content of CaO in Class F is less than 20 %, therefore impacting its use as a replacement material in cement. It needs a cementing agent such as ordinary Portland cement, quicklime or hydrated lime with the presence of water to react and produce cementitious compounds. Class F CFA primarily consists of an aluminosilicate glass, quartz, mullite and magnetite. The total amount of ferric oxide, silicon dioxide and aluminium oxide is greater than 70 % for class F, while Class C is characterised by contents between 50 and 70 % of these minerals as depicted in Table 1. Class F require a cementing agent such as ordinary Portland cement, quicklime or hydrated lime with the presence of water to react and produce cementitious compounds [63]. The South African CFA is mainly composed of an amorphous silica-alumina glass phase (62.1 %), crystalline phases quartz (6.1 %), mullite (31.8 %) and classified as class F [64]. The South African CFA is mainly used as a pozzolan in ordinary Portland cement applications.

Classification, crystallinity, mineralogy, morphology, and chemical composition reveal the properties of this material and these provide guideline for its reusability [9,53,65,66]. Depending on pH, CFA is classified as acidic ash (pH 1.2–7), mild alkaline ash (pH 8–9) and strongly alkaline ash (pH 11–13) [9,60,65]. The pH values of CFA differ depending on the sulphur/calcium ratio. The concentrations of constituents vary according to the type of coal and they influence its colour [20]. The grey colour of CFA is explained by the high percentage of unburned carbon, while iron contributes to the brownish colour and tan to light is associated with lime [67]. Other properties of CFA are presented in Table 1S (Supplementary material). Unacceptable levels of toxic elements including As, Ba, Cr, Cd, Ni, Pb, and Hg were reported in CFA highlighting serious health risks due to its disposal [68,69]. The constituents of CFA include also a small number of radioisotopes such as ^{235}U , ^{226}Ra , ^{222}Rn , ^{238}U , ^{232}Th , and ^{40}K , which present radiation risk depending

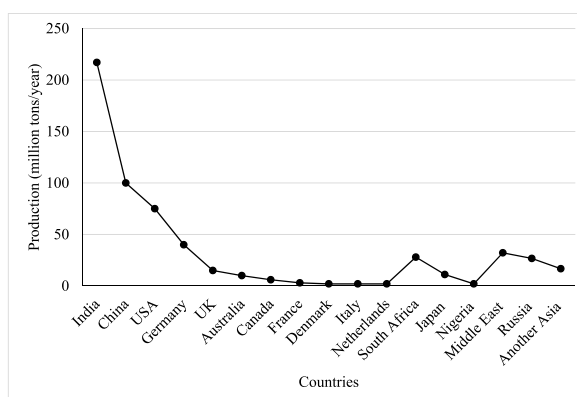


Fig. 1. Global production of CFA (million tons/year).

Table 1
Physical properties and chemical composition of Class C and F coal fly ashes.

Physical properties	Class C	Class F
Fineness, Retained on #325 Sieve (%)	15.9	25.7
Water Requirement of Control (%)	89	103
Pozzolanic Activity Index (%)	79	93
Soundness, Autoclave Expansion (%)	0.11	0.08
Specific gravity	2.58	2.30
Chemical composition		
Aluminium Oxide (Al ₂ O ₃)	19.4	24.0
Silicon Oxide (SiO ₂)	32.9	49.9
Total SiO ₂ +Al ₂ O ₃ +Fe ₂ O ₃	57.7	88.0
Iron Oxide (Fe ₂ O ₃)	5.4	14.4
Calcium Oxide (CaO)	28.9	3.23
Sulphur Trioxide (SO ₃)	3.8	0.88
Potassium Oxide (K ₂ O)	0.3	2.46
Magnesium Oxide (MgO)	4.8	0.98
Loss on Ignition	0.6	3.50
Moisture Content	0.8	0.11
Alkalis (potassium and sodium), and sulphates (SO ₄)	High	Low

Table 2
CFA production (million tons per year) and use (%) in different countries.

Countries	Production (million tons/year)	Utilization (%)	Approximate amount remaining (million tons/year)	References
India	217	77	49	[8,9,]
China	100	45	55	
USA	75	65	25	
Germany	40	85	6	
UK	15	50	7.5	
Australia	10	85	1.5	
Canada	6	75	1.5	
France	3	85	0.5	
Denmark	2	100	0	
Italy	2	100	0	
Netherlands	2	100	0	
South Africa	28	7	26	[11–13]
Japan	11.1	96	0.4	[9,]
Nigeria	2	54	0.9	
Middle East	32.2	10	29	
Russia	26.7	18	21.9	
Another Asia	16.7	66	5.7	

on distribution and concentrations [55]. Radioactivity level is a key environmental factor to consider for safe utilization of solid wastes [70,71].

The organic matter found in CFA include unburned carbon and various organic compounds. During the coal combustion processes, changes that occur result to the formation and emission of various organic compounds. The polycyclic aromatic hydrocarbons (PAHs) are among those emitted and these compounds are carcinogenic and mutagenic [72]. Relatively higher amounts of PAHs are found in CFA than that in the raw coal [73]. Low molecular weight-PAHs predominate in CFA samples [74]. Research also found that polychlorinated biphenyls (PCBs), aliphatic hydrocarbons and small number of dioxins are also present in CFA [59,75,76].

The organic pollutants in CFA are of significant environmental concern as they may exert harmful effects on the neighboring terrestrial, aquatic and aerial bionetworks [77,78].

4. Coal fly ash disposal

CFA is disposed of in lagoons/landfills/ash dams constructed near the power stations requiring large land area. These disposal sites are situated near the residential areas, agricultural fields, posing serious risks of environmental pollution. The two main CFA disposal mechanisms that are used by several C-FPPs worldwide include the wet and dry method. In the wet method, CFA is mixed with water and discharged into the fly ash settling ponds [8]. The disposal of CFA requires a huge amount of water burdening the already stressed water systems [79]. Although wet ash handling systems are closed circuits allowing for the ash water to be recycled, the recirculating water is lost through seepage, evaporation and absorption on the ash [11]. In case of dry disposal system, CFA is directly disposed of in basins and landfills covering several areas of valuable land near the power plants [80,81]. With the use of the dry system, the ashes can be dispersed due to wind in the surroundings which affect respiratory system and other human parts leading to cancer, bronchitis, anemia and asthma [43], [82]. To reduce the spread of CFA dust, suppression systems or water sprinklers are installed but these is not a sustainable strategy due to high water consumption. These common disposal practices are regarded as unsightly, converting lands to

non-productive sites, environmentally unsafe and are not feasible financially [52,83]. The disposal of high volume of CFA from C-FPPs consumes too much water and fuel during transport from the plant to the dumping sites. CFA contains a high concentration of salts, heavy metals and metalloids that can leach out into the environment through water and wind erosion, which further cause soil or water contamination in the surroundings of power stations [43,84]. The eco-friendliest alternative for CFA disposal sites is revegetation establishment. Vegetation could serve a variety of functions including stabilization of ash against wind and water erosion, reduction of leaching through water loss as evapotranspiration, provision of shelter and habitat for various life forms and restoring a degraded environment.

5. Coal fly ash utilization

To overcome challenges related to CFA disposal, many countries are reusing in a variety of applications and this has been beneficial for the health of the environment and contribution to economic growth. The recycling and utilization of CFA has increased over the years, for some countries as depicted in Fig. 2 and Table 2. In China, CFA is mainly used in production of building materials, paving, mine backfilling and low-end building accounting to 45 % utilization rate [85]. More than 60 % of produced CFA is recycled in the USA and is mainly used to produce cement products [86]. According to Yousaf [39], India produced 217 million tons between 2018 and 2019 and utilized 168.40 million tons of CFA for cement production, construction of dams and road, suggesting a utilization of more than 70 %. The EU has reported utilization rate over 90 %, which can be attributed to small volumes produced and the involvement in projects aimed at turning waste into valuable materials. For example, Netherlands, Denmark and Italy use up to 100 % of the produced CFA while in Asia, Japan only utilizes above 90 % [40].

CFA utilization is currently commercially feasible in the building and construction industry [13]. In South Africa, Eskom has embarked on CFA utilization project aimed at identifying markets that can require high usage of CFA and have proposed four applications including mine drainage treatment and backfilling, soil amelioration and land reclamation, road construction and brick and cement development [12].

5.1. Building and construction material

CFA is considered as a value-added material in construction and building industry (concrete and cement) including grout, block and brick development, geopolymers, environmental management such as backfilling and mine drainage treatment, land reclamation and soil amelioration, road construction, agriculture, raw material for glass-ceramics production, rubber industry, and recovery of valuable minerals [8,29,87]. CFA does not swell when used as a base material for buildings [88], and this makes it a suitable material in the

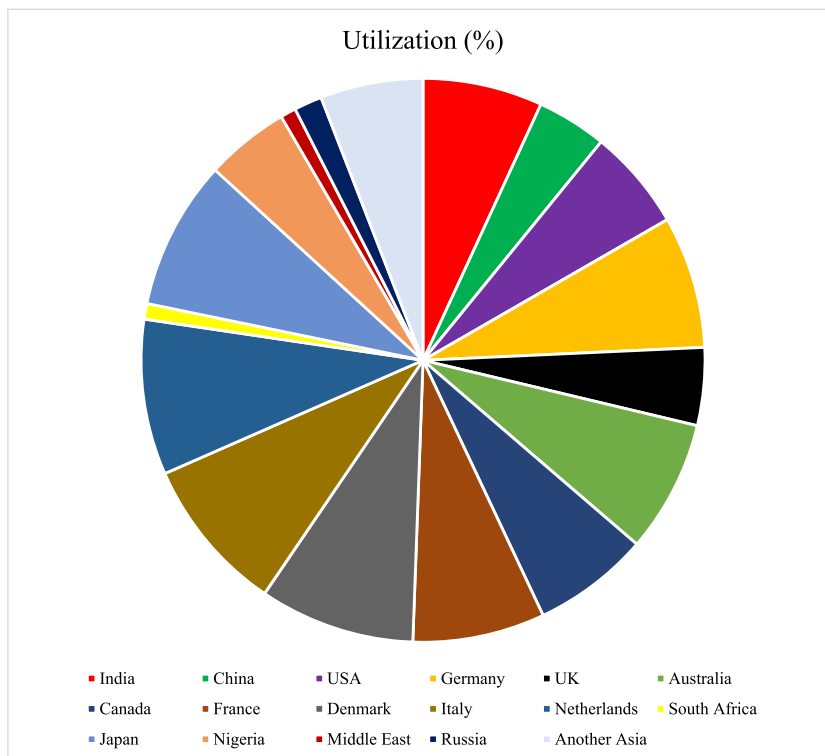


Fig. 2. Global utilization rate of CFA.

construction sector. The cement and concrete industries contribute greatly to the recycling of CFA accounting for 44.19 % of its utilization, followed by the road construction (15.25 %) and 12.49 % is reported for remediation of landfills [89]. The use of 40 % CFA in replacing cement resulted to cost reduction in the process of producing concrete production [40]. Supplementing with CFA results to hardened concrete through pozzolanic and hydraulic activity. An even higher amount of CFA up to 60 % can be used in structural uses [90].

5.2. Treatment of heavy metals and organic pollutants

CFA is also used to synthesize zeolite that can be used in agricultural activities and water treatment processes. The South African CFA has been confirmed to be a good substrate for silica nanoparticle and zeolite syntheses [91]. Zeolites have high sorption capacity due to the smaller particle size that increases the specific surface area ranging between 2500 and 4000 cm²/g [92]. The characteristics of zeolite include noteworthy ion-exchange capacity, high surface area and unique pore characteristics and therefore have been applied for treatment of heavy metals from industrial sludges, acid mine drainage and wastewater [55,93–95]. Nanomaterials synthesized from CFA demonstrated effectiveness for removal of heavy metals and organic pollutants [96,97].

5.3. Geopolymerization

CFA can be applied as a geopolymer concrete binder material due to its wide availability, greater potential and low cost for preparing geopolymers [98–100]. Geopolymeric materials synthesized from CFA can offer valuable solutions as they can immobilize toxic heavy or radioactive metals through process such as physical encapsulation and chemical stabilization [101]. However, CFA-based geopolymer materials may be affected by many parameters which are considerably related to the primary materials and their physical and chemical properties such as chemical composition, fineness and glassy phase content [99,102]. The CFA/kaolinite ratio, the water content and metal silicates utilized significantly affect the final geopolymer product [103].

5.4. Recovery of valuable minerals

While natural resources are decreasing with time, CFA contains a variety of beneficial metals and mineral substances whose demand is increasing in industries [104]. CFA is a potential alternative source of rare earth elements (REEs) and carbon nanoparticles, which are important components for various applications. A variety of consumer goods such as cell phones, computers, fluorescent lighting, catalysis, permanent magnets, advanced defense technology, and medical devices are made from these materials [105–107]. CFA also contain a valuable amount of alumina which offers a wide range of applications in various domains. Alumina has high mechanical strength, corrosion and wear resistance, the capability to withstand thermal stresses and high temperatures, high electric insulation, and improved dielectric properties [108,109]. Advancements in different sectors, such as automobiles, construction, defense, and industrial result in an increasing demand for alumina. It can be recovered from CFA using pyro-hydrometallurgical routes. Pyrometallurgical route can be used to change the mullite phase to a less stable state while the hydrometallurgical route can be used to dissolve the less stable form of alumina. Hydrometallurgical route has advantages such as low-temperature operation, high treatment capacity, and high removal capacity of different impurities, whereas pyrometallurgical route has environmental friendliness and low chemical consumption [109].

5.5. Agriculture

CFA has demonstrated utilization potential in agriculture as it contains a series of essential elements that contribute to improved soil properties. Essential elements such as Ca, P, K, S, Na, Mg, Zn, Fe, Cu, Co, Mo, and B that are beneficial for crop development are present in CFA [110,111]. In addition to improved nutrients availability, amendment of soil with low concentration of CFA improves soil characteristics such as pH, electrical conductivity, porosity, water holding capacity, and positively impacting plant growth [112]. Lower quantities of CFA on poor soil has been reported to enhance microbial activity. A study by Mandpe et al. [113] reported CFA as an additive for enhancing microbial and enzymatic activities in in-vessel composting of organic wastes. Diverse microbes respond differently to CFA dosage while actinomycetes and fungi inhabitants decrease during application of soil ameliorants [114]. A study by Leclercq-Dransart et al. [115] reported that addition of low CFA dosage enhanced fungal activity. CFA-based zeolites have the ability to retain water and cation exchange capacity and therefore widely used for the controlled release of fertilizers in agriculture [116]. Although there are benefits with the use of CFA as an amendment, this product is characterised by toxic elements that may be taken-up by crops and therefore entering into the food chain. Furthermore, toxic metals can suppress microbial activity [114]. The use of CFA in agriculture is also restricted because its repeated applications to the soil may lead to build-up of toxic metals over time and therefore careful monitoring strategies should be implemented. Converting CFA into reusable materials is an excellent strategy of reducing high volumes that need to be dumped into landfills, and consequently reducing the need for more storage space that is costly [97]. However, presently far less CFA is recycled and the low utilization rate in South Africa and many other countries highlights the need for effective management strategies to prevent further degradation of the ecosystem.

6. Treatment methods of contaminated sites

Several remediation strategies are being applied for clean-up of contaminated soils and ash disposal sites but most of them are

ineffective and expensive [117,118]. Methods for the treatment of contaminated sites include biological, physical and chemical methods which have advantages and disadvantages as well. Remediation methods are dependent on the contaminant and environmental factors, therefore there is no single or combined treatment methods that can be effective for remediation of all pollutants under same environmental conditions [119]. In addition to the use of landfills, the major treatment and disposal strategies of CFA include stabilization and solidification, chemical treatment and heat treatment [118,120]. The chemical and thermal techniques are technically difficult and not suitable for practical applications, because they are often associated with high cost, poor efficiency and are dependent upon the pollutants in the substrate [121].

6.1. Chemical remediation

This technology aims to alter the chemical characteristics of pollutants by decreasing their hazardous properties. The most commonly known chemical remediation methods include chemical oxidation and extraction, nanoremediation, ion-exchange, soil amendment, precipitation, and chemical leaching [118]. Chemical fixation fixes toxic heavy metals in the contaminated site through adsorption resulting to less availability of them. It is not a permanent treatment as heavy metals can get released into the environment under conducive conditions of weathering. It also consists of addition of reagents into the polluted sites to form slightly insoluble materials [122,123]. Chemical leaching is based on dissolving heavy metal ions into the acids such as H_2SO_4 , HCl or HNO_3 and extracting them out [124]. The limitation of this method is the need of large quantities of acids, which later require pre-treatment and therefore too costly. In the soil amendment technique which also called chemical fixation, the mobility and solubility of toxic heavy metals are reduced using chemical reagents such as CFA [118]. Nano-remediation is suitable for clean-up of hazardous elements in industrial waste. The technology involves the complex formation of heavy metals with modified nanoparticles, followed by co-precipitation, and ultimate removal of toxicants from the contaminated media [118,125]. The chemical oxidation is a remedial technique applied for *ex-situ* and *in-situ* effectively for the treatment of organic pollutants [126–128]. These methods require the use of oxidizing agents such as ozone, potassium permanganate, chlorine dioxide, hypochlorite, and hydrogen peroxide. The limitations of these methods include the production of secondary pollutants or by-products that need further processing and the need to use chemical reagents [129,130]. Furthermore, the use of these treatments is associated with high costs [131].

6.2. Physical remediation

The treatment of contaminated sites via physical remediations include soil replacement, excavation and off-site disposal technology, vitrification and heat treatment [118,119,132]. Soil replacement technique is based on the reduction of contaminant levels through completely or partially replacing the polluted soils. Physical technique for migrating contaminated land and disposal in landfills is labour-intensive, quite expensive, time consuming and not commercially sustainable [123]. Vitrifying technology consists of removing heavy metals through weathering process of organic matter by maintaining high temperature in the soils. This is a complicated technique which requires more energy, making it expensive and it has limited application. Thermal desorption is suitable for treatment of most semi-volatile and volatile pollutants such as PCBs and PAHs. In this method, these organic pollutants in contaminated media are heated first to an appropriate temperature by indirect or direct heating under vacuum or into carrier gas to separate target pollutants from the substrate. The advantages of this technique include possibility of recycling soil and contaminants, high safety, high efficiency, short treatment period, and capability to treat different types of pollutants. However, this method produces off gases which can result in secondary contamination. This treatment method requires soil excavation and transportation that can create new contamination sources such as rainwater contamination, dust and noise [133]. In addition, the other limitation is the high energy consumption and the resulting costs. In the excavation and off-site disposal technology, polluted soils are physically excavated and transported to disposal sites and treatment facilities, but this technology is not cost-effective [119]. Incineration method is a complex system representing an integrated system of components for waste preparation, feeding, combustion, and emission control. Thermal technology such as incineration method is a complex system that require utilizing electricity and therefore not a viable option for South Africa which is currently experiencing energy crisis. Remediation of polluted sites by biological methods can be advantageous to other methods as it does not require chemicals, cost-effective and no possibility of producing secondary pollutants.

6.3. Biological remediation

Bioremediation techniques involve the use of microorganisms and plants for clean-up of polluted environments. Microorganisms are readily available, easily defined, highly diverse, widespread, and can feed on various hazardous substances [134]. Despite the slower process which requires microorganisms to adapt to the environment, microbial metabolism has proven its success in decontaminating sites, through the production of enzymes and metabolites, without causing secondary damage to the environment [135]. Bacteria are the most commonly listed bioremediation agents among fungi, algae and yeast [136]. *Bacillus* spp. is a bacterial genus studied for bioremediation strategies, including biosorption, extracellular polymeric substance (EPS)-mediated biosorption, and bioaccumulation [137]. *Bacillus* spp. strains have proven their ability to reduce the concentration levels of heavy metals (Cd, Cr, Pb, Hg, As, and Ni) from contaminated substrates. *Rhodococcus* showed its effectiveness in the management of heavy metals which is due to the presence of appropriate enzyme systems, resulting in high decontamination rate [138]. Bacteria have been evaluated for potential to reduce the concentration of toxic elements and organic pollutants such as oil, PAHs and pesticides [139]. *Pseudomonas* species have demonstrated effectiveness in the treatment of contaminants attributed to their varied metabolic networks and ability to secrete biosurfactants to make hydrophobic substrates more bioavailable, thus enabling degradation of pollutants [140]. *Phanerochaete*

chrysochlorium, a fungal species is effective for degrading PAHs and dyes due to its effective mechanisms [141]. Furthermore, algae also demonstrated potential to degrade hydrocarbons and can also absorb heavy metals and other pollutants from water. Although little-discussed, earthworms and insects have been listed as soil animals which are equally valuable indicators of soil contamination, both before and after the application of bioremediation methods. The introduction of earthworms such as vermiremediation [142], and insects such as collembolans, ants, beetles, termites, and larvae, is an efficiently cheap method to break down organic pollutants in soil which enhance microbial activity and in turn degrade organic pollutants [143]. Vermiremediation and insect-driven remediation have received increasing research interest and thus more studies are reported widening the different biological remediation technologies [144]. The advantages offered by microorganisms include affordability, less or no by-products and its reusability potential.

7. Phytoremediation technology

Phytoremediation processes involve the use of native or indigenous plant species (shrubs, trees, herbs, aquatic plants, and grasses) and associated microorganisms, together with agronomic and chemical techniques (chelating agents) for decontamination of polluted sites in the environment [145,146]. Phytoremediation is an emerging environmentally-friendly, economical and sustainable technology applicable to large areas [146–148]. Vegetation is effective in restricting the spread of pollutants, reducing wind dispersion and water erosion.

Different phytoremediation mechanisms include phytoextraction, phytostabilization, phytovolatilization, phytodegradation, and rhizodegradation [149,150]. Understanding mechanisms involved in phyto-strategies is fundamental for successful implementation of these technologies. Metal-contaminated sites are mainly rehabilitated using phytoextraction and phytostabilization. In phytoextraction, plants referred to as hyperaccumulators are used to remove toxic metals or organic pollutants from the soil by accumulating them in the aboveground parts of the plant [151,152]. Phytoextraction technology can be achieved via natural means and the chemically enhanced phytoextraction, which involve the use of chemical agents [153]. Phytostabilization processes make use of plants to limit the movement of pollutants, thereby localising them in the roots [154]. The phytostabilisation process can be aided through incorporation of amendments into the soil to reduce the labile pollutants and phytotoxicity prior to establishment of suitable species [155]. While conventional methods disturb the physical properties of soil, the use of phyto-strategies maintains and improves the soil quality and structure [156,157]. Phyto-technologies maintain soil development processes, stabilises microbial communities, therefore promoting soil ecosystem roles through reduction/stabilization of pollutants [155]. Experimental trials, studying plant tolerance towards metals toxicity and tests under various amendments are essential before establishing a field-scale system. The ability to accumulate toxic metals varies greatly between species, and even between cultivars within a species [156,158]. For the successful practical application of phytoremediation, the production of biomass, the metal content of the plant material and the time span (number of annual crops) needed to achieve the desired degree of remediation of the soil are considered [159].

In phytodegradation, organic pollutants such as PAHs are taken by roots and broken down into less toxic substances, such as CO₂ and H₂O through metabolic processes. Organic pollutants are also removed or reduced from the environment by phytovolatilization process in which organics are absorbed from the substrate and converted to gaseous substances that are released into the atmosphere through transpiration [160,161]. The effectiveness of phytoremediation technology depends on the selection of suitable plant species and a variety of environmental factors. Spatial variability of soil factors, e.g. total concentrations and labile pools of contaminants, texture and depth of soil layers, nutrient and water availability, problems with the homogeneous input of amendments, differences in rooting depth and density, variability of soil moisture, impacts of pests, pathogens and herbivores can affect both treatment efficiency and plant responses in the field [162]. Microorganisms improve substrate properties by effecting chemical and biological changes and therefore widely applied to improve phytoremediation performance. Plant growth-promoting bacteria (PGPB) bioaugmentation has been proposed as a strategy to improve innate heavy metal phytoremediation capacity in plants, since it has been proven to promote plant growth, and is tolerant to metal stress [163–166]. Specific inoculum of microbes can be added to contaminated soils. Endophytic bacteria and Rhizospheric bacteria are two distinct categories of bacteria commonly reported to facilitate plant phytoremediation. These species have the capacity to enhance plant development and stress tolerance through the production of siderophores and secondary metabolites. The secondary metabolites including phenolic compounds play a role in the plants defence mechanism. Endophytic bacteria are considered a subclass of rhizospheric bacteria that can improve plant health by targeting pests and pathogens with antibiotics, hydrolytic enzymes, nutrient limitation and by priming plant defences [167,168]. Rhizodegradation is a phytoremediation strategy that uses roots of plants that enhance microbial and fungal activity in the rhizosphere and breakdown organic pollutants [169]. Various laboratory or green house studies have employed *Helianthus annuus* with and *Bacillus* species and the reports revealed enhanced accumulation of metals in their tissues [170]. Controlled inoculations of plants with growth-promoting microorganisms can significantly improve plant growth and stress tolerance in contaminated soils, therefore facilitating the phytoremediation process [171]. Synergistic use of plants and microbes results to more efficient clean-up processes [172]. The combination of *Brassica juncea* with bacteria such as *Pseudomonas*, *Bacillus* sp. and *Azotobacter chroococcum* were effective for clean-up a Cr contaminated soil and the enhanced plant growth was attributed to phosphorus and potassium solubilisation and the associated N fixation process [173]. A pot trial by Mesa-Marin et al. [174] reported a microbe-assisted phytoremediation strategy under greenhouse conditions, in which plant growth promoting and heavy metal resistant bacteria improved metal uptake therefore, assisting the performance of phytoremediation. Plant-associated bacteria improve phytoremediation processes by altering the solubility, availability, and transport of heavy metal and nutrients by reducing soil pH, release of chelators, P solubilisation, or redox change [169].

7.1. Selection of plant species

Depending on the substrate and pollutants, selection of suitable plants species is a foundational need to be addressed in phytoremediation strategies. Species suitable processes should be tolerant to the heavy metal toxicity and be able to produce a large biomass [175]. Fast growing species offer an advantage as they form massive green cover in reasonable less time and conserve moisture [176]. Plant that require less maintenance under field conditions and adapt to environmental factors including low water availability and low nutrients availability is more desirable [177]. Native plants have the advantage of tolerance to local growth medium condition. For restoration of CFA deposits, it is more desirable to select species that limit movement of metals depending on the pollution load. In the avoidance strategy, plants limit uptake of heavy metals and restrict their movement into aerial parts through mechanisms including root sorption, metal ion precipitation and metal exclusion [178]. Excluders restrict or limit the roots to uptake metals and translocation in high concentration of pollutants inside the plant tissue over a wide range of growth medium (soil or CFA) or substrate concentrations [179–182]. They stabilize metals in the growth medium (phyto-stabilization), reduce the mobility and bioavailability of organic and inorganic pollutants, preventing pollution of surface and groundwater and subsequent entry into the food chain [183,184]. Studies have profiled a number of species growing in the vicinity of CFA dams, but these should be tested in the field trials for longer periods in order to determine their effectiveness in phytoremediation processes. It is also important to select species based on their ecological importance, dominance at the site under investigation and socio-economic importance for community upliftment [38].

7.2. Nanotechnology-assisted phytoremediation

Phytoremediation, in combination with nanotechnology, has potential for efficient clean-up of polluted sites [185]. Nano-phytoremediation is a new emerging technology and gaining interest for restoration of polluted sites through the use of bio-synthesized nanoparticles [186]. Nanomaterials have unique physicochemical properties such as large surface area, more active sites, and high adsorption efficiency that enhance the application of phyto-strategies for clean-up of polluted substrates [187]. This approach has shown potential to enhance biomass production for plants growing in a polluted environment and improve plants capability to accumulate toxic metals in their tissues [188,189]. Nanoparticles can enhance plant growth and stabilize contaminants through augmenting antioxidant activities [190]. A variety of laboratory trials have been reported for the application of nanomaterials on plants for removal of toxic pollutants [191]. Silicon nanoparticles application is effective in increasing wheat grain biomass and yield while reducing toxicity of Cd in plants [192]. Nanoscale zero-valent iron has proven to be effective in phytoremediation processes and therefore widely investigated in remediation of contaminated soil and groundwater [171]. Huang et al. [193] discovered that application of zero-valent iron nanomaterials improved Pb accumulation in ryegrass, therefore enhancing the removal of Pb from the environment. Although nanomaterials have demonstrated advantages in restoration processes, they are potential environmental hazards, especially in soil microbial communities [171,194]. There are toxicity concerns with the use of nanomaterials in phytoremediation processes, this can be a beneficial strategy and therefore further research is needed to develop effective and safer approaches for its applications.

8. Challenges of establishing vegetation in CFA-polluted sites

Revegetation has been reported as an efficient approach for stabilization and restoration of polluted sites, thereby protecting the environment. However, studies on CFA polluted sites are still poorly reported, possibly due to unfavorable physicochemical properties of CFA that do not support plant development. Although CFA contains some essential nutrients, they are not bioavailable due to high pH values resulting from presence of hydroxides, carbonates, calcium, and low Sulphur content [195,196]. Essential cations including Ca, Mg and K are available for other plant physiological processes, but low levels of nitrogen and phosphorus limit plant growth [25]. Furthermore, high concentration of soluble salts is phytotoxic and requires a washing step, in order for plants to thrive. The available phosphorus content in CFA is usually 0.05–0.20 % due to complexation with Al and Fe. Nitrogen is estimated to be below 0.05 % as most of it, is lost by volatilization and therefore restricting plant growth [197]. Although Boron (B) is an essential element, CFA is characterized by toxic levels of B which severely hinders plant growth [198]. Plants accumulate high levels of B in its tissues due to excessive uptake and therefore resulting to phytotoxicity [199,200]. Alkalinity of CFA can lead to increased release of selenium and toxic levels, and this may hinder plant growth, resulting in chlorosis, burning of leaves and oxidative stress [104]. Depending on the coal used, CFA contain high levels of metals (As, Be, B, Cd, Cr, Co, Cu, Ni, Fe, Mn, Pb, and Hg) and organic pollutants (PAHs, PCBs, pesticides, etc.), which may be toxic to plants and therefore hindering plant growth [201–203]. The fine particles of CFA form compacted layers which leads to low aeration and water infiltration [36]. CFA is characterized by low water holding capacity, low organic carbon and lower community of microorganisms [25,43]. Microorganisms excrete root exudates needed by other microbial communities that in exchange for the provision of essential organic compounds, providing support for plant growth and therefore aid plants to thrive in harsh environments [204]. Plant-microbe interactions contribute significantly to improved soil structure, nutrient cycling and nutrient uptake [205].

9. Approaches in phytoremediation of CFA-polluted sites

CFA disposal has resulted in significant loss of flora and destruction of the ecosystem due to its severe conditions that restrict the establishments of vegetation in CFA disposal sites. Various amendments are required to improve unfavorable CFA physicochemical properties such as high pH that is linked to unavailability of nutrients [195,206]. High salinity due to dissolved salt has led to

phytotoxicity negatively impacting the establishment of vegetation on CFA disposal sites. CFA has low content of organic matter and this play a variety of roles including reduced erosion, improved substrate structure and porosity allowing unrestricted movement of air and water [207]. The organic matter retains moisture and allows slow release of nutrients and is the source of food for microorganisms. Organic matter sources include plant based organic fertilizers such as sewage sludge, wood chips, straw, biosolids, compost, sludges, manures, bacterial inoculum and dairy sludge [208]. The use of compost as an organic amendment effects the plants differently for each species. Compost consists of nutrients for the native microbes present in the polluted substrate and can increase the water-holding capacity and the cation-exchange capacity [209]. The composts can act as both bioremediation technique and soil amendment agents, in which the microbes employ these nutrients for the remediation of pollutants present. Amendment through the addition of compost results in improved toxic metal uptake [210]. To increase efficiency, contaminated sites were found more effectively remediated by implementing the microbes isolated from the polluted sites. Microbes assist in increasing the absorptive surface area of the plant species. The addition of bioinoculants and microbes can establish an ecological system including complex substrates and plants [208]. Nutrient support through fertilizers and manures provides the possibility of effective remediation and the nutrients to the matrix combination within a lesser time. Application of fertilizers in phytoremediation can improve plant biomass and improve the plant absorption of toxicants [211]. Inorganic fertilisers contain the three main nutrients (N, P, K) which are lacking in CFA, but their excessive use can lead to a build-up of salts in the substrate leading to contamination problems. Treatments such as the addition of fertilizers, composts and manure are effective in enhancing the availability of nutrients, but can also increase levels of toxic metals that will be source of pollution problems over time [212]. Therefore, the safety of the selected amendment should be established prior to application. The strategy to clean up restore CFA disposal sites through phytoremediation is promising, but currently, such studies are sparse, possibly due to the harsh conditions of CFA that hinder plant growth. These problems can be addressed by altering the substrate

Table 3
Species with potential in eco-restoration of CFA disposal sites.

Plant species	Country	Pollutants	Remarks	References
<i>Tamarix tetrandra</i> , <i>Robinia pseudoacacia</i> , <i>Populus alba</i> , <i>Amorpha fruticose</i>	Serbia	As, B, Cr, Cu, Mn, Ni, Se, and Zn	Case study assessing the phytoremediation potential of planted and spontaneously colonized woody plant species on CFA disposal sites	[184]
<i>Acacia nilotica</i> L., <i>Acmella oleracea</i> L., <i>Bacopa monnieri</i> L., <i>Cynodon dactylon</i> (L.) Pers., <i>Cyperus rotundus</i> L., <i>Dactyloctenium aegyptium</i> L., <i>Digitaria sanguinalis</i> L., <i>Trianthema portulacastrum</i> L., <i>Typha latifolia</i> L. and <i>Portulaca oleracea</i> L.	India	Not listed	Vegetation by naturally colonizing plants for eco-restoration of CFA disposal area	[214]
<i>Nicotiana glauca</i> , <i>Protea burchellii</i> , <i>Ipomoea pestigridis</i> , <i>Althernathera pungens</i> , <i>Eurphobia hirta</i> , <i>Amaranthus spinosus</i> , <i>Cynodon dactylon</i> , <i>Dactyloctenium aegyptium</i>	Botswana	Cu, Pb and Zn	Naturally growing in CFA dumpsite	[215]
<i>Typha latifolia</i> , <i>Saccharum spontaneum</i> , <i>Prosopis juliflora</i> , <i>Ipomea carnea</i> , and <i>Saccharum spontaneum</i>	India		Physiological profiling of invasive plant species for ecological restoration of CFA deposits	[216]
Barley (<i>Hordeum vulgare</i>), Sudan grass (<i>Sorghum bicolor</i>), canola (<i>Brasica campestris</i>), rapeseed (<i>Brassica napus</i>), alfalfa (<i>Medicago sativa</i>), and perennial ryegrass (<i>Lolium perenne</i>)	North Dakota USA	Co, Cr, Cu, Sr, Ti, Tl, and V	Environmental health aspects of coal ash phytoremediation by selected crops	[217]
<i>Agrostis stolonifera</i> , <i>Calamagrostis epigejos</i> ,	Poland	Cu, Co, Mn, Cd, Ni, and Pb, Fe, and Zn	Heavy metal and nutrient uptake in plants colonizing post-flotation copper tailings	[218]
<i>Saccharum spontaneum</i> -grass and <i>Cymbopogon citratus</i>	India	Mn, Cu, Zn, Cd, Ni, Cr, and Pb	Studies on colonization of CFA disposal sites using invasive species and aromatic grasses	[16]
<i>Ricinus communis</i> -castor <i>Amaranthus watsonii</i> , <i>Solanum lumholtyianum</i> , <i>Bromus catharticus</i> , <i>Acacia farnesiana</i> , <i>Gnaphalium leucocephalum</i> , <i>Brickellia coulteri</i> , <i>Baccharis sarothoides</i> , <i>Prosopis velutina</i> , and <i>Boerhavia coulteri</i> -shrubs	Mexico	Al, Ag, Ba, Bi, and Sb	<i>Cistus ladanifer</i> phyto-stabilizing soils contaminated with non-essential chemical elements	[219]
Seedlings of rye, wheat, oats, barley, triticale, and regreen (hybrid between wheat and ryegrass)	USA	As, Cd, Co, Cr, Li, Mn, Pb, Ni and V	Preliminary study to assess potential of cereal crops for restoration of CFA deposits	[220]
<i>Brassica juncea</i>	India	Pb, Mn, Cr, Zn, and Ni	Phytoremediation of metals from CFA through bacterial augmentation	[221]
<i>Dalbergia sisso</i> , <i>Bougainvillea glabra</i> , <i>Casuarina equisetifolia</i> , <i>Delonix regia</i> , and <i>Thuja occidentalis</i>	India	Fe, Cu, Zn and Mn	Remediation of CFA landfills through plantation	[196]
<i>Tamarix chinensis</i>	China	P, K, Na, Al, Cu, Fe, Mn, Pb, Zn, and B	Natural revegetation of CFA in a highly saline disposal lagoon	[222]
<i>Amaranthus hybridus</i> , <i>Digitaria eriantha</i> , <i>Grasses</i> ; <i>Brachiaria serrata</i> , <i>Heteropogon contortus</i> , <i>Tristachya leucothrix</i> , <i>Setaria sphacelata</i> , and <i>Cynodon dactylon</i>	South Africa	Fe, Mn, Cu, and Zn	A comparative analysis of the vegetation and topsoil cover nutrient status between two similarly rehabilitated CFA disposal sites	[223]
<i>Atriplex</i> , <i>Enchylaena tomentosa</i> , <i>Halosarcia</i> , <i>Mesembryanthemum</i> , <i>Nitria billardieri</i> , and <i>Scaevola Colloris</i>	Australia	Not listed	Revegetation on CFA lagoons	[224]

properties and careful selection of plants species. To establish vegetation in CFA ash dams, factors including low organic matter, availability of nutrients, microbial community, toxic elements and high salinity should be addressed. Investigating these environmental factors can be used to predict plant response to stressors and therefore establish sustainable and successful restoration programs.

10. Plant species with potential in the phytoremediation of CFA-polluted sites

Toxicity of CFA negatively impacts the plant normal physiological processes and hence the selection of plant species plays a critical role in the success of phytoremediation processes [80]. The occurrence of naturally-growing plants near CFA dumpsites has opened doors for opportunities to explore phytoremediation strategies for reclamation of these unproductive sites [60]. These plants species are able to self-propagate successfully with no additional inputs [213]. Native plants that grow in CFA deposits are often characterized by high levels of toxic metals in their plant tissues because they develop survival mechanisms appropriate for the respective environmental condition. Naturally colonizing plants can adapt to harsh conditions and are capable of improving properties of the rhizospheric CFA [214]. Numerous studies revealed that CFA is polluted with heavy metals and species listed in Table 3 demonstrate potential of native plants for their potential in colonizing CFA ash dams. These species provide valuable information that can be useful in identifying ideal species for a particular environment.

The species that grow around the vicinity of ash dumps present an opportunity for selection of suitable species for phytoremediation strategies. It is noteworthy to mention that, the phytoremediation process depends on the species, concentration and type of pollutants and environmental conditions [155]. The tree species (*Betula pendula* Roth, *Populus tremula* L.) and herbaceous species (*Solidago virgaurea* L.) growing on coal combustion waste landfills were effective in accumulating Cd and Zn, but with low accumulation for Pb and Cu [10]. There are no plants that have universal applications for phytoremediation processes due to differences in site conditions, and therefore co-cropping systems should be explored to improve the phytoremediation capacity of plants by taking advantage of distinct crop capabilities. The number of differentially abundant rhizosphere-associated bacterial species were higher in co-cropping system including a legume in comparison to monocultures [225]. These highlight the need for careful selection of organic amendment such as the use of legumes that impact the microbial communities. Implementing the microbes isolated from the polluted sites can be beneficial, as such microorganisms have adapted in the particular environment. Therefore, plant-microbe dynamics should be investigated to guide the selection of suitable inoculum.

10.1. Grasses

In the initial stage of phytoremediation processes, the species selected must be tolerant or resistant to pollutants present or conditions in the substrate. Grasses have wide fibrous root systems, which prevent erosion and tolerate harsh growing conditions, have high biomass production and therefore suitable for restoration strategies [226]. They stabilize toxicants through absorption in their roots and translocation to the aerial parts and therefore their cultivation can diminish water, land and air pollution and prevent dust flow. *Cynodon dactylon* pers. (L.) (Bermuda grass) demonstrated phytoremediation potential for the phytoextraction of Cd and degradation of PAHs [171]. This species was reported to be amongst the species that naturally colonised CFA dumps and accumulated toxic concentration of Cd [227]. *Cyperus rotundus* L., is a metal-tolerant plant species which grows well on metal-contaminated CFA deposits [228]. The phytoremediation potential of *Azolla caroliniana* (water fern), which naturally occur in CFA dams was demonstrated by high bioconcentration factor (BCF) values for Pb, Fe, Mn, Ni, Cr, Zn, Cd, and Cu that ranged from 1.7 to 18.6 [195]. Vetiver (*Vetiveria zizanioides*) demonstrated phytostabilization potential after being grown on CFA for a period of three months and the plant had massive growth of roots [229]. The high porosity and low density of CFA facilitate root growth in *Vetiver zizanioides*, which is a commercial part of these plant. Higher biomass may be obtained from CFA dumps as compared to other wastelands because poor soil structure restricts root development and growth, and root breakage occurs during its digging [38]. Field trial by Pandey et al. [195] showed that *Saccharum munja* grows well on abandoned CFA lagoons and its roots increased from 3.48 to 4.57 m deep and produced high biomass. *Saccharum spontaneum* is a tall (100–600 cm) perennial grass with deep roots and high biomass productivity that is suitable for restoration and stabilization of ash deposits. Similarly, Kumar et al. [230] reported the accumulation and translocation of Ni, Zn, Cu, Fe, and Pb by *Cynodon dactylon* and *S. munja*. Vegetation with *S. spontaneum* improves porosity, water holding capacity, decrease pH and conductivity and consequently enhances nutrients availability. Grasses can be mixed with trees, as they present financial gains. It is imperative that planting is approached in a manner that minimize competition for water and nutrients between trees and grasses.

10.2. Legumes

Very low levels of nitrogen and reduced nitrogen-fixing microorganisms limit plant growth in CFA. Although some nutrients are present in high levels, they are not bioavailable due to various factors including high pH. This phenomenon can be overcome by the use of leguminous species that release hydrogen ions, thereby reducing the pH of CFA which enhances bioavailability of other nutrients. Legumes have gained interest in phytoremediation strategies due to the nitrogen fixing capabilities, which modify the physicochemical properties of the CFA [231]. Legumes change soil properties through the symbiotic association with microorganisms, such as rhizobia, which fix the atmospheric nitrogen and therefore increasing the available nitrogen in soil [232]. The presence of ammonium ions and nitrates contributes greatly to plant development. Leguminous species are heavy-metal resistant and can significantly reduce organic pollutants such as PCBs, PAHs and herbicides from soil by degradation mechanism [152,198,231,233]. Traits of ideal species for

phytoremediation is fast growth and accumulation of biomass, but studies report unavailability of nutrients as the limiting factors for plant growth on CFA media. Legumes have great potential in phytoremediation processes because they enhance the number of microbial populations and can make fixed nutrient available to other crops [25].

The use of legumes in phytoremediation strategies provides economically feasible and environmentally-friendly strategy to reduce the use of chemical amendments in a bid to improve soil characteristics. For examples, *Lotus corniculatus* belonging to the Leguminosae family was successfully used for soil restoration contaminated with crude oil [234]. *Medicago lupulina* has phytostabilization potential due to its ability to accumulate high concentration of Pb, Ni and Zn in its root tissues and high biomass production [235,236]. *Acacia nilotica* is an ecologically, economically and socially important Leguminosae tree species that has phytoremediation potential of contaminated soils with Cd [237,238]. The products derived from this species such as fuel, gum, fodder, and drugs can be used as green manure to enhance soil fertility, to adapt to climate change and to fight against societal poverty in the rural areas [238]. Such approach is beneficial for sustainable development of communities. A variety of species have been tested for clean-up of polluted soils (Table 4), but little attempts made for CFA deposits restoration. *Medicago sativa* has been evaluated for its potential to restore CFA disposal site contaminated with Co, Cr, Cu, Sr, Ti, Tl, and V [220]. The capabilities of legumes to change the properties of CFA highlight immense potential in the restoration of CFA deposits. These species can be cultivated alongside the other plants and modify the substrate properties, therefore making the growth media conducive for plants to thrive. The leguminous species listed in Table 4 occur in many parts of the world and have been investigated for potential in cleaning-up metal-contaminated soils and mine tailings. Therefore, legumes are promising candidates that deserves to be explored for their application in the restoration of CFA dams. The advantage of these species are their capabilities to improve substrate fertility without any external input as compared to non-leguminous species.

In Southern Africa, the legume family is comprising of 133 indigenous genera and 1620 indigenous species [253]. *Trifolium repens* which occur in many countries including South Africa was evaluated for its effectiveness for the removal of organic pollutants and restored polluted soil by improving its properties [254]. The influence of co-cropping two plant species, *Chrysopogon zizanioides* (vetiver grass) and the legume *Medicago truncatula* (barrel clover) demonstrated improved the plant's tolerance toward metals toxicity [255]. Developing co-cropping system is more beneficial as they increase biomass of the plants and improve microbial diversity of the rhizosphere, therefore effective for vegetation of barren lands.

10.3. Grass-legume mixture

Strategies are being explored with the use of microbial inoculants and inclusion of different plants such as legume-grass mix and inclusion of wastewater [208]. Grass-legume mix is efficient as they can readily colonize the polluted site and develop a thick vegetation cover within a reasonable timeframe. The advantage of these combination is the creation of nitrogen balance in the substrate. By comparing with other species grown in low nutrient media, legumes have a higher decomposition over grasses and therefore thrive much better [256,257]. Legumes survive harsh conditions caused by contaminants better than grasses [198]. The grass-legume mixture is effective in the restoration of waste disposal site [176]. This approach has shown potential in restoration of CFA deposits, and therefore should be explored for the benefit of the environment and restoration of the ecosystem.

10.4. Aromatic plants

Phyto-strategies are now being approached in a manner that enable solving problems simultaneously, i.e. restoration and revenue generation. Aromatic plants are widely used for their aroma and flavour and also used for their medicinal properties. These plants produce essential oils (EOs) which are characterised by a variety of secondary metabolites dominated by terpenes [258]. The EOs that are used in perfumery, aromatherapy, personal care products, detergents and soap manufacturing, pharmaceutical, food industries, and as insect repellents. These species are gaining interest in phytoremediation strategies as they can grow in metal-contaminated sites and can also produce high value EOs [38,259]. These species can simultaneous address pollution problems due to metal toxicity, while generating revenues from commercialization of EOs extracted from harvested biomass [260,261]. The EOs are extracted through hydrodistillation and therefore the oil produced is free from metals [262–264]. The use of EOs provide renewable sources of valuable

Table 4
Legumes with potential in eco-restoration of CFA deposits.

Plant	Country	Mycorrhizal fungi	Element	References
<i>Anthyllis cytisoides</i>	Italy	<i>Glomus macrocarpum</i> , <i>Glomus mosseae</i>	Lead, Zinc	[239]
<i>Astragalus sinicus</i>	China	<i>Glomus mosseae</i> , <i>Glomus intraradices</i>	Cadmium	[240]
<i>Canavalia ensiformis</i>	Brazil	<i>Glomus etunicatum</i>	Zinc	[241]
<i>Glycine max</i>	Brazil	<i>Glomus macrocarpum</i>	Lead	[242]
<i>Leucaena leucocephala</i>	China	<i>Glomus</i> spp.	Lead, Zinc	[243]
<i>Medicago truncatula</i>	France	<i>Glomus intraradices</i>	Cadmium, Zinc	[244]
<i>Pisum sativum</i>	France	<i>Glomus intraradices</i>	Cadmium	[245,246]
	India			
<i>Sesbania cannabina</i> , <i>Sesbania rostrata</i> , <i>Medicago sativa</i>	China	<i>Glomus mosseae</i>	Copper, Zinc	[247]
<i>Trifolium pratense</i>	China	<i>Brevibacillus</i> sp., <i>Glomus mosseae</i>	Zinc, Lead	[248,249]
<i>Trifolium repens</i>	Spain	<i>Glomus mosseae</i>	Zinc, Iron, Lead, Cadmium	[250]
<i>Trifolium repens</i>	Spain	N/I	Cadmium	[251]
<i>Vigna radiata</i>	Pakistan	N/I	Chromium	[252]

plants products, protection of plant biodiversity and restoration of degraded sites [265]. Aromatic grasses are perennial in nature with multiple harvests and tolerant to stress conditions such as drought, heavy metal toxicity and unfavorable pH [266]. Aromatic grasses such as *Vetiveria zizanioides*, *Vetiveria nigriflora*, *Cymbopogon martinii*, *Cymbopogon winterianus*, and *Cymbopogon flexuosus* have been evaluated for their effectiveness in phytoremediation strategies [38,260]. These plants are high-value plants due to their high biomass production and abilities to produce EOs [38,267–269]. Shrubs have also been tested in phytoremediation strategies and have shown their effectiveness in many phyto-strategies. *Vetiver zizanioides* and *Mentha arvensis* were effectively planted in CFA used in conjunction with mycorrhiza and 20 % farmyard manure [270–272]. Many field trials have focused on monoculture, limiting survival of species. This can be overcome by establishing a diverse plant community and inoculation of various microorganism. Aromatic shrubs can be grown together with grasses as they provide vegetation cover, immobilizing pollutants, whilst shrubs are being established. Shrubs and trees establish a deeper root network to prevent erosion and they may enhance nutrient availability for grasses, whilst reducing water deficiency and improving soil properties [213].

Aromatic plants are not edible and are not being consumed directly by animals and humans like vegetables, pulses and cereals and therefore there is a low risk of entering into the food chain. Furthermore, grazing animals do not feed on aromatic plants due to their essence and are therefore abundant and can be used on a large scale [266]. The global demand of EOs is expected to increase from around 226.9 kilotons–404.2 kilotons between 2018 and 2025 [267]. According to Verma et al. [38], the global request of herbal products, in which EOs from aromatic herbs contribute considerably is projected to reach 5 trillion U.S. dollars by the end of year 2050. Tests should be conducted on EOs produced to ensure the safe use of these products. Cultivation of aromatic plants on CFA dumping sites is economically viable and recommended management of CFA landfills [261]. Although there are economic advantages and low contamination risk of growing EO producing plants on CFA disposal sites, research on these species is poorly investigated and deserves attention.

10.5. Biomass producing plant-energy resources

The use of energy crops in phytoremediation technology is another innovative way of attaining added value from the process. Energy plants are able to accumulate high biomass in a short period of time, resistant against abiotic stress and can take up toxic substances. Plants producing oil or biomass for biofuel, fibres (e.g. flax, *Miscanthus* and hemp) and quality hardwood can provide financial returns [268]. The forestry sector uses CFA dumping sites for growing trees that have economic benefits such as paper and pulp trees, timber wood, biodiesel crops, etc. as a way to preserve the environment in a sustainable manner [16,195]. Rapeseed varieties have been successfully reported as oil crops and biofuel resources and therefore represent a sustainable option for land use [268]. Seeds and oil are known to be characterised by negligible concentration of toxic metals and therefore food chain contamination is limited. *Helianthus annuus* and *Miscanthus sinensis* have been identified as ideal candidates for both phytoremediation and bioenergy production [273]. Two willow species (*Salix viminalis* and *S. purpurea*) were studied to assess their capability for removal of As, Sb and Pb. Although, they accumulated the contaminants in their roots, these were tolerant toward metal toxicity and demonstrated potential for vegetation cover [274]. Studies have demonstrated promising results for the use of energy crops in the efficient uptake of these REEs minerals [255]. CFA is highly characterised by minerals including Al and REEs, and therefore the aspect of phytoextraction for remediation and recovery of valuable minerals present multi-purpose application of phytoremediation. Leguminous species are known to alter the pH of the growth medium and enhancing metal uptake. These may suggest improved uptake for valuable minerals and therefore more studies are needed to study the impact of low pH on uptake of valuable minerals from CFA. The application of phytoremediation through the use of energy crops have potential for remediation of contaminates sites, while also focusing on the recovery of valuable minerals, therefore future research needs to focus on studying the impact of improved properties of CFA on the recovery of minerals.

11. Conclusion and future prospects

The production of CFA will continue to increase globally due to much dependency on coal combustion for energy generation. Although CFA utilization is reported, more than 200 million tons of CFA remain untreated yearly and problems related to their disposal will continue to pose environmental problems. CFA disposal leads to land degradation, loss of vegetation, worsening of air quality and negatively affecting the health of humans, animals and the environment. The use of phytoremediation strategies by cultivating plants on CFA dumpsites has great potential for reclamation of degraded lands and generation of revenues. An engineered sustainable ecosystem should be developed with the aid of tolerant plant species which would provide vegetative cover. Unfavorable conditions that limit plant development can be overcome by a mix of legumes with other species as they can improve CFA properties to support the growth of plants. In this paper, we recommend that legumes should be included in rehabilitation strategies involving plants for ecological benefits and socio-economic advantages. Organic amendments can overcome problems relating to nutrient deficiency and improved microbial communities, while exploring nanotechnology can assist in enhancing plant growth and reducing toxicity of metals in plants. The use of aromatic plants and energy crops offers an advantage as they produce commercially valuable resources which can convert unproductive lands to valuable sites. Phytotechnology can offer cost-effective tools and environmentally-friendly solutions for restoration of polluted sites, tools to reduce global warming through carbon sequestration, means for recovery of valuable products that can contribute to sustainable development of communities. The insights provided in this review is relevant in guiding future research for restoration of CFA dumping sites, particularly in countries that report low usage.

CRediT authorship contribution statement

Maria Fezile Banda: Writing – review & editing, Writing – original draft, Supervision, Conceptualization. **Dithobolong Lovia Matabane:** Writing – review & editing, Writing – original draft, Conceptualization. **Alexis Munyengabe:** Writing – review & editing, Writing – original draft, Conceptualization.

Data availability statement

The datasets presented in this study can be found in online repositories. The links (DOIs) of the repository/repositories and accession number(s) can be found in the article (reference section).

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.

Acknowledgments

The authors would like to thank the Tshwane University of Technology for supporting this project and the National Research Foundation of South Africa (Grant no. 129752) for funding.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.heliyon.2024.e40741>.

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