Title: Ecological restoration of fly ash-dumped area: Challenges and Opportunities

Running title: Ecorestoration of fly ash deposits

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Abstract

Fly ash (FA) is the 80% of coal burnt by-product of thermal power plants (TPPs), its disposal in landfills causes environmental and health issues. The amount of FA production is increasing continuously to fulfil the worldwide energy for demand, which possibly never find a practically safe method for FA dumping. Its fine particle size disperses in the air and causes air pollution and water pollution is resulted due to slurry erosion from FA dumps and contamination by leachate. Health issues and environmental concerns due to fly ash landfills/dumpsites can be prohibited by covering with phytoaccumulator plant species. Limitations of plant growth in FA includes alkaline pH, contain metals such as Cr, Cd, As, Hg and Pd, toxic level of B, pozzolanic properties of FA and lack of microbial activity. Generally, the phytoremediation process is slow therefore, to accelerate the phytoremediation process FA require organic amendments and bio-fertilizers. This article focuses on the role of naturally occurring plants in stabilization of FA dumpsite and physiochemical changes in FA. This review summarises the different holistic approaches of rehabilitations of FA landfills and also compiles how to convert FA landfills into useful landfills for bioenergy productions. Utilization of organic matter and industrial waste has been proved to provide essential nutrients for plant establishment and heavy metal accumulation. The outcomes of this learning are beneficial for classifying site-specific ecological restoration of FA landfills through holistic approach.

Keywords Coal combustion residue**,** toxic metals, restoration, opportunities, plant responses,

1. Introduction to Fly ash dumps

Fly ash (FA), as a primary discharge from the Coal Thermal power plants (CTPPs), makes use of it in hot steam to generate electricity. It is about 80% of coal burnt by-product (Temuuj*in et al*. 2019) and its production is growing continuously to mitigate the ever growing energy requirements across the globe, which could not yet discover a harmless way of dumping. In general, there are two ways of FA dumping viz. dry ash and wet ash dumping. Wet ash dumping in form of slurry requires large amount water that too leads to shear wastage of another valuable resource (Madhumit*a et a*l. 2018). FA disposed land thus invades and contaminates thousands hectares of valuable agricultural land adjacent to the TPPs over the world. The dry FA disposal contaminates the air and causes air pollution. The fine particles disperse in the air and cause pollution. The water pollution results due to slurry erosion from FA dumps and contamination by leachate (Chakraborty & Mukherjee, 2009). The primary cause of FA pollution is due to toxic heavy metals such as Pd, B, Hg, Cd, Cr present in it (Pande*y et al*. 2014). FA contains hazardous organic pollutants besides radionuclides (Celi*k et al*. 2007; Ruh*l et a*l. 2009). The organic pollutants include dioxin, polycyclic aromatic hydrocarbon, polychlorinated biphenyls, chlorinated benzofurans, and methyl sulphate (Wheatley & Sadhra, 2004; Jambhulka*r et al*. 2018a). These elements bioaccumulate and enter into the food chain and cause serious health issues for the living organisms that need to be remediated.

FA dumps are required to be covered with vegetation so as to remediate such hazardous organic and inorganic pollutants. However, t some physiochemical and biological limitations of FA landfills hinder plant establishment and further growth. Alkaline pH, soluble salts, lack of nitrogen, phosphorus, organic carbon (OC), presence of toxic level of boron, absence of bacterial and fungi activities, and pozzolanic property of FA making in compacted layers are few major limitations of FA that need serious attention (Pandey & Singh, 2010). FA landfills can be converted into concentrated vegetation cover by utilising organic manures and microbial strains during plantation (Mulhern et al. 1989). The properties of FA can be of better-quality by adding of suitable amendments for plant establishment. Additionally, biofertilizers can be used for improving plant- biomass and canopy cover. ??? FA carries nutrients such as organic carbon (OC), nitrogen (N), and available phosphorous (P) for supplementing plant growth and development, and reducing the toxicity of heavy metals in soil (Haynes, 2009). Earthworms are the first colonizers in FA landfills as studied by Eijsackers (2010).

Establishment of plants on such contaminated lands are still very challenging mission for the preservationists, biologist, conservationists and law makers. Nonetheless, it can be achived through repetitive afforestation with different approaches (Singh *et al*. 2011). Transformation of FA landfills in biodiversity rich ecosystem facilitates alleviation of hazardous pollutants. Ultimately, conversion of contaminated land into a resourceful and bioenergy productive land is the requirement of present scenario of the growing environmental issues. This review paper compiles all possible ways to remediate/reclaim the FA contaminated land with different organic amendments and bio-fertilizers. It also focuses on the role of naturally growing plants on FA dumps to pave the way forward for undertaking further afforestation programmes.

2. Physicochemical properties of Fly Ash

The physical and chemical properties of FA are mostly based on the characteristics of the parent coal, combustion method, type of emission device, storage and treatment, and methods of combustion (Ukwattage *et al*. 2013). Based on different types of coals such as bituminous, lignite, and anthracite coals, fly ashes are classified into two categories viz. class F and class C. The classification is according to the presence of silica, alumina, and iron. The burning of lignite and subbituminous coal produces class C ash and the burning of anthracite and bituminous coal produces class F ash. The class C has higher concentration of CaO than class F (Da Costa *et al*. 2004; Ahmaruzzaman, 2010). FA consist of very fine particles with average diameter of less than 10 micron and the average particle size ranges in between 0.01 to 100 mm, and thus FA is easily leachable (Pandey & Singh, 2010). Generally, FA is alkaline in nature and its pH ranges from 4.5 to 12 owing to the presence of S, Ca, and Mg in it. Coal containing higher concentration of S produces acidic pH on combustion, whereas, alkaline pH is due to higher concentration of hydroxide and carbonate salts of Mg and Ca in parent coal. Bituminous and lignite coal have low S concentration therefore, produces alkaline pH of 8.50-9.90 (Weber *et al*. 2015); 9.40, (Pandey & Singh, 2014). Whereas, in India, Jambhulkar and Juwarkar (YEAR) reported 5.01 pH of FA from TPP (Jambhulkar & Juwarkar, 2009).

FA contains essential nutrients such as P, K, Ca, Mg, and S along with heavy metals like B, Ni. Cu, Cd, Cr, Mn, Mo, As, Se and Al. The Ca is dominantly present in FA than Mg, Na and K. Due to smaller particle size, FA is easily leachable for heavy metals and thus become bio-available (Ukwattage *et al*. 2013). FA is devoid of N, organic carbon and available P that are highly essential for plant growth and development (Ram & Masto, 2014). Practically, FA is observed to have changed the electrical conductivity of soil on unweathering of FA than weathering. However, the N, available P and organic carbon contents are low in unweathering FA than on weathering (Gajić *et a*l. 2016). Collectively, it confers that the physicochemical properties of soil changes due the plant species that create favourable conditions for further plantations. The above-mentioned section defines the variability in physicochemical properties of FA that is required before plantation and for undertaking further amendments on land for expected outcomes.

3. Limitation for plant growth on FA dumpsite

Plantation on FA to recover its ecological status is an upsurging strategy being employed across the world. Several successful attempts have been made at different degrees of land-mass cover by FA in various parts of world. Despite of success, the slow processing rate is major drawback of this approach (Pandey *et a*l. 2009). Plants required specific physicochemical and biological environments for healthy growth. Harsh environment of FA creates considerable hindrance in the growth of plant that affects the growth rate and limits the success rate of phytoremediation.

4. Fly ash dumps restoration

4.1 Phytorestoration

Naturally growing plant-based restoration–Phytoremediation is a holistic method which was introduced before few decades ago, using plants for rehabilitation of contaminated land. Although, several chemical and extraction methods are available for reclamation of heavy metal contaminated land but they are not economically suitable for vast contaminated land. Generally, FA landfills do not support plant stabilization due to its harsh physiochemical conditions. In many plants, growth is restricted due to alkaline pH, toxicity heavy metals, lack of nitrogen, lack of organic material and unavailable phosphorus (Pandey and Singh 2010). Still numbers of naturally growing plants survive on FA landfills after few years because of their innate capability. In this aspect, worldwide, many naturally occurring plants have been detected on FA dumps (Pandey 2012b, a, 2013; Gajić et al. 2019). On the bases of phytoremediation, the naturally growing plants have potential for phytostabilization and phytoextraction. Phytostabilization involves reduction of the mobility of metals and phenols in substrate and prevent metal bioavailability in the food chain (Prasad 2006; Nwoko 2010). Itis a productive method that can be functional for heavy metal contaminated land such as, FA dumps. The phytostabilization potential plant species for phytostabilization must have enlarged root structures and capability to arrest contaminants at the rhizosphere level and prevents the passage of metals in above parts of plant (Baker 1981). Phytoextraction is a process in which plants extract heavy metals from the substrate to plants tissue (Keller et al. 2005). Phytoextractor plants are hyperaccumulator that sequester high amount of heavy metals from root to shoot, higher biomass, fast growth and tolerance against heavy metal (Tong et al. 2004). According to the field survey by (Gupta and Sinha 2008), five naturally growing plants *Sida cardifolia, Calotropis procera, Chenopodium album, Blumea lacera* and *Cassia tora* are found to be a phytostabalisers and phytoaccumulators in FA dumpsites. In this study the translocation of metals (Cr, Mn, Fe, Cu, and Cd) was reported in *Sida cardifilia* and metal (Ni, Pb, Co and Zn) in *C. album* it shows that *S. cardifolia* and *C. album* are suitable for phytoextraction of metals in FA landfills. The Phytostabilisers of FA dumps are *Blumea lacera* (Co and Mn), *Cassia tora* (Cr, Cu, and Fe), *Calotropis procera* (Zn and Co), *C. album* (Cr, Mn and Cd), and *S. cardifolia* (Pd, Ni and Co). The maximum bioconcentration factor (BCF) was recorded in *S. cardifolia* and *C. procera*. Several researches in the literature have reported that the naturally growing plants on FA dumps are more potential to sustain and tolerate harsh conditions. Therefore, it has been suggested in many case studies that naturally colonised plants are suitable for phytoremediation/ revegetation of toxic metal contaminated lands (**Table 1).** The maximum of flora studies discovered that a large variety plants are naturally colonized plants and planted on FA landfills for revegetation belong to Asteraceae, Brassicaceae, Chenopodiacea, , Poaceae, Rosaceae and Fabaceae families (Gajić et al. 2019). Leguminous and grassy plantshave fast proliferating and self-improving properties therefore havepotential for revegetation and rehabilitation of FA landfills (Pandey and Singh 2010; Pandey 2015)). Naturally growing plants in FA landfills such as *Saccharum munja, Cassia siamea,* and *R. communis* perform well as primary coloniser for phytoremediation (Pandey and Singh 2011). The above-mentioned plants also accelerate the rehabilitation process on FA landfills and accumulate metal concentrations in plant tissue. According to the restoration potential of naturally growing plants more research is required for identification of the restoration potential of these plants. There are many research studies on remediation of FA dumps by introducing plant species. However, these approaches have limited opportunities due to the long-term process and low possibilities of economic return. Therefore, assisted phytoremediation through naturally occurring and economic plants will be the facilitator for fast green capping of the FA dump sites.

In India, naturally occurring plants on FA deposits are *Acacia nelotica,., Achyranthes aspera. Aerva lanata, Ageratum conyziodes , Amaranthus spinosus , Amaranthus viridis Azolla caroliniana, Bergia ammannioides, Blumea lacera, Calotropis gigantea, Calotropis procera , Cassia occidentalis , Cassia tora ,. Chenopodium album . Croton bonplandianum. Cynodon dactylon, Cyprus esculentus , Dactyloctenium aegyptium Datura metal, Delbergia sissoo, Desmodium triflorum, Dicliptera paniculate, Eclipta prostrata, Eichhornia crassipes, Eragrostis nutans, Eragrostis Pilosa, Eragrostis unioloides, Euphorbia hirta, Fernandoa adenophylla, Ficus racemose, Fimbristylis bisumbellata, Glinus oppositifolius, Holoptelea integrifolia, Ipomoea aquatica, Ipomoea carnea, Kyllinga brevifolia, Lantana camera, Launaea procumbence, Leucas aspera, Lowsonia inermis, Parthenium hysterphorus, Phragmites karka, Phyla nodiflora, Physalis minima, Physalis peruviana, Polygonum glabrmum, Prosopis juliflora, , Sida cordifolia, Sacchrum munja Saccharum spontaneum, Sonchus asper, Solanum nigrum, Saccharum bengalense, Solanum virginianum, Tephrosia purpurea, Tridex procumbens, Typha latifolia, Ziziphus nummularia* (Gupta and Sinha 2008; Rau et al. 2009; Pandey 2012b, a, 2015; Kumari et al. 2013; Pandey et al. 2015a, b; Madhumita et al. 2018; Maiti and Pandey 2020)**.**

Naturally growing plants have been listed across the world. In Australia, the ecologically potential plants on fly ash deposits are *A. hybridus, B. serrata, C. bienis, C. dactylon, C. esculentus, H. contortus, D. eriantha, Eragrostis sp., H. hirta, L. bonariensis, L. cunea, Setaria sphacelate, T. leucothrix* (Van Rensburg et al. 2003;(Morgenthal et al. 2001) *Atriplex, E. tomentosa, Halosarcia, Mesembryanthemum, N. billardieri* and *S. colloris* (Jusaitis and Pillman 1997). Various plants species found on FA dumps are *C acuminatum, E. indica, N. reynaudiana, T chinensis, P. aquiilinum, F. polytrichoides, P. repens* (Chu 2008)**.** In South Africa, plant species identified on fly ash dump sites *are A. hybridus, C. bienis, C. gayana., C. dactylon, Eragrostis sp. C. esculentus, D. eriantha, H. hirta, L. bonariensis, L. cunea, B. serrata, H. contortus, T. leucothrix, S. sphacelate* (Morgenthal et al. 2001; Van Rensburg et al. 2003). Summarise data could pave way to frame restoration strategies at newly form FA catena as per its respective spatial distribution of plants. Furthermore, timely documentation should be the process in newly formed FA dumps with emerging thermal power plant.

Tree planting-based restoration–FA is the waste disposal of TPPs and its quantity is increasing to achieve global demands, which has not yet been able to find the suitable mechanism. The formation of green belt on FA landfills is the ecological approach to cope up with the pollution due to FA. Suitable approach is required to reduce the ecological and environmental issues due to FA by establishing potential tree species around the FA dumps. Such afforestation protects the environment through different ways. Firstly, it supports heavy metals stabilization to prevent leaching of metals to groundwater and secondly it generates bioenergy and lastly increasws carbon sequestration. Thus, the restoration of FA landfills with potential tree species would be an active ecological approach to preserve the environment. After a suitable period, it paves the path in formation of the ecosystem and raises socio-economic standards of the localities. Several studies have focused on the potential of the assisted vegetation and naturally growing tree species on FA landfills as mentioned in Table 2. It is widely known that naturally occurring plants take long time to cover the FA landfill it occurs slowly due to the unfavourable condition of FA landfills (Pandey and Singh, 2012). Therefore, plantation of potential grass, leguminous plant and tree species control soil erosion and facilitate heavy metal uptake by the plants (Pandey et al., 2012, Pandey et al., 2015a). The goal of this learning is to evaluate the revegetation of the tolerant tree species andto expand the knowledge about the rejuvenation status of planted and naturally growing trees under reformed FA ecosystems. The study suggests that the suitable tree species producing higher biomass on FA landfill such as *Acacia*, *P. juliflora* are grown for generating profit. as their growth does not get affected by harsh properties of FA. In this favour, a conceptual research presents the role of afforestation for the mitigation of environmental pressures of FA landfills accompanying advantages for ecological and economic benefits.

4.2 Mycorrhizoremediation

In several studies, the phytoremediation practices have been resulted in unproductive outcomes in decreasing the death rate of planted seedlings due to the lack of essential available nutrients like N, available P, high soluble salt, low soil microbial activity, and pozzolanic property of FA makes cement layers on FA landfills (Selvam and Mahadevan 2002). The lack of symbiotic association between plants and mycorrhizathat increases the nutrient source to the host plants and decrease biotic stress, may be responsible for the difficulties in revegetation (Sylvia and Williams 1992). Mycorrhizal fungi are universal in nature as a soil microorganism that make symbiotic association with plant roots (Smith and Read 2008). Ecologically, Arbuscular mycorrhizal fungi (AMF) play principal part in the rehabilitation of contaminated landfills by enchaining the fertility of polluted land and plant nutrition (Chen et al. 2001). Utilization of potential AMF strains may accelerate the rate of rehabilitation and productivity of phytoremediation process (Vosatka 2001). Mycoremediation is the bioremediation technology to overcome the limitation of phytoremediation. Arbuscular mycorrhizal fungi (AMF) develops the mutual symbiotic association with plant roots and facilitate the uptake of P, several soil nutrients and increases the stress tolerance of host plants (Yang et al. 2016). In the symbiotic association between the fungi and plant roots in which fungi obtain source of carbon from the roots of the plant and it also has been estimated that 20% of total carbon sequestered by the plants are released as root exudates in the rhizosphere (Ely and Smets 2017). In return, fungi promote the phosphate solubilization, fixing N and forming plant growth regulator which accelerate the plant growth and development. Several trials have been conducted to study the role of fungi in phytoremediation of FA dumps (Suhara et al. 2003; Awoyemi and Dzantor 2017). (Selvam and Mahadevan (2002) have isolated and identified 15 Arbuscular mycorrhizal fungi (AMF) species from different plant species grown in FA dump such as *A. leucophloea, A. occidentale, A. indica, C. equisetifolia, T. indica* and *T. grandis*. The isolated AMF were *Acaulospora gerdemannii, Gigaspora sp. G. decipiens, G gigantean, G. margarita, Glomus sp., G. citricola, G. formosanum, G. fasciculatum, G. fulvum, G. mosseae, G. maculosum, G. magnicaule, G. tenebrosum, Scutellospora erythropa, Scutellospora fulgida, and Sclerocystis pachycaulis.* Moreover, few endomycorrhizal association have also been observed in naturally growing plants on the same FA dump site (Selvam and Mahadevan 2002).

Babu and Reddy (2011) have reported different AMF from dominant plants grown in FA deposits in Damnjodi, Odisha, India. The isolated AMF were *Glomus species, G. etunicatum, G. heterogama, G. maculosum, G. magnicaule, G. multicaule, and G. Rosea, and Scutellospora heterogama and Scutellospora nigra*. It has been observed that the AMF diversity varies with the rhizospheres of different plant species on FA. According to the above identified AMF Glomus fungal species is abundantly present in FA dump and tolerant to FA (Babu and Reddy, 2011). Furthermore, the inoculated plants with AMF improves the plant growth, chlorophyll content, and total P content of *Eucalyptus tereticornis* saplings grown on FA, and decreased Al, Cu, Zn, and Fe translocation. Therefore, it is suggested that the utilization of FA adapted AMF supports the remediation of contaminated sites. Moreover, the researchers at The Energy and Resources Institute (TERI) proposed mycorrhizal mediated reclamation technology. They have isolated, and multiplied different strains of AMF under green cover conditions (Das et al. 2013). The isolated and multiplied AMF along with farm yard manure (FYM) were inoculated with plants on three different FA dumps at 200 infectious propagules rate per plant (Das et al. 2013). It improved the physico-chemical and biological properties significantly by accepting mycorrhiza technology. There were manyfold increase in organic matter and rise in microbial activitiesy (Das et al. 2013). Hrynkiewiez et al. (2009) conducted the pot experiment and estimated the Salix plant growth inoculated with a mycorrhiza-associated bacterial strain (*Sphingomonas sp.* 23L) and observed enhancedshoot and root growth of a *S. viminalis* x *S. caprea* hybrid. The inoculation with mycorrihza promoting bacterial strains might be a suitable method to promote mycorrhizal formation with autochthonous site-adopted ectomycorrhizal fungi in FA and thus to progress re-vegetation of FA dumps with *Salix spp*. (Hrynkiewicz et al. 2009).

Another research study conducted by Ultra (YEAR) on *Delonix regia*, in pot experiment by planting *Cymbopogon citratus* in FA with different amendments such as mycorrhiza, NPK fertilizers, compost and there the different combinations. NPK alone resulted manyfolds of yield and growth than control.Compost resulted in lowering the metal availability andprovided nutrient availability while mycorrhiza improved the nutrient and water uptake. Combined utilization of NPK, compost and mycorrhiza decreased the concentration of Cr, Cu, Ni, and Pb in shoots and in essential oil. Overall, the combined application improves the phytostabilization potential of *C. citratus* in FA (Ultra, 2020). Inoculation of *Rhizophagus fasciculatus* fungi in *Paspalum scrobiculatum* with different doses of FA.

The plant growth improved with FA doses of 20 mg kg^{-1} with inoculated AM fungi (Channabasava et al. 2015). Results depicted that both FA amendment and AMF inoculation significantly improved the number and weight of grains and suggested that the inoculation of AMF could be beneficial for rehabilitation of FA dumps.

In another research study (Ultra and Manyiwa, 2020) conducted to evaluate the growth and phytoaccumulation capacity of three dominant *Acacia species* in mine tailing amended with FA and mycorrhiza. The result of this study revealed that mycorrhizal inoculation boosted the existence of *A. luederitzii*, and *A. albida* and reduced the *A. tortilis* survival in mine tailings and the combined mycorrhizal inoculation and FA amendment boosted the establishment of *A. luederitzii* (Ultra and Manyiwa 2020). The symbiotic association of mycorrhiza and plants root has powerfully co-evolved over a time and depends on each other. The AMF support the root system enlargement of the host plants and improve their nutrient up-take capacity such as N, P and water more efficiently than roots alone. In summary, literature reports evidenced that AMFs isolated from heavy metal contaminated area show high tolerance and hence, can be utilised for phytoremediation of FA dumps. Mycorrhizal symbiotic association can greatly improve host plant heavy metal tolerance by improving plant nutrient acquisition and by inducing the fate of the metals in both the plants and substrate (Hrynkiewicz et al. 2009).

4.3 Bacterial mediated remediation of FAdumps

FA disposal and dumps are the serious problems across the world. Phytoremediation is a holistic approach to remediate and improve it when coupled with microbes (Ullah et al. 2015). Various plant growth promoting bacteria (PGPB) have been isolated from naturally growing plants on FA dump-sites as detailed in Table 1. The Phytobacterial association is more productive than the phytoremediation alone in removal of heavy metals in stressful condition. *Enterobacter sp.* and *Bacillus sp.* are predominantly present in FA dumps that aretolerant to heavy metals (Tiwari et al. 2008; Kumar et al. 2009; Kumari and Singh 2011; Raja and Omine 2013; Mukherjee et al. 2017). Plant growth promoting Rhizobacteria (PGPR) efficiently enhance yield and growth of plants by N fixation and acceleratedgrowth hormone generation *i.e.* indole-3-acetic acid (IAA) thatimprovesthe availability of plant nutrients by producing siderophore and solubilising phosphate. Ethylene hormone has inhibitory role on root system and plant growth (Lv et al. 2018). PGPR lowers the ethylene concentration through release of 1-Aminocyclopropane-1-carboxylic acid deaminase enzyme which favours in plant growth in stressed conditions (Ullah et al. 2015; Chen et al. 2017). Soil is rich in P in unavailable form and plants cannot absorb it directly. Bacteria release organic acids that promote the P solubilization. Phosphate-solubilizing bacteria include *Rhizobium sp.*, *Bacillus sp.,* and *Pseudomonas sp.* (Chen et al. 2006). The studies mentioned in Table 3. describe that the FA associated microbes are well adapted to heavy metal contaminated sites and increase the bioavailability of metalloids from FA to complement phytoremediation.

The habitat of microorganism is mainly in the rhizospheric region of plants which play important role in solubilization of organic and inorganic pollutants. Inoculation of PGPR and AMF in revegetation is beneficial because they have incredible ability to improve plant growth, increase biomass and defendplants from phytotoxicity. It is required to isolate /study/ multiply such bacteria and fungi tolerant to harsh substrate. Active metabolism of microflora in response to organic extract from roots subsidizes the rhizofilteration. It can be determined that potential strains of bacteria and fungi have the ability to access metals that should be included in the consortia for further progress in phytoremediation of contaminated and barren land. However, phytoremediation is slow and spontaneous process and takes long time for desired results however through this approach rehabilitation speeds up. Along with it selection of native plant species is desirable in phytoremediation and research should focus on elevating the effectiveness of native varieties by identifying and studying microflora from the rhizospheric microorganism of these plants throughseveral biotechnological approaches.

4.4 Organic amendments

FA dumping is a serious problem for the environment which causes soil, water and air pollution. Therefore, it is required to restore the FA dump-site with phytoaccumulating plant species to control the heavy metal leaching to the groundwater and dispersion of FA into the air. Although, FA contains micro nutrients, but are devoid of essential macro element such as N, available P and organic carbon (Pandey and Singh 2010). Therefore, plants face difficulties to cope with harsh physiochemical condition of FA. Due to the lack of organic matter in FA, it is necessary to add organic matter into the FA to maintain the nutrient cycle and also to maintain the concentration of N, available P and organic carbon (Jambhulkar and Juwarkar 2009). Reforestation is tough on FA dumps unless the organic amendments and bio-fertilizers are applied (Pandey and Singh, 2010). A number of field trials and green house experiment have been performed with selected plant species and with various organic and industrial waste amendments on FA landfills worldwide andthe results of various studies have been discussed in (table 4). Some organic amendments such as vermicompost, farm yard manure, animal manure, industrial wastes, sewage sludge, and press mud are well proven for the making of FA suitable for plant growth (Ram and Masto 2014). Various studies have been proposed with positive role in reclamation and revegetation of FA dumps and have been put forth for effective remediation such as 1) suitable plant species 2) nutrient rich organic amendments and industrial waste, and 3) bio-fertilizers (Pierzynski et al., 2004; Gupta et al., 2007).

5. Morphological responses

The study on the morphological character of plant adaptation on contaminated habitat is required for revegetation of plants at disturbed lands (Pandey et al. 2020). According to the studies, the unfavourable properties of FA for the plant growth such as high pH, lack of N may significantly affect the morphology of plants. The naturally growing plants *Withania somnifera* at Badarpur thermal power plant (BTPP) resulted in significant decrease in the leaf morphology such as leaf length, leaf area and leaf width (Qadir et al. 2020). Similarly, the leaf length, leaf area and leaf width of naturally growing *Azadiracta indica* presented a decreasing trend from the control site near to BTPP. The leaves of *A. indica* also showed chlorosis and necrosis (Qadir et al. 2016). In contrast, the study of two Betula species growing on FA landfills shown 1.5-2 folds more in leaf area ratio and leaf mass ratio compared with the Betula species growing in forest near FA landfills. This reflects the growth of a larger leaf assimilating surface per tree. Leaf thickness was higher in both Betula species on FG dump sites (Kalashnikova et al. 2020). Due to the presence of toxic concentration of As, the morphological impairment has been observed in leaves of *D. glomerata* grown on FA dump site in the form of chlorosis like yellowing and light green as well as leaf necrosis, wilting, reduction in root and leaf growth (Gajić et al. 2020). These plant and contaminated landfills interactions convert the plant’s metabolism and leaf morphology to adapt in new habitats. Therefore, plants can be used as a bio-indicator and bio-itigator for environmental contamination.

6. Physiological responses

Wide gaps about the knowledge related to the potential of the physiological tolerance of the invasive plant species towards the harsh macro and micro- climatic stress of FA deposits. More concentrated screening of physiological response of naturally growing plants on FA dump site for ecorestoration and revegetation of contaminates landfills. The plants grow in stressful environmental condition such as low and high temperature, lack and excess of heavy metal, deficiency of essential nutrients, and salt stress have great distinctions in their physiological mechanisms. These physiological mechanisms are photosynthesis, respiration, relative water content, water saturation deficit, sap osmosis, chlorophyll, and carotenoids get effected due to environmental stress (Pandey et al. 2020). Physiological potential/responses have been observed in naturally growing plants on FA deposits. Physiological measures have been observed in naturally occurring plants on FA dump-sites such as *Typha latifolia, Ipomea carnea, Prosopis juliflora, and Saccharum spontaneum. Saccharum spontaneum* taht are most effective in carbon sequestration it which reflected its maximum photosynthetic rate (29.29 ± 0.46 μmol CO_2 $m^{-2}s^{-1}$). The maximum water use efficiency (9.3 ± 0.5 mmol CO_2 mol H_2O^{-1}), high carboxylation, the lowest transpiration rate (3.15 ± 0.13 mmol H2O m^{-2} s^{-1}) and stomatal conductance (0.2 ± 0.004 mol H_2O m^{-2} s^{-1}) as compared to other three species (Pandey et al. 2020). Chlorophyll content were low at 50 and 100 % FA in *Cicer arietinum* but carotenoids concentration improved (Pandey et al. 2010a). Also, chlorophyll and carotenoid level reduced in leaves of *Lolium perenne* in FA amended soil (Lopareva-Pohu et al. 2011). Lower content of carotenoids and chlorophyll reflects the sensitivity of photosynthetic pigments towards the harsh stress factors of FA.

However, movement of Mg in the chlorophyll molecule structure with metal ions leads to the chlorophyll in non-functional molecule (Van Assche and Clijsters 1990). Moreover, less content of carotenoids in plants can be advised in decreased PSII which leads to the dropping of photosynthetic efficiency of plants (Gajić et al. 2016). The negative impact of 100% FA amended with *L. leucocephala grown* revealed less content of chlorophyll, carotenoids, protein, and in-vivo nitrate reductase activity as compared to 100% soil and FA and 50% press mud amendments (Gupta et al. 2000). The efficiency of photosynthetic unit PSII in plants were lower than the optimal range of plants growing on FA landfills (Björkman and Demmig 1987). The lower value of PSII reaction centre has been observed in various plants species grown on FA landfills such as

Epilobium collinum, Eupatorium cannabinum, Verbascum phlomoides, Cirsium arvense, Calamagrostis epigejos, Crepis bienis, Oenothera biennis, Festuca rubra and tree species Populus alba, and *Robinia pseudoaccacia and shrub species such as Tamarix tetandra, Amorpha fruticosa,* and *Spiraea van-houttei* (0.429–0.727) (Mitrović et al. 2008; Pavlović et al. 2004) Gajić et al. 2013) (Gajić et al. 2016), (Kostić et al. 2012). Oppositely, *Festuca rubra* growing for 11 years on FA lagoons showed improved photosynthetic efficiency (PSII) thatreflected the adaptive nature that increases the tolerance of the photosynthetic apparatus to stress after some time (Gajić et al. 2016). Moreover, the relative water content has been observed high in plants growing on FA landfills such as *Verbascum phlomoide, Crepus setosa, Epilobium collinum,* and *Eeupatorium cannabinum* but it was low in *Populus alba* (Pavlović et al. 2004). The relation between the plant physiology and eco-restoration offers the new point of view on the association between plant physiological response to stressful environment condition and ecorestoration. Physiological and biochemical tools can be used to predict plant response to stressful environmental conditions and towards the success of restoration projects.

7. Antioxidant responses

In several studies, the response of antioxidants has been observed to fight against the oxidative stress due to the overproduction of reactive oxygen species (ROS). FA, consisting of metals and metalloids like Fe, Ni, Pb, Cr, Pb, Cu and Cd are the alarms of toxicity which not only pollute the environment but also result in oxidative stress with the production ROS such as peroxide and superoxide radicals (Qadir et al. 2019). It creates oxidative stress by electron transfer and inhibition of metabolic reaction (Pandey et al. 2010b). The raised level of Malondialdehyde in plants is the indicators of ROS production which is the authentic key to measure oxidative stress level in host plants (Randjelovic et al. 2016). Plants have a defence mechanism to control the ROS through enzymatic and non- enzymatic antioxidants (Hossain et al. 2012), and also, the plants grow in contaminated and toxic metal substrate having high antioxidant capacity (Randjelovic et al. 2016). In several studies, the role of antioxidants have been noticed due to the presence of higher concentrations of As enhanced the concentration of malondialdehyde in the leaves of *Festuca rubra* grown on FA deposits (Gajić et al. 2016), *Zea mays* [(Stoeva et al. 2003), *Adianthum capilillus-veneris* (Singh et al. 2010).

In a comparative study of plants growing in Badarpur Thermal Power Plants (BTPP) with non-polluted site by Qadir et al. (2020) the plants indicated the signs of oxidative stress by higher levels of malondialdehyde and electrolyte leakage from cells. Furthermore, the oxidative stress has been observed in *Withania somnifera* through noteworthy and higher production of reactive oxygen species (ROS). In this study, the ability of *Withania somnifera* to well coordinate ascorbate peroxidase, glutathione reductase, superoxide dismutase, and activities involved in the foraging of ROS along with the enhanced increment of nonenzyme activities (total ascorbic acid, proline, and oxidized glutathione) could be related to FA stress tolerance in *W. somnifera* (Qadir et al. 2020). In the observation of field survey on FA dump site near Nikola Tesla Thermal power plant the antioxidants properties have been noticed in *D. glomerata*. The level of Malondialdehyde, is higher in leaves that can occur due to the presence of As in leaves and the content of total phenolics. Ascorbic acid is higher in leaves and root of *D. glomerata* sown on FA dump as compared to the control site. The activation of antioxidant properties reflects the stress condition in plants growing in FA deposits. The DPPH scavenging activity is also higher in the roots of *D. glomerata* growing on FA deposits (Gajić et al. 2020). Though, higher concentration of Malondialdehyde indicates the sign for stimulation of antioxidant biosynthesis increasing the total phenolics, ascorbic acid, and scavenging capacity, which in turn provides recovery of PSII photochemistry and systematic electron transfer that supports in reducing the oxidative stress (Gajić et al. 2020). These studies indicated that the above-mentioned plant species i.e. *W. somnifera* and *D. glomerata* are potential species for phytoremediation of FA landfills and other contaminated sites. However, more studies and research are required to know about the potential plant species whereas to identify their role in oxidative stress that occur due to the contaminated substrate. The lack of explanation of complete mechanism of the role of antioxidants during the stressful condition is urgently required.

8. Opportunities in fly ash dump restoration

Several prospects exist for improving FA landfills quality by application of different organic amendments such as FYM, press mud, sewage, sludge, cow dung, vermicompost and bio-fertilizers that improve the physicochemical properties of FA (Pandey and Singh, 2010). Phytoremediation is a low input approach that decreases the greenhouse gases production and accelerates the carbon sequestration (Pandey et al. 2016b). Combined utilization of potential mycorrhiza and bacteria would be effective in establishment of plants in harsh conditions of FA landfills and also support in dropping the level of HMs (Lewandowski et al. 2003; Dzantor et al. 2013; Pandey et al. 2016a; Pandey 2017). Continuous rejuvenation of FA dumping area with biomass production and accumulation of heavy metals through phytomanagement is a holistic approach that will also help in climate change mitigation (Pandey et al. 2016a). FA landfills restoration promotes the enhancement of above and below ground biodiversity. The economically and ecologically valuable plant-based green technology such as phytoaccumulation and phytostabilization not only covers the FA dumps with the reduction of the toxic effect of HMs in FA but also generates revenue through phytoproducts (i.e. essential oil, biomass, biofuels, biodiesel, fibre, wood biomass for paper and packaging material, etc.) produced from phytoremediation (Pandey 2017; Pandey, 2020).

9. Future prospects and conclusion

Revegetation/rehabilitation of huge FA landfills is the present research priority. FA landfills can be utilised for both ecological and socio-economical purposes. The ecological purposes can be solved by utilising of potential plant species for phytoremediation which have good biomass, higher N_2 fixing capacity and flourishing the microbial diversity of rhizosphere and soil. The socio-economical purpose is dependent on biomass production and market value of the biomass of the planted bioenergy crops. Ther is a need to create general awareness among the localities and farmers for utilization of such contaminated landfills into bioenergy crop for economical balance. To achieve ecological and socio-economic benefits more field scale work are required with analysis of both environmental issues and cost: benefit ratio. In biological point of view, isolation and identification of potential microorganism from re-vegetated FA landfills with the genomic markers is the latest dimension of upcoming research. This combined study will help to understand the physiological and molecular levels to attempt the proper scientific method for restoration of FA landfills. The expected outcomes of this review paper compile different holistic approaches to accelerate the phytoremediation process. As it is well known that the remediation is very slow unless a suitable approach is undertaken to accelerate the remediation process. The suitable approache involves the utilization of organic amendments, bio-fertilizers and potential plant species. More research work needs to done on field scale to explore the outcomes of amended plantation. Flora and fauna play active role in rehabilitation of the FA landfills. Therefore, regular monitoring of the successions of the on-going rehabilitated FA landfills for better understanding towards stability and sustainability is indispensable. However, a very fewer studies are traceable on the bioindicators of the remediated FA landfills.

Abbreviations FA, Fly ash; CTPPs, Coal thermal power plants; CEA, Central electricity authority; VAM, Vesicular arbuscular mycorrhiza; OC, Organic carbon; MTs, Million tonnes

Acknowledgement The authors are thankful to the Director, CSIR-NEERI, Nagpur, India for encouragement and providing necessary facilities.

Authors' contributions Swati Yadav (compiling and writing—original draft), Vimal Chandra Pandey (conceptualization, visualization, writing—review and editing), Lal Singh (writing—review and editing).

Competing interests The authors declare that they have no competing interests.

Funding information No external funding was received for this review assignment.

Data availability statement The authors confirm that the data supporting the findings of this study are available within the article.

Conflict of Interest There is no conflict of interest among the authors.

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Figure captions

Figure 1 Bamboo plantation on fly ash dumpsite near Koradi Thermal Power Plant (Nagpur, Maharashtra) (A) Fly ash dumpsite before revegetation (B) Rehabilitated fly ash dumpsite after one year

Figure 2 Toxic metals enriched fly ash in the environment and their impacts on flora and fauna