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Pedological properties and ecological implications of substrates derived 3 and 11 years after the revegetation of lignite fly ash disposal sites in Serbia

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ABSTRACT

The revegetation of disposal sites of lignite fly ash (FA) offers the best way to overcome the extremely serious problems which their deposition via sluicing into settling ponds can pose for the environment and human health. Even so, plant survival and development is limited by toxicity and the highly unfavourable physical, chemical and nutritive characteristics of FA. Hence, initiating soil formation processes at the ash deposit sites is of inestimable significance for the success of this reclamation process.

This case study investigates the influence of weathering and vegetation development on changes in selected physical and chemical properties of FA, as well as assessing the environmental risks of FA at three deposit lagoons (L0, L1 and L2) at the 'Nikola Tesla-A' thermal power plant in Obrenovac, Serbia, weathered for 0 (raw ash), 3, and 11 years respectively. The raw FA is characterised by a sandy texture, alkaline pH, high electrical conductivity (EC), a low cation exchange capacity (CEC), and low total nitrogen (N) and available P and K content, while the content of elements such as As, B, Cr and Cu is in the critical range for plants. Weathering and revegetation processes have brought about an increase in the clay and silt fraction, a reduction in alkalinity and salinity, and an increase in the cation exchangeable capacity (CEC) and N, P and K content, particularly in the surface layer (0–10 cm) at L2. Even though these changes point only to the initiation of soil formation processes, they lead to colonisation and in later stages an increase in the diversity of spontaneously colonising plants (55 species at L1 and 80 species and a greater value on the Shannon index of diversity at L2). At the same time, a reduction in the total content of As, B, Cr, Cu, Mn, Ni and Zn and their mobility was noted, as well as in individual and total contamination of FA over time. In this regard, our study, conducted at a large coal ash disposal site, can contribute to knowledge on the effects of weathering and the development of vegetation directly on ash itself and on changes in the physical and chemical properties of ash as important indicators of soil initiation and development on this substrate, with these processes being of exceptional importance for the successful ecological reclamation of ash deposit sites.

1. Introduction

Fly ash (FA) as a by-product of coal combustion is a hazardous material, dumped at open-air sites (either by wet or dry methods) due to its low utilization. According to some estimates, this type of disposal site covers approximately 3235 km^2 of fertile, potentially agricultural land across the world (Pandey and Singh, 2012). The possibility that fine particles of FA will be dispersed into surrounding habitats and toxic material leached into groundwater means that these disposal sites are a constant source of pollution and a serious global environmental and ecological threat for air, water and soil (Weber et al., 2017). A cost-effective and non-invasive method centred on the planting of grass, shrubs and tree species, which ensures the ash's physical and chemical stabilisation, has been favoured over the last few decades for the mitigation of environmental problems and the ecological reclamation of degraded areas arising from fly ash deposition (Pandey et al., 2012; Haynes, 2009; Mitrović et al., 2008; Pavlović et al., 2004). Even so, physical and chemical limitations (unfavourable texture, phytotoxicity due to high pH and high contamination of micro elements, high content of soluble salts, deficit of essential elements N and P, and reduced microbial activity) inhibit plant establishment and survival (Ram et al., 2015; Haynes, 2009; Mitrović et al., 2008; Pavlović et al., 2004; Gupta et al., 2002). Besides offering physical protection, vegetation development at ash disposal sites also contributes to increasing the content of organic matter in the substrate through the input of litter and fine roots. Organic matter improves the physical (water holding capacity,

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aeration, temperature, and structure) and chemical (cation exchange capacity and nutrient content) properties of the substrate (Arocena et al., 2010), and through its decomposition and mineralisation, nutrient content and flow is regulated, meaning the nutritive and biological characteristics of this substrate improve significantly over time (Pandey and Singh, 2012; Walker and del Moral, 2009; Djurdjević et al., 2006). Initial vegetation improves soil conditions for colonisation by subsequent species and hence, for the further successful development of the ecosystem, knowledge of how soil develops during the ecological reclamation process is of particular importance (Frouz et al., 2008). The importance of initiating soil formation processes is particularly great if revegetation takes place directly on ash, without it first being covered with a layer of soil in order to reduce costs (Zhao et al., 2013; Alday et al., 2012; Arocena et al., 2010; Zikeli et al., 2002). However, soil formation on ash deposits is a very slow and long process (spanning several decades) (Weber et al., 2015; Alday et al., 2012; Šourková et al., 2005; Shaw, 1992), with the following parameters the best indicators of its quality: texture, pH, EC, properties of the adsorption complex, total organic carbon (C) and nitrogen (N), soil organic matter and nutrient availability (Uzarowicz et al., 2017; Weber et al., 2015; Courtney et al., 2009).

The total metal content, which in FA is often at excessive levels, is a significant indicator of the total level of contamination, but it alone is not enough for a true assessment of the actual ecological risk FA poses to the living world. The leaching and bioavailability of elements is affected not only by their total content, but also by their different chemical forms, their binding state, metal properties, environmental factors, organic matter content and substrate properties (Zhipeng et al., 2015; Chaudhary and Banerjee, 2007; Jankowski et al., 2006; Bódog et al., 1996). This means that the content of mobile metal fractions in FA reflects the most important characteristics of the substrate and represents the soil equilibrium (Gupta et al., 1996). The management and long term disposal of FA in the open exposes it to the constant influence of natural factors, leading to changes in its physical, chemical and mineralogical characteristics (silt and clay content, pH, EC, soluble salt content, cation exchange properties, and organic matter content), which can have a significant impact on the behaviour of contaminants and the leaching and mobilisation of FA species (Eze et al., 2013; Zikeli et al., 2004). Furthermore, the cementitious or self-hardening properties of raw FA when reacted with water reduces its porosity and increases the length of its exposure to weathering factors (Weber et al., 2015; Ahmaruzzaman, 2010; Haynes, 2009). For this reason, application of a three-step sequential extraction procedure, by which metals are fractionated into the acid soluble/exchangeable (F1), reducible—Fe and Mn oxide bound (F2), oxidisable—organic matter bound (F3), and residual—lattice bound (F4) fractions through imitating the various extraction processes that can occur in the environment, allows for a prediction of their behaviour under environmentally relevant conditions (Krgović et al., 2014; Jankowski et al., 2006; Querol et al., 1996; Ure et al., 1993). All the phases except the residual one can become available to biota under changing environmental conditions (Chaudhary and Banerjee, 2007).

Most studies of revegetation have focused on the choice of species and techniques for vegetation establishment (Żołnierz et al., 2016; Pandey, 2015; Kostić et al., 2012; Mitrović et al., 2008), while changes in the physical and chemical characteristics of the substrate during deposition and revegetation, as well as soil development and the impact of these changes on the behaviour of contaminants has not been given enough attention. There has been a lot of research recently into changes in soil parameters during vegetation development and vegetation succession, but this is mainly focussed on other types of coal and metal mine waste sites, while such research for lignit ash deposit sites is very rare (Uzarowicz et al., 2017; Weber et al., 2015; Zhao et al., 2013; Alday et al., 2012; Arocena et al., 2010; Djurdjević et al., 2006; Zikeli et al., 2002).

The aim of this study was to describe and evaluate the effects of weathering and vegetation on the development of substrate properties, as well as the environmental implications of FA deposition at the disposal site of the largest thermoelectric power plant 'Nikola Tesla A' (TENT-A) in Serbia. For this reason, a chronosequence of FA lagoons (FA aged 0, 3 and 11 years) was selected to determine property changes. Hence, the detailed objectives of this study were to establish the changes at FA lagoons of different ages in terms of: (1) plant species composition, richness and diversity, (2) the main physical and chemical properties of FA, (3) concentrations of selected chemical elements (As, B, Cr, Cu, Mn, Ni and Zn) in FA, and (4) the mobility and availability of selected elements including an assessment of contamination levels and the environmental risk.

2. Materials and methods

2.1. Site description and fly ash sampling

Research was carried out at the fly ash deposit site, which is located in the municipality of Obrenovac (lat. 44°30′ N, long. 19°58′ E, average altitude 80 m), on the right bank of the River Sava, 41 km upriver from Belgrade, the capital city of the Republic of Serbia. This region is characterised by a moderate continental climate with a mean annual temperature of 12.5 °C and mean annual precipitation of 690.1 mm.

TENT-A consists of 6 generator units with a combined capacity of 1726.5 MW. Each year it burns an average of 12×10^9 kg of low calorie lignite, from the open-cast mines in the Kolubara Coal Basin, and produces in excess of 8×10^9 kWh of electrical energy. Approximately $2.2–2.5 \times 10^9$ kg of ash are produced as a by-product of lignite combustion. To date, almost 66×10^9 kg of ash and slag has been deposited at the disposal site, which was designed to hold 112×10^9 kg of waste material. In terms of its chemical characteristics, the electrofilter ash produced by lignite combustion is aluminosilicate (approximately 80%) with a significant proportion of Fe, Ca, Mg and K oxides (approximately 16%) and toxic content of As, Cr, Cu, Ni and Zn (Supplementary information, Table S1).

Coal ash (FA and bottom ash) is deposited over an area of 400 ha. Of the three lagoons, which are of approximate equal size, only one is active (settling pond—L0) at any one time, while the other two are temporarily passive (L1 and L2) (Fig. 1).

The ash is mixed with water (1:10, but also 1:15 in practice) and transported as slurry through pipes to the active lagoon. The coarser fraction (bottom ash) is separated out by means of hydrocycling and is used to form the side dyke, while the finer fraction (FA) runs into the active lagoon in the form of a pulp. The spread of the pollutants from the ash deposits is exacerbated by north-western and western winds that carry the ash towards the surrounding settlements, the central zone of the municipality of Obrenovac and agricultural fields (Ćujić et al., 2016). Dispersal of ash from the deposit site is prevented by maintaining a layer of water at the active lagoon, wetting the ash with the aid of 80 water cannons and 124 sprinklers, and implementing a process of revegetation at the passive lagoons (MP TENT, 2004).

Revegetation is undertaken through the sowing of a grass-legume mixture directly onto the ash (without the application of topsoil), with the use of agronomic measures (800 kg/ha^{-1} of 15N:15P:15K mineral fertilizer and the wetting of the seeded area until the formation of plant cover), (MP TENT, 2004). The grass-legume mixture (sowing density: 270–300 kg/ha) comprises grasses tolerant to the unfavourable chemical characteristics of the ash and the unfavourable water regime (*Secale cereale* L., *Arrhenatherum elatius* (L.) P. Beauv., *Lolium multiflorum* Lam., *Festuca rubra* L., *Dactylis glomerata* L., *Vicia villosa* Roth., *Lotus corniculatus* L. and *Medicago sativa* L.). Once grass cover has been established, during the second or third year, cuttings of tamarix (*Tamarix tetrandra* Pall.) are planted within the lagoon, while the side dykes are further stabilised by planting cuttings of *Robinia pseudoacacia* L., *T. tetrandra* Pall., *Populus × euramericana*/Dode/Guinier and *Salix alba* L. The ash deposition process at the disposal site is cyclical, which