Low-Rank Coal as Humic Substances

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Humic substances (HS), as important environmental components, are essential to soil health and agricultural sustainability. The usage of low-rank coal (LRC) for energy generation has declined considerably due to the growing popularity of renewable energy sources and gas. However, their potential as soil amendment aimed to maintain soil quality and productivity deserves more recognition.

low-rank coal brown coal soil amendment Humic substances

1. Low-Rank Coal (LRC) Types and Properties

Coal is considered one of the world’s most abundant and most important fossil fuels for power generation [1]. There are different types of coal that are characteristically distinct in a few specific features, such as origin, composition, and coalification level [2]. Brown coal, known as lignite, and sub-bituminous coal are classified as LRC due to the short formation time and low-grade metamorphism. Both have relatively low heat value and high ash content [3]. In addition, LRC, especially brown coal has a high moisture content, in the range of 25–65%, most of which exists as free water that rapidly evaporates under dry conditions [4].

The natural oxidation (weathering) of brown coal takes place on a large scale when the coal is in the seam or occurs during transportation/storage and significantly affects its physical properties and chemical composition [5]. As a result of oxidation, the valuable properties of fossil fuels deteriorate leading to extremely fast fragmentation and low calorific value. The resulting type of coal is called leonardite, named after Arthur Gray Leonard in recognition of his research contribution [6]. The interaction of coal with the atmosphere is a cause of great concern for the power sector and industry due to its gradual destruction, dispersion, and redeposition [7]. Moreover, LRC may combust spontaneously during mining and utilization, thereby causing air-polluting emissions [8]. Up to date, thousands of hectares of previously-fertile land functioning ecosystems are disturbed by coal mining and coal waste [9].

2. Impact of LRC on Soil Quality and Health

The application of LRC and its derivatives to agricultural soil is becoming a very common practice. Due to high levels of SOM in LRC, there is steady great interest in its use as a soil amendment and conditioner. Many studies have traced a wide range of benefits of applying LRC and its derivatives provisioning for physical, chemical, and biological functions of soil in mainly short-term practice. It is important to note that the labile and humified organic matter of LRC has a decisive impact on soil health and fertility [10][11].

The SOM of LRC is characterized by its high content (<90% d.w.) of humic substances (HS) [12][13]. HS are mixtures of humic acid (HA, only soluble in water under alkaline conditions), fulvic acid (FA, soluble in water under all pH conditions), and humin (HM, neither soluble in alkali nor in acid) [14]. HS can be extracted from coal using alkali, acids, and organic solvents [15]. HS are relatively stable complexes and display diverse functional groups that help to create a healthy soil environment by improving soil aggregation, microbial activity, enzymatic functionality, carbon sequestration, nutrient retention, and pollutant immobilization [16][17]. The LRS-specific HS exhibit more carbonyl carbon (about 16%) and less aliphatic carbon (27%) compared to the typical soil-specific HS, containing about 11% and 31% respectively [12].

2.1. Effects of LRC on Soil Physical Properties

LRC accrue benefits for soil structure by enhancing its water retention ability, aggregate stability/porosity, aeration, and bulk density. The water holding capacity of brown coal HS due to its partial hydrophilicity and porous character is well-understood [18]. Piccolo et al. showed that coal-derived HS can improve the structure and water retention of degraded arable soils and argued that the higher the HS content, the better the water retention of soil [19]. Cihlar et al. [20] suggested that modification of brown coal HS by formaldehyde cross-linking may provide an effective strategy for achieving high water uptake kinetics. Oxidation may enhance the HA content of coal sources to be used as soil conditioners. Two independent experimental studies showed that nitric acid (HNO_3) oxidation of brown coal leads to the increase of HA content with richer functional groups and ensures the retention rate of nutrients, which consequently improves soil aggregate stability and associated structure [21][22].

Brown coal-derived humic acid can reduce the disaggregating effects of cyclic wetting and drying on soil structural stability [23]. Soil porosity, an essential component of the soil skeleton structure and site productivity, can be maintained after coal mining by conducting site reclamation [24]. Being a rich source of carboxylic acid and phenolic hydroxyl functional groups, HS can provide reactive sites, increasing the CEC and the pH buffering of soils [25]. The high CEC of brown coal results in greater retention of NH_4^+ and consequently lowers the NO_3^- leaching loss [26].

Most of lignin oxygen-containing functional groups result in a low pH level when ionized in solution [27]. For this reason, LRC can be rather effective in neutral-to-alkaline soils. However, in combination with lime, brown coal is well suited for application to soils with a low pH. Imbufe et al. [28] have found LRC humates to be effective for increasing pH and electrical conductivity in acidic soils. Further acid ameliorating effects by LRC have been studied in different conditions [29]. According to another recent report, the LRC at a dose of 5 kg m^{-2} contributed to the decrease of electrical conductivity and sodium adsorption ratio of saline-sodic soils, whereas pH levels and bulk density displayed no significant changes [30].

2.2. Effects of LRC on Soil Organic Matter

The high content of TOC and its relatively slow mineralization suggest LRC be attractive for increasing plant nutrient supply in the soil the same way as known organo-mineral fertilizers [21]. The application of LRC by B. Dębska with colleagues [31] resulted in an increase in TOC content (by ~300%) and elevated soil organic carbon with higher aromaticity (38.6% compared to 35.4% in controls), which implies higher C sequestration potential and recalcitrance. Along with LRC, the LRC-derived humic acid products can outperform conventional organic wastes such as farmyard manure (FYM) in ameliorating soil quality and fertility [32]. Enhancement of SOC content and sequestration following LRC-derived HS is well-documented by R. Spaccini et al. [33].

2.3. Effects of LRC on Soil Heavy Metals and Other Pollutants

The good adsorptive properties of LRC have aroused intense interest for its potential as a versatile environmental adsorbent. The utilization of the coal-based HS in soil remediation [14][34][35][36] and water treatment systems (municipal wastewater and acid mine drainage) [37][38][39] are recently well-documented. Detoxification studies by research groups led by Qi and by Skłodowski [27][40] have employed LRC as an attractive low-cost adsorbent for the removal of different pollutants from the aquatic and terrestrial environments. The complex and heterogeneous coal matrix is created by amorphous polymers containing double- or triple-substituted aromatic rings which makes LRC highly suitable for immobilizing di- and trivalent metals in soil, consequently reducing their uptake by plants. Brown coal-derived HA has been used already multiple times for the environmentally beneficial adsorption of metal ions (Al^{3+}, Pb^{2+}, Fe^{3+}, Ca^{2+}, Mn^{2+}, Mg^{2+}, Cu^{2+}, Ni^{2+}, Co^{2+}, Cd^{2+}), that strongly reduced their mobility, bioavailability, and phytotoxicity [41][42][43][44][45].

The system of interactions between HA and dissolved metal ions creates a complex supramolecular network given by their heterogeneous, polyelectrolyte, and polydispersive character [46]. In comparison with HA/HS isolated from various soils, HA/HS from brown coal exhibit a remarkably high sorption capacity and a low desorption profile [47]. A. Pusz [48] showed that brown coal can be especially effectively employed on soils strongly contaminated with heavy metals, and suggested using it at the dose of around 90 t ha^{-1} (roughly equivalent to a dose of 150 g pot^{-1} in their studies).

Coal-derived humic substances are considered to be effective for the extraction and concentration of many organic pollutants as well. The recovery degree of phenols using magnetic Fe_3O_4 nanoparticles modified with HA from natural sources (brown coal, peat, chernozem, and sapropel) exceeded 94% [49]. The sorption rate of polar organic pollutants can be strongly influenced by the degree of the HS aromaticity [50]. Brown coal amendment to soil contaminated with the pesticide pentachlorophenol resulted in a distinct improvement of its biodegradation, enhancing the growth of the inoculated bacterial strain *Comamonas testosteroni* [51].

2.4. Effects of LRC on Soil Microbial and Biochemical Qualities

The application of exogenous organic matter is often critical to improving soil fertility and nutrient management. Only such a treatment can substantially stimulate microbial activity, root respiration, enzyme turnover, and many other biological processes in soil. Studies assessing the impact of LRC on soil microbial community structure and activity are scarce. However, existing reports consistently show that the LRC amendment increases soil microbial activity, manifesting in elevated soil respiration, higher enzyme activity, and larger CEC [52][53][54]. The high specific surface area and porosity of LRC promotes ventilation and moisture retention, providing a favorable habitat for the growth and activity of microbial communities [55]. Activity levels of various hydrolytic and ligninolytic enzymes (including esterases, peroxidases, phenol oxidases as well as supporting enzymes, e.g., H_2O_2-generating oxidases; all predominantly of fungal origin) are strongly positively correlated with the enrichment of soil with LRC [56].

Due to its chemical and physical properties LRC act as a “storehouse” for nutrients that attracts soil microbial communities. Microorganisms with different physiological properties and metabolism transform LRC and generate HS through the so-called “ABCDE-system” (A = alkali, oxidative; B = biocatalysts; C = chelators; D = detergents; E = esterases) [57]. Microbially produced chelators and alkaline substances attack the macromolecular coal matrix and dissolve HS [57].

Metagenomic analyses revealed that both endophytic and epiphytic microorganisms are abundant in the LRC environment, as coal is generally originated from plant materials and therefore exhibits inherent plant interaction abilities [58][59][60]. LRC supplementation usually promotes the relative abundance of *Actinobacteria.* Due to their filamentous nature, these bacteria favor and can readily colonize the leonardite-rich environment [52][61]. Many members of *Actinobacteria* and *Firmicutes* are able to solubilize and depolymerize coal matrix [62][63][64]. However, further data regarding microbial functional responses to LRC exposure are inconsistent. Victorian brown coal had a short-term effect on the soil microbial community after 60 days of application, i.e., it temporarily increased the peroxidase and phenol oxidase activities, suppressed the heterotrophic respiration, and induced shifts among microbial populations [65]. Bekele et al. [66] observed that leonardite amendment had no effect on microbial biomass carbon (MBC) of the receiving subsoil, while application together with labile organic mix resulted in intermediate MBC values. It is important to note that the current understanding of microbial colonization and its activity is mainly drawn from short-term studies; thus, more testing should be done yet, especially focusing on long-term studies.

3. Impact of LRC on Plant Growth and Crop Yield

Among the main benefits of using LRC as soil amendments are the enhancement of plant growth and stress resistance. Some coal-derived HS is promoted commercially as plant growth stimulants and regulators. However, despite multiple publications showing the positive effects of coal derivatives on plant growth, the success of commercial coal-derived products in agriculture varies and so there is a relative lack of statistical evidence of its effectiveness. Furthermore, most of the commercial products are highly complex and contain mixtures of organic matters as well as added plant nutrients, which makes it difficult to identify the individual effect of HS [67].

While several studies have confirmed the beneficial role of coal-derived products on plant growth, only a few have specifically examined the direct impact of LRC. Part of the reason for this is the wide range in physicochemical and functional properties that make LRC substantially less predictable regarding plant-stimulating behavior compared to other soil amendments of a known chemical structure [68]. In addition, depending on the used LRC type, the selected plant, and soil, as well as on environmental conditions, the actual efficiency of an amendment can vary dramatically.

The majority of applications were conducted in hydroponic, soil-less, or field conditions. On the one hand, in most cases, significant plant-growth stimulation was observed in response to LRC derivatives/compositions. For example, Amoah-Antwi et al. [69] reported that LRC applications provide long-term soil quality benefits and adequate protection against pollution, which results in reduced net abatement costs. On the other hand, the observed effects were inconsistent across the studies, depending on the type of plant treated, soil classes tested and the manner of product application.

Rose et al. [67] ranked the factors contributing to positive plant-growth promotion using a boosted regression tree (BRT) and demonstrated that application rate, HS source, and plant type were the key factors regulating HS impact on the shoot and root growth, while the growth media employed and the location of application played a negligible role. HS can influence plant growth directly, by acting on physiological and metabolic plant processes, and indirectly, by modification of soil characteristics [70][71].

For example, hormone-like and catalytic activities of HS directly stimulate the shooting and rooting of plants [72]. Moreover, some studies suggest that HS may directly stimulate the activity of H+-ATPase and ion transporters in the root plasma membrane, consequently enhancing nutrient acquisition [73][74]. The best documented indirect effects of HS include improvement in soil structure, pH buffering, CEC, and water retention capacity, as well as enhancement in nutrient bioavailability (particularly P, Fe, K, Zn, and N) and reduction of toxicity of heavy metals [75][76]. The presence of abiotic environmental stress factors, such as salinity, nutrient deficiency, and heavy metal toxicity plays a big role in shaping the root growth response to HS [77]. The high content of (coal-derived) HS, alleviates salinity stress presumably by binding excess cations [67].

A general conclusion that can be drawn from the studies listed here is that the response of crop yield to LRC is mainly affected by its origin, level of coalification, rate/dose, form/mode of application. Crop yield is also dependent on specific plant responses, soil type, and environmental conditions.

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