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Review

Low-Rank Coal as a Source of Humic Substances for Soil Amendment and Fertility Management

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Abstract: 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. LRC, a highly heterogeneous material in nature, contains large quantities of HS and may effectively help to restore the physicochemical, biological, and ecological functionality of soil. Multiple emerging studies support the view that LRC and its derivatives can positively impact the soil microclimate, nutrient status, and organic matter turnover. Moreover, the phytotoxic effects of some pollutants can be reduced by subsequent LRC application. Broad geographical availability, relatively low cost, and good technical applicability of LRC offer the advantage of easy fulfilling soil amendment and conditioner requirements worldwide. This review analyzes and emphasizes the potential of LRC and its numerous forms/combinations for soil amelioration and crop production. A great benefit would be a systematic investment strategy implicating safe utilization and long-term application of LRC for sustainable agricultural production.

Keywords: low-rank coal; brown coal; soil amendment; soil health; crop yield; soil remediation

Citation: Akimbekov, N.S.; Digel, I.; Tastambek, K.T.; Sherelkhan, D.K.; Jussupova, D.B.; Altynbay, N.P. Low-Rank Coal as a Source of Humic Substances for Soil Amendment and Fertility Management. *Agriculture* **2021**, *11*, 1261. https://doi.org/10.3390/agriculture11121261

Academic Editors: Vladimír Frišták and Martin Pipíška

Received: 1 November 2021
Accepted: 9 December 2021
Published: 13 December 2021

1. Introduction

Managing soil health is essential for its functional biodiversity, environmental sustainability, and crop productivity. Nowadays, around 33% of the global land is degraded and virtually unproductive due to various factors. The negative impact of intensive land use on soil properties and productivity is evidenced by a significant shift in the balance of humic substances (HS) and nutrients in arable soils over past decades [1–3]. This situation thereby affects the livelihood and food security of billions of people worldwide [4]. The annual demand for organic soil amendments is constantly increasing across the globe and makes it impossible to fulfill it with traditional types of organic matters. Sole utilization of chemical fertilizers hardens the soil, reducing soil fertility and polluting the natural environment [5].

Alternative sources for soil fertility management can be low-rank coal (LRC) and its derivatives. LRC is rich in a wide range of macro- and microelements and is also a valuable source of organic matter containing a high amount of HS [6]. For energy or industry applications, LRC is not economically feasible and therefore in the course of mining, it is usually sent to dumps. The amount of such low-rank coals is estimated during its detailed exploration and evaluation, while depends on the deposit structure and its location. However, there is no doubt that the overall LRCs amount is huge and has severely disturbed the natural environment [7]. LRC is enjoying rapidly growing application in agriculture as a fertilizer synergist due to its ability to ameliorate a broad range of soil properties. Particularly, LRC-derived HS, amended to the soil at specific rates and combinations can provide additional benefits for soil productivity [8].

In light of the potential value of LRC, we sought to gain a better understanding of using LRC as an amendment/conditioner for soil health and plant development (Figure 1). The objectives of this review were to examine and to summarize various effects of LRC (i) on soil chemical characteristics including soil carbon (C) and nitrogen (N), soil organic matter (SOM), pH, cation exchange capacity (CEC); (ii) on soil physical properties, such as moisture, porosity, and bulk density; (iii) on soil biological properties, including microbial community, mineralization, and enzyme activity; and (iv) on plant growth response and crop productivity. The beneficial impact of LRC in addressing polluted soil is also considered (v).

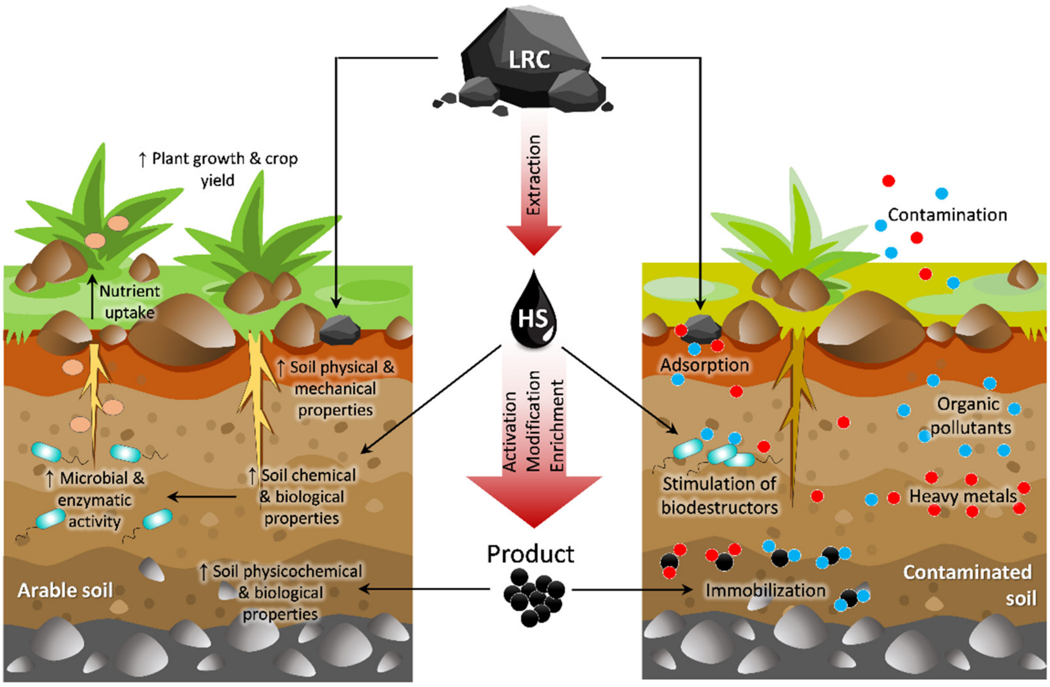

Figure 1. Low-rank coal and its derivatives provide a highly functional additive for soil fertility maintenance and plant growth stimulation. In addition, it is highly effective to immobilize and degrade various pollutants in soil, reducing their availability to plants.

2. LRC Types and Properties

Coal is considered one of the world's most abundant and most important fossil fuels for power generation [9]. There are different types of coal that are characteristically distinct in few specific features, such as origin, composition, and coalification level [10]. 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 [11]. 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 [12].

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 [13]. 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 [14]. 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 [15]. Moreover, LRC may combust spontaneously during mining and utilization, thereby causing air-polluting emissions [16]. Up to date, thousands of hectares of previously-fertile land functioning ecosystems are disturbed by coal mining and coal waste [17].

3. 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 (Table 1). It is important to note that the labile and humified organic matter of LRC has a decisive impact on soil health and fertility [18,19].

Table 1. Selected short- to long-term effects of LRC and its derivatives on soil properties.

Experimental Type	Soil Type	Type of LRC (Dose)/Origin	Study Duration (Years)	Effects and Inference	Reference
Laboratory	Acid yellow sand	Brown coal (1%, 2%)/Indonesia and Australia	0.1	The treatment ameliorated the effect of acidity, Al phytotoxicity and increased root growth of acid-sensitive wheat. However, any decrease in Al activity was solely dependent on increased soil pH, which may provide the basis for evaluating the value of brown coal in ameliorating soil acidity.	[20]
Greenhouse	Grey-brown podzolic (Luvisol)	Brown coal-based preparation (brown coal—85%, peat—10%, brown coal ash—4%) 320 g per pot containing 6.4 kg soil/Poland	2	The preparation created the HA with higher aromaticity and higher resistance to thermal distraction, consequently increasing CEC, which is particularly important in processes of heavy metal bonding. The application of brown coal resulted the C:N ratio increase, showing the necessity of soil fertilization with N.	[21]
Experimental station	Loam	Leonardite (10 and 20 Mg ha^{-1})/Turkey	2	SOM content increased significantly ($p < 0.01$) compared to control (no leonardite); and could be used as soil conditioner material in soils with lower SOM content and to increase the yield of crop. However, no effect on soil EC, pH, and lime was observed.	[22]
Field pots	Grey-brown podzolic	1. Brown coal 140 g per pot (56.4 kg soil). 2. Brown coal-based preparation (brown coal—85%, low peat—10%, brown coal ash—4%, mineral fertilizers–1%) 180 g, 360 g, 720 g per pot (56.4 kg soil)/Poland	7	Soil: higher C contents, and consequent higher C/N ratios were recorded, particularly in the soil with the highest dose of preparation. Soil HA: higher content of carboxyl groups and a more aromatic character were noted, particularly HA from the soil with the highest dose of preparation. These results may be attributed to the increasing content of simple aromatic moieties of HAs.	[23]
Microcosm	Clay loam, sandy soil, clay	Brown coal (10 t ha^{-1}) + Urea (50 kg ha^{-1})/Australia	0.4	The treatment had a minor, temporary effect on N-cycling, microbial activity, and community composition in different soil types with or without urea application. Brown coal reduced the CO_2 emissions, primarily by inhibiting respiration. Periodic increases in Px and PO activities in treated soils were also observed. Thus, under circumstances where brown coal is applied to soil for beneficial effects, it is unlikely to substantially contribute to increased greenhouse gas emissions or significantly disrupt soil microbial processes in the short term.	[24]

Table 1. *Cont.*

Experimental Type	Soil Type	Type of LRC (Dose)/Origin	Study Duration (Years)	Effects and Inference	Reference
Field	Salidic Calciustolls	Brown coal (5 kg m^{2-}) + CSB: *Bacillus mycoides*, *Microbacterium* sp. and *Acinetobacter baumannii* (1×10^8 bacteria mL^{-1} at a dose of 100 mL m^{-2})/Colombia	0.5	Increase in the soil respiration, microbial and enzymatic (LiP, MnP, and Lac) activity was recorded. Decrease in the EC, SAR and ESP was shown. The results suggest the possibility of using brown coal as an OM source for the rehabilitation of degraded saline soils and in the dry lands influenced by open-pit coal mining.	[25]
Laboratory	Sandy	Brown coal slow-release fertilizer (Activated brown coal + polymeric compounds) 100 g per 500 g soil/China	0.3	Activated brown coal revealed the high adsorption ability to NO_3^-, NH_4^+, $H_2PO_4^-$, and K^+. In addition, it improved the soil water-retention property and showed the nutrient (N, P, K) slow-release characteristics. The findings suggest that the newly developed slow-release fertilizer has great potential to be used in plant cultivation and production systems.	[26]
Experimental field	Loamy	Brown coal-urea (BU) granules with C:N ratios of 1–10. Each different BU granule (5 ± 0.1 g) per 60 g soil/Australia	0.1	N-release from BU granules was slower than from urea, resulting in higher N retention. Addition of BU blends and brown coal alone increased water holding and retention capacity of the soil. These findings support the hypothesis that BC is suitable for developing slow-release N fertilizers.	[27]
Glasshouse	Clay loam, sandy loam	Granulated brown coal with urea (BCU) (40–54% C and 5–22% N) 250 mg N kg^{-1} (either from granulated or urea) soil/Australia	0.2	Decrease in the release of fertilizer-N and substantially increase in the mineral and PMN by decreasing its gaseous and leaching losses. The granules containing higher proportions of brown coal maintained better N retention. The results suggest that BCU granules enhanced efficiency fertilizer for increasing availability and use efficiency of N by crops.	[28]
Greenhouse	Medium (silt) and coarse- textured (loamy sand)	Brown coal HA (liquid fertilizer—actosol) 6.2 g and 12.4 g per 5.0 kg soil/Poland	2 and 3	Sorption complex characteristics, SOM quality, and dehydrogenase activity were improved in both soils. The soil quality index increased for the loamy sand: from 0.16 (control) to 0.29 (actosol), while for silt, from 0.19 (control) to 0.28 (actosol). Although the positive effects were visible in both soils, the more robust improvement of soil properties was especially marked in coarse textured soil, rendering low grade lignite-derived humic acids a valuable product, especially with poor soils.	[18]
Field	Haplic Luvisol	HA from leonardite, brown coal, alkalized leonardite, and alkalized brown coal. Total HA: 39.91, 19.39, 89.16 and 303.75 kg ha^{-1}, respectively/China	0.1	All HA increased cumulative NH_3 losses by 147.7, 278.5, 113.9, and 355.3%, respectively, compared with the control (no HA). A significant increase in cumulative CO_2 losses was recorded only under alkalized brown coal HA treatment, by 14.44–24.90% compared with all other treatments. Soil urease and sucrase activity was higher under alkalized brown coal HA treatment. Since humic acid from pulverized leonardite caused no increase in NH_3 volatilization or CO_2 emissions, it is therefore thought to be the most suitable humic acid for field application.	[29]
Laboratory	Silty clay loam	Brown coal HA (0.5, and 1.0 kg ha^{-1})/Pakistan	0.1	The addition of 0.5 and 1.0 kg ha^{-1} HA promoted CO_2 evolution, increased bacterial population by 355% to 476%, fungi 610 to 716%, and CEC of the soil by 13.8 to 28.9%. The results suggest that the brown coal HA addition caused the improved biochemical environment of the soil.	[30]

Table 1. *Cont.*

Experimental Type	Soil Type	Type of LRC (Dose)/Origin	Study Duration (Years)	Effects and Inference	Reference
Greenhouse	Saline-sodic	Brown coal and CSB (*Bacillus mycoides*, *Acinetobacter baumannii* and *Microbacterium* sp.) at 1% (1×10^8 bacteria mL^{-1} g^{-1} brown coal) of soil/Colombia	0.6	An increase in the soil respiration, microbiological activity, CEC, and activity of the enzymes LiP and Lac was observed. A decrease in the EC, SAR, and ESP was noted. The findings suggest the possibility of using brown coal as a possible organic amendment in saline-sodic soil, where the microbial activity can accelerate the biotransformation processes of coal to contribute to the rehabilitation of the disturbed soils.	[31]
Pot experiment	Sandy loam soil	Coal-derived commercial humates (CH1 and CH2 with ~40% C and ~1% N; CH3 with ~20% C and 4.26% N)/200, 400, 700, 1500, and 3000 kg ha^{-1}/Russia	0.1	All treatments resulted in a slight HA accumulation and a minor accumulation of FA. Fungi, actinomycetes, and bacteria in CH-soil mixtures were highly stimulated by the rate of 700 kg ha^{-1}. Soil oxidative processes were also activated, which in turn enhanced soil aerobic properties. The data obtained characterize CH as a valuable microbial fertilizer, although one should bear in mind that at high rates CH can possess microbial toxicity as well.	[32]
Glasshouse	Stony Creek (SC) and Cranbourne (CB)	Brown coal-derived products (humate granule (4 kg ha^{-1}), humate powder (10 L ha^{-1}), blend (1 t ha^{-1}), granule (50 kg ha^{-1}), conditioner (1 t ha^{-1}), raw brown coal (5 t ha^{-1}))/Australia	0.2	Microbial colonization was higher in SC, while only humate granule resulted in higher colonization in CB. The addition of products generally had a positive effect on microbial biomass in CB soil. The pH of CB (7.4–7.6) was higher than that of SC (4.6–4.7). This finding highlights the need for soil specific optimization when applying these amendments.	[33]
Greenhouse	Subsoils: Clay, loam, and sand	Leonardite (humalite)/53.1 (loam), 14.3 (clay), 9.1 (sand) g kg^{-1} + fertilizers and labile organic mix/Canada	0.3	Subsoils had higher organic C than control, regardless of soil type. Treatment increased microbial biomass and decreased geometric mean diameter of the dry soil aggregates. Humalite-only amendment on these soil properties was not significant relative to control. However, long-term field studies are required to ascertain the longevity of the desirable properties and to assess effects associated with aging of humalite in the soil.	[34]
Phytotron chamber	Smelter-polluted soil and post-mining soil	Coal slurry (Cs) from coal preparation plant (2%) and Lake Chalk (LC) from a brown coal mine (2%)/Poland	1.5	The highest values of OCC were recorded for lake chalk amended both soils. The immobilization of heavy metals in smelter-polluted soil with lake chalk was noted. The reduction in the bioavailability of heavy metals (Zn, Cd) in both soils was observed. The results suggest that the additives used in experiment may be a valuable fertilizer source for supporting plant growth and development.	[35]
Greenhouse	Dark-chestnut soil	Leonardite—L (1.5 g kg^{-1}) and Leonardite HA—LHA (1 g kg^{-1})/Kazakhstan	0.3	The pH of L-soil (6.9) was lower than that of LHA-soil (7.1) and control soil (7.4). Metagenomic analysis displayed the high microbial diversity and richness of LHA-soil compared to the control. The significant changes in the bacterial population structure of L-soil were observed. The findings highlight the importance of amending leonardite-based humic products for maintaining the biogeochemical stability of soils and keeping their healthy microbial community structure.	[36]

Table 1. *Cont.*

Experimental Type	Soil Type	Type of LRC (Dose)/Origin	Study Duration (Years)	Effects and Inference	Reference
Greenhouse	Loam and silt loam	Brown coal HA (50, 100, 150 and 200 mg kg^{-1})/Pakistan	~0.8	HA improved soil nutrient status by increasing organic matter (9%), total N (30%), available P (166%) and available K (52%), indicating a substantial increase in soil nutrient status. The improvement in soil fertility in response to humic acid observed in this study is critical in the degraded and eroded soils.	[37]
Laboratory	Silty-clay soil	Leonardite HA (1000, 2000, 4000 and 8000 mg kg^{-1})/Italy		The doses had no effect on soil shrinkage and water-stable microaggregates, but rather they determined deterioration of physical characteristics of the soil. The findings indicate that the influence of humic acids on soil properties is likely to depend on the origin and characteristics of the humus fractions used as amendment.	[38]
Pot trials	Sandy	Leonardite humate (250, 500, 1000, 2000 and 4000 mg kg^{-1})/Italy	0.4	The doses up to 2000 mg kg^{-1} resulted in a progressive stimulation of bacterial growth. In addition, slight effects on soil actinomycetes were evidenced while filamentous fungi did not differ. However, at high concentrations have confirmed some negative effects on soil biota.	[39]
Laboratory	Silty sand	HS from various LRC (HS/soil weight ratio 1:20)/Greece		Brown coal samples provided the best results concerning the HA concentration, as well as the CEC improvement of the amended soil. The results enable an initial correlation among the different parameters and a rating of the samples according to their suitability for soil-amelioration agents.	[40]
Field	Saline-sodic	Brown coal HA (1.5 Mg ha^{-1}) with flue gas desulfurization gypsum (3.2 Mg ha^{-1})/China	5	The SOM, porosity, microporosity, MWD, water-stable macroaggregate, and AWC were increased by 22.8, 6.34, 23.2, 48.1, 55.5, and 15.8%, respectively, while the BD was decreased by 5.9% compared to no amendments applied. The authors suggest a great potential for ameliorating saline-sodic farmland soil by using combined amendment of brown coal HA and flue gas desulfurization gypsum.	[41]
Soil columns	Acid red podzol	Calcium-saturated coal-derived organic products (80 or 160 g Ca m^{-2})/Australia	8	Amendment was effective in decreasing exchangeable A1 and increasing pH and exchangeable Ca to depth, the extent being a function of amendment and rate applied. The formation of inorganic and organic complexes were assumed to be responsible for the movement of Al out of the column in the leachate.	[42]

Note: AWC—available water content; BD—bulk density; CSB—coal solubilizing bacteria; DWB—dry-weight basis; EC—electrical conductivity; ESP—exchangeable sodium percentage; FA—fulvic acid; Lac—laccases; LiP—lignin peroxidase; MWD—mean weight diameter; OCC—organic carbon content; PMN—potentially mineralizable N; PO—phenol oxidase; Px—peroxidase; SAR—sodium adsorption ratio.

The SOM of LRC is characterized by its high content (<90% d.w.) of humic substances (HS) [43,44]. 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) [45]. HS can be extracted from coal using alkali, acids, and organic solvents [46]. 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 [47,48]. 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 [43].

3.1. 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 [49]. 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 [50]. Cihlar et al. [51] 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 [52,53].

Brown coal-derived humic acid can reduce the disaggregating effects of cyclic wetting and drying on soil structural stability [54]. Soil porosity, an essential component of the soil skeleton structure and site productivity, can be maintained after coal mining by conducting site reclamation [55]. 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 [56]. The high CEC of brown coal results in greater retention of NH_4^+ and consequently lowers the NO_3^- leaching loss [57].

Most of lignin oxygen-containing functional groups result in a low pH level when ionized in solution [58]. 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. [59] 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 [20]. 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 [25].

3.2. 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 [52]. The application of LRC by B. Dębska with colleagues [21] 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 [60]. Enhancement of SOC content and sequestration following LRC-derived HS is well-documented by R. Spaccini et al. [61].

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

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 [45,62–64] and water treatment systems (municipal wastewater and acid mine drainage) [65–67] are recently well-documented. Detoxification studies by research groups led by Qi and by Skłodowski [58,68] 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 [69–73].

System of interactions between HA and dissolved metal ions creates a complex supramolecular network given by their heterogeneous, polyelectrolyte, and polydispersive character [74]. 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 [75]. A. Pusz [76] 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 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 a magnetic Fe_3O_4 nanoparticles modified with HA from natural sources (brown coal, peat, chernozem, and sapropel) exceeded 94% [77]. The sorption rate of polar organic pollutants can be strongly influenced by the degree of the HS aromaticity [78]. 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* [79].

A summary of the effects of various LRC types on heavy metals mobility and uptake by plants in different soils is provided in Table 2.

Table 2. LRC application effectively reduces the bioavailability of heavy metals in the soil and their uptake by plants.

Experiment Type/Location	Soil	Heavy Metal (Concentration in mg kg^{-1})	LRC Type/Applied Dose (% Is a Mass Percentage)	Plant	Effects	References
Pot experiment/Poland	Clayey silt (I) and heavy silty loam (II)	Soil I: Cu (4985), Pb (1236), Zn (294.6), Cd (2.82); Soil II: Cu (1008), Pb (413.0), Zn (194.5), Cd (1.51)	Brown coal/50, 75, 100, 150, and 175 g pot^{-1}	Composed plants: ryegrass (40%), red fescue (35%), Italian ryegrass (15%), meadow-grass (10%)	The increasing doses of brown coal caused statistically significant decrease of heavy metals content in plants while change of total heavy metals content in soils was statistically insignificant. Among analyzed doses it is suggested to use dose of 150 g pot^{-1}.	[76]
Pot experiment/Poland	Acidic soils	Cu (<1686) and Zn (<368)	Brown coal/50 g kg^{-1}	Red fescue	Application of brown coal to soil caused increased accumulation of heavy metals in plants, i.e., useful for phytostabilization of Zn in polluted soils	[80]
Greenhouse/New Zealand	Pallic, orthic, rendzic, sandy	Cd (1.1)	Brown coal/1, 3.4, and 7.1%	Ryegrass	1% brown coal amendment reduced plant Cd uptake by 30%, without adversely affecting biomass or the uptake of essential nutrients including Cu and Zn.	[73]
Pot experiment/Poland	Haplic Luvisols	Cd (0.80), Pb (60.4) and Zn (90.0)	1. Brown coal product "Rekulter" (85% brown coal, 10% peat, 4% brown coal ash, 1% mineral); 2. Brown coal/180, 140, 390 and 630 g per pot	Rye	TOC amounted to 12, 15 and 8 g kg^{-1} in contaminated soils amended with brown coal, Rekulter and control, respectively. Contamination caused a high decline in the yield of fresh and dry mass, respectively, 79 and 76% compared with objects without heavy metals. By adding the Rekulter and brown coal, the negative influence of heavy metals on yield was neutralized. The highest yield was in case where Rekulter was applied.	[81]
Pot experiment/Pakistan	Sandy loam	Cd (25)	1. Brown coal/1% OC; 2. Brown coal + rice husk biochar/0.5% OC + 0.5% OC; 3. Brown coal + FYM/0.5% OC + 0.5% OC	Wheat	Amendments were highly effective in enhancing the wheat growth and yield as well as in minimizing the phyto-available fraction of Cd and its transfer to edible tissue of wheat.	[82]
Field/Pakistan	Loamy sand	Cd (7.35)	1. Brown coal/0.1%; 2. Brown coal + limestone/0.05% each; 3. Brown coal + biochar/0.05% each.	Wheat and rice	Amendments increased the grain and straw yields as well as gas exchange attributes compared to the control. No Cd was detected in wheat grains with the application of amendments. The lowest Cd harvest index was observed brown coal + biochar treatment for rice.	[83]

3.4. Effects of LRC on Soil Microbial and Biochemical Qualities

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 LRC amendment increases soil microbial activity, manifesting in elevated soil respiration, higher enzyme activity, and larger CEC [36,84,85]. 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 [31]. 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 [86].

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) [87]. Microbially produced chelators and alkaline substances attack the macromolecular coal matrix and dissolve HS [87].

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 [88–90]. 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 [36,91]. Many members of *Actinobacteria* and *Firmicutes* are able to solubilize and depolymerize coal matrix [92–94]. 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 [24]. Bekele et al. [34] 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.

4. 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 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 [95].

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 (Table 3). 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 [96]. 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.

Table 3. Response of various plants to soil supplementation with LRC derivatives/compositions.

Location	Media/Soil Type	Plant	LRC Type (Applied Dose% *w*/*w*)	Plant Health Effects and Inference	Reference
Shandong, China	Sandy loam soil	Apple trees	Brown coal slow-release fertilizer (SAF) (Activated brown coal + polymeric compounds)	Treatment improved the leaf chlorophyll content values, stem length, and trunk girth. These results clearly demonstrated that activated lignite enhanced SAF through improving its slow release and water-retention capabilities and thus stands as a strong candidate as an alternative nutrient vector for increasing fertilizer use efficiency and promoting the growth of apple.	[26]
Hokkaido, Japan	Hydroponics	Barley	Brown coal HA (10 and 25 mg-C L^{-1})	Brown coal HA was more effective in promoting the barley root growth (~33 cm) than those of compost derived from cattle manure (~23 cm). The antioxidant enzymatic activities (catalase and ascorbate peroxidase) increased. Using HA as a supplement can be effective in enhancing antioxidation enzymatic activities, while the appearance of the effects is retarded because of the decomposition and release of auxin-like compounds from HA by organic acids from the plant roots.	[97]

Table 3. *Cont.*

Location	Media/Soil Type	Plant	LRC Type (Applied Dose% *w/w*)	Plant Health Effects and Inference	Reference
Pretoria, Republic of South Africa	In vitro/medium	Cantaloupe, lettuce and onion	Coal-derived sodium humate (500, 1000, 5000, and 20,000 mg L^{-1})	Stimulation in the root growth of seedlings of cantaloupe at 1000 mg L^{-1}, lettuce, and onion at 500 and 1000 mg L^{-1}, as well as hypocotyl growth of cantaloupe at 1000 mg L^{-1} were observed. Growth at optimal humate concentrations was significantly increased above that of nutrient solution controls, indicating that the stimulatory effect cannot be ascribed to a supply of nutrient elements by the coal product.	[98]
Pisa, Italy	Pot/sandy soil	Chicory	Leonardite humate (250, 500, 1000, 2000, and 4000 mg kg^{-1})	Stimulatory effect was directly correlated with the amount of amendment. Humate at 2.000 mg kg^{-1} promoted plant growth for 39 cm and fresh weight for 18 g compared to the control (33 cm and 12 g), respectively. However, at concentrations higher than 2.000 mg kg^{-1}, appeared quite toxic to the plants, indicating that the choice of the optimal concentration is crucial.	[39]
Victoria, Australia	In vitro/compost extract	Cress	Compost extract (Brown coal HA + FYM) (3 mL per a Petri dish)	Brown coal addition improved the germination index of the final compost: 90–113% compared to 71% for FYM only. Future large-scale field studies assessing the agronomic value of lignite-amended manure compost are recommended.	[99]
Navarra, Spain	Growth chamber/nutrient solution	Cucumber	Purified leonardite HA (PHA) (2, 5, 100, and 250 mg of organic carbon L^{-1})	The higher doses of PHA caused a transient increase in the expression of the CsHa2 (plasma membrane H^+-ATPase) for 24 and 48 h. The higher doses up-regulated CsFRO1 (Fe (III) chelate-reductase) and CsIRT1 (Fe (II) high-affinity transporter) expression for 48 and 72 h; whereas these genes were down-regulated by PHA for 96 h. These results stress the important relationships existing between HS effects on plant growth and plant Fe uptake mechanisms.	[100]
Navarra, Spain	Growth chamber/nutrient solution	Cucumber	Purified leonardite HA (5 mg L^{-1} and 100 mg L^{-1} of organic C)	The root application causes a significant increase in shoot growth that is associated with an enhancement in root H^+-ATPase activity, an increase in nitrate shoot concentration, and a decrease in roots. The findings indicate that the beneficial effects of HA on shoot development could be directly associated with nitrate-related effects on the shoot concentration of several active cytokinins and polyamines.	[101]
Merelbeke, Belgium	Field/loamy sand, sand, sandy loam	Herbage	Leonardite HS (1. 8.3 kg ha^{-1} (liquid form); 2. 3.6 to 6.4 kg ha^{-1} (incorporated))	A significant proportional increase of 0.14 ($p < 0.05$) with the incorporated treatment and a non-significant increase of 0.08 with the liquid treatment were observed compared to the control. In general N, P and K uptake at the first grass cut was higher after application of HS but only in one experiment was this increase statistically significant.	[102]
Moscow, Russia	Model experiment/sandy loam soil	Lettuce	Coal-derived commercial humates (CH1 and CH2 with ~40% C and ~1% N; CH3 with ~20% C and 4.26% N)/200, 400, 700, 1500, and 3000 kg ha^{-1}	The CH samples with similar properties (CH1 and CH2) exhibited different growth-stimulating effects; CH2 was less effective. The least effective was CH3 despite the highest N content. High application rates of CH inhibited plant development despite the higher nutritional value. This leads to the conclusion that either CH bound N is unavailable for plants, or the amount and quality of HA are more important for growth-stimulating effects of CH than the total amount of nutrients.	[32]
Clayton, Australia	Pot experiment/Stony Creek (SC) and Cranbourne (CB)	Lucerne and ryegrass	Brown coal-derived products (BDP) (humate granule (4 kg ha $^{-1}$), humate powder (10 L ha^{-1}), blend (1 t ha^{-1}), granule (50 kg ha^{-1}), conditioner (1 t ha^{-1}), raw brown coal (5 t ha^{-1})	Lucerne: the effect of BDP on the shoot weight varied considerably, only blend product caused a positive root growth effect, but this occurred only in CB soil. Ryegrass: No strong positive shoot growth responses to any BDP in either soil. Blend product gave a significantly positive root growth response in the CB, whereas the reverse was true in the SC. There were significant differences between the effects of each soil on plant nutrient uptake. Given the variable responses of the plant species and soil types to the amendments applied the further mechanistic studies are needed to help understand how these amendments can be used to greatest effect.	[33]

Table 3. *Cont.*

Location	Media/Soil Type	Plant	LRC Type (Applied Dose% *w/w*)	Plant Health Effects and Inference	Reference
South Sulawesi, Indonesia	Greenhouse/oxisols	Maize	Brown coal HS: (300 ppm (H1), 600 ppm (H2), 900 ppm (H3))	The higher the dose of the HS given, the higher the plant height, the sum of leaves and dry weight value. However, the plants still showed P deficiency symptoms. The study discovered the effect of HS from lignite to the availability of soil P in the oxisols that problemed with soil chemistry.	[103]
Brno, Czech Republic	Hydroponics	Maize	Brown coal potassium humates (40 mg L^{-1} and 2 mM $CaCl_2$)	The biological activity (root growth, mass increment, root division) of the humates was related to the nature of self-assemblies, while the chemical composition had no direct connection with the root growth. However, full control of chemical and physicochemical properties and biological activity still remains a challenge.	[104]
Mikulčice, Czech Republic	In vitro/medium	Maize	Brown coal potassium and ammonium humates (100 mg kg^{-1})	Humic samples with the lowest molecular size (0–35 kDa) showed no correlation with bioactivity (Pearson coefficient (PC) from 0.05 to −0.4), middle-sized (35–175 kDa) showed a highly significant positive correlation (PC up to 0.92) and the highest molecular-sized (275–350 kDa) showed a negative correlation (PC up to −0.75). The appropriate and most efficient combination of HS/pre-treatment agent to simulate more effectively the molecular re-aggregation in parental lignite should be considered.	[105]
Uvalde, USA	Growth chamber/sandy greenhouse/sandy and clay	Pepper (bell)	Brown coal HS (0.5 kg m^{-2}; both chamber and greenhouse)	HS increased plant tolerance to water stress conditions due to the reduction of leaf moisture loss and stimulation of root development. More specifically, it increased root development and soil bacteria population in moderate and no stress conditions. Physiologically, HS decreased leaf stomatal conductance and transpiration after imposing severe or mild stress. Due to their capacity to improve plant root growth and microbial activity, application of HS might have long-term benefits in agricultural systems.	[84]
Bangkok, Thailand	In vitro/solution	Riceberry	Leonardite HA (1000 mg L^{-1})	The increase in the root (25 cm) and shoot (38.6 cm) lengths compared with untreated (20 and 33.5 respectively) were observed. Weights of root and shoot in treated were higher than untreated at 44 days plantlet. However, the impact of long-term HA application on riceberry growth should be considered.	[106]
Shandong, China	Greenhouse/solution	Rice	Brown coal HA activated with molybdate-phosphorus hierarchical hollow nanosphere (Mo-P-HH) catalyst (10 mg L^{-1})	The rice germination rate reached 90% after 5 days of incubation. Seedlings displayed longer root and shoot compared to the other groups. The contents of Mo and P elements were higher than that in other treatments. The study provided a high-performance hierarchical hollow nanocatalyst for activation of HA and also offered the theoretical basis for the application of HA in agriculture.	[107]
Tamil Nadu, India	Field/Vertisol and Alfisol	Rice	1. Brown coal HA soil application/10 or 20 kg ha^{-1} 2. Foliar spray/0.1% 3. Root dipping/0.3%	Soil application at 10 kg ha^{-1} + foliar spray (0.1%) + root dipping (0.3%) provided the high nutrient (NPK) availability in both soils compared to the other treatments. The increased availability of micronutrients due to the addition of HA might be attributed to the ability of HS to form chelating compounds.	[108]
Nanjing, China	Hydroponic nutrient solution	Snap bean	Leonardite HA with different molecular weights (400 mg/L)	Plants treated with low-molecular-weight HA had significantly greater root length (2–65%), root surface area (6–83%) than those treated with other HA, while leaf growth was affected mainly by HA with high molecular weight. Uptake of K by shoot was higher in plants treated with low-molecular-weight HA. It is concluded that low molecular weight fraction of HA appeared to promote the production of snap bean due to an enhancement in the physical growth of leaf and root.	[14]

Table 3. *Cont.*

Location	Media/Soil Type	Plant	LRC Type (Applied Dose% *w/w*)	Plant Health Effects and Inference	Reference
Elsen Tasarkhai, Mongolia	Field/calcic kastanozems	Tree species (*Populus sibirica*, *Salix ledebouriana*, and *Acer tataricum*)	Leonardite HA (2000, 10,000, and 20,000 mg L^{-1})	Compared to monthly RHGR over four years, the treatment yielded significantly better tree growth. Significant differences were observed between the humic fertilizer concentrations, which varied depending on the species. Further studies will be needed for long-term monitoring, including those in which species of trees and soil types necessary for specific objectives in different ecological conditions.	[109]
Gembloux, Belgium	In vitro/culture medium	Tree species (silver birch and black alder	Leonardite HA (10, 50 or 100 ppm)	HA affected root growth, mainly lateral roots formation, and primary root length. At 10 ppm, HA stimulated especially primary root growth. At 100 ppm did not affect alder root growth but increased root growth in birch. The high molecular weight fraction was more effective at promoting root development than the lower one. The stimulation of root development was mainly due to HA fraction.	[110]
Overton, US	Field/fine sandy loam, fine loamy, siliceous	Turnip and mustard	Leonardite (56.1, 112.1, 224.3 and 445.6 kg ha^{-1})	No significant interactions occurred among treatments and the number of applications in fresh weight of leaves or roots, soluble solids, percent dry weight, or size distribution. Detectable amounts of humic acid were not found in the soil after the experiment was concluded, probably due to the small amounts of leonardite applied.	[111]
Puławy, Poland	Greenhouse/neutral soil	Wheat	1. Brown coal-based fertilizer (50% HA) with ammonia (25.06 g per 7 kg soil); 2. Brown coal-based fertilizer (50% HA) with magnesite (29.06 g per 7 kg soil)	There was no significant influence of the fertilizer type on spike number per plant and plant height measured at the booting stage of wheat development. Wheat responded positively to soil application of the brown coal-based fertilizer. Additional studies should be conducted to select a special binder and appropriate raw material ratios to increase the particle hardness.	[47]
Clayton, Australia	Glasshouse/loamy sand	Wheat	Urea-enriched brown coal granules (1:3 and 1:10)/nominal delivery of 230 mg N kg^{-1} soil	The granules significantly reduced the amount of nitrate and ammonium lost through leaching and reduced the emission of nitrous oxides from the soil, whilst not reducing the plant available N. The study provides a proof of concept for the pilot-scale production and use of brown coal blended organo-mineral fertilizer granules.	[112]
Clayton, Australia	Glasshouse/acid soil (Dermosol)	Wheat	1. Brown coal (1% and 2.5%, equivalent C basis); 2. P (5, 10, and 25 kg ha^{-1})	When no P was applied, addition of brown coal increased shoot height. The addition of both resulted in additive effects, with increased shoot height, tiller number, shoot dry matter and tissue P uptake. Further study is required to assess whether this growth response translates to improvements in grain yield at feasible agronomic and economic rates of addition.	[113]
Azad Kashmir, Pakistan	Greenhouse/calcareous and a non-calcareous haplustalf	Wheat	Brown coal HA (30, 60 and 90 mg kg^{-1})	The largest increases in plant height and shoot fresh and dry weights were found with 60 mg kg^{-1} treatment, being 10%, 25%, and 18%, respectively, as compared to the control. The wheat growth and N uptake in the non-calcareous soil were higher than those of the calcareous soil. These results have the potential to be applicable in wheat growing regions of both soils.	[114]

Note: CP—crude protein; DM—dry matter; FYM—farmyard manure; HA—humic acid; P—phosphorus; RHGR—relative height growth rate; SOC—soil organic carbon.

Table 3 overviews the studies on plant growth-promoting activity of LRC and its products. 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. [8] 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. [95] 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 [115,116].

For example, hormone-like and catalytic activities of HS directly stimulate shooting and rooting of plants [117]. Moreover, some studies suggest that HS may directly stimulate activity of H+-ATPase and ion transporters in the root plasma membrane, consequently enhancing nutrient acquisition [101,118]. 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 [119,120]. 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 [121]. High content of (coal-derived) HS, alleviates salinity stress presumably by binding excess cations [95].

The studies devoted to the impact of LRC supplementation specifically on the crop yield are addressed in the Table 4. 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.

Table 4. The crop yield response to application of LRC derivatives/compositions.

Location	Media/Soil Type	Crop	LRC Type/Applied Dose	Yield Change	Quality Effects and Inference	Reference
Lublin, Poland	Field/Loamy sand	Arnica	Leonardite/2, 4 and 6 kg ha^{-1}	Fresh and air-dry matter of flower heads (g m^{-2}) 419 (351 control) and 75 (63 control), respectively.	LRC positively affected the activity of enzymes (dehydrogenases, acid phosphatase, urease, and protease) catalyzing the transformation processes of SOM. It was recorded a significant increment of the number of flowering stems and inflorescences per plant resulting in raw material yields increase along with increasing leonardite dose. As a consequence, raw material yield's increment was obtained.	[85]
Murcia, Spain	Pot/Calcareous soil	Barley	Leonardite HA/5, 100 and 200 mg C kg^{-1}	from 38 to 62%	It significantly enhanced plant growth compared with the control in every dose applied. It had a less favorable effect on N and P absorption as the doses increased. This may suggest that the leonardite contains HS partly formed of high stable compounds.	[122]
Victoria, Australia	Glasshouse/Tenosol (pH 7.24) and dermosol (pH 5.4) soils	Beet (silver)	Brown coal-urea blend (100 kg N ha^{-1} and 50 kg N ha^{-1}) + P (40 kg ha^{-1}) and K (60 kg ha^{-1})	27% and 23% in neutral and acid soil, respectively	Increase in the N uptake by silver beet and SOC. The blends with higher brown coal (17% N) had higher biomass yield, better N uptake and maintained higher mineral N in soil compared to the blends with lower brown coal (22% N). Blending of urea with brown coal can strongly reduce N losses via gaseous emissions, as a result greater amount of N was available to beet, increasing the N uptake and use efficiency.	[123]
Coimbatore, India	Pot/Alfisol soil	Blackgram	1. Brown coal HA soil application/10, 20, 30, and 40 kg ha^{-1}; 2. Brown coal HA foliar spray (0.1%) and seed soaking (1%) application	from 7.23 to 9.46 g pot $^{-1}$	Among the various dose of HA, 20 kg ha^{-1} recorded a significantly higher seed yield. Among the methods of application, soil amendment of HA performed better than seed soaking and foliar spray. Confirmatory results should be obtained in the field experiment.	[124]
Manitoba, Canada	Greenhouse/Purple spring sandy loam soil	Canola, wheat, and green beans	Leonardite (0.5, 1, 5 and 10 g to 3 kg of soil) + Nutrients (per kg soil: 100 mg N, 50 mg P, 20 mg S, 100 mg K, 4 mg Zn, 4 mg Fe, 2 mg Mn, 1 mg Cu, 1 mg B and 0.4 mg Mo).	1 and 10 g of leonardite caused 15 and 27% canola increase, respectively	Uptake of S, N, P and K by canola were significantly affected by the leonardite amendment. However, the application of leonardite had no significant effect on the yield of wheat and green beans. It can be concluded that leonardite increased the yield of canola by supplying S directly and by possibly facilitating the uptake of other nutrients. The lack of response of wheat and green beans to leonardite was attributed to their lack of response to S.	[125]
Tokat, Turkey	Experimental station/Loam soil	Climbing bean	Leonardite (10 Mg ha^{-1}) + N (130 kg ha^{-1}) and P_2O_5 (100 kg ha^{-1}) fertilizers	6.099 kg m^{-2}	The effects on the pod number and pod length were not significant. Leonardite could be used as soil conditioner in soils with lower organic matter content and to increase the yield of climbing bean.	[22]
Victoria, Australia	Field/Loamy soil	Canola and wheat	Brown coal-urea (BU) granules (5 ± 0.1 g) with different C:N ratios (1–10) per 60 g soil	Canola: ~1750 kg ha^{-1} than in the control (500 kg ha^{-1})	No significant differences in the grain yield of wheat between any treatments, but slightly increased with the increase in N application rate. However, the grain protein content of wheat was increased. BU granules containing 8–17% N with a C:N ratio of 5.4 to 2.7 were the most suitable. The findings support the hypothesis that brown coal is suitable for developing slow-release N-fertilizers.	[27]
Krakow, Poland	Pot/Silt and loamy sand	Celery and leek	Brown coal HA (liquid fertilizer: actosol)/6.2 g and 12.4 g per 5.0 kg soil	Loamy sand: the yields were <4 fold higher than control. Silt: the difference was ~2.5-fold.	The application of 12.4 g HA significantly promoted the growth of shoots and roots of the plants in the loamy sand, while in the silt, the crops in both 6.2 g and 12.4 g treatments were almost equal. The use of brown coal HA for fertilizing soils for vegetable cultivation can be an economically reasonable and environmentally justified way to enhance both agricultural productivity and soil quality, especially with coarse textured soils.	[18]

Table 4. *Cont.*

Location	Media/Soil Type	Crop	LRC Type/Applied Dose	Yield Change	Quality Effects and Inference	Reference
Jinju, Republic of Korea	Hydroponics	Lettuce	Commercial leonardite HA (Mycsa (USA)/1 g L^{-1} of the plant nutrient solution	Fresh and dry weights were increased over control (without HA)	Coincubation of isolated microbes (*Bacillus* and *Aspergillus* genera) from HA with lettuce resulted in a significant increase in plant biomass and enhanced resistance to NaCl-related abiotic stresses. The microbiological factors could be considered when coal-related HS is applied in hydroponic crop cultivations.	[126]
Portici, Italy	In vitro/solution	Lettuce (L) and tomato (T)	Leonardite-derived HS/40, 100, 1000, and 5000 mg L^{-1}	L: fresh weight was enhanced at high content of HS. T: dry weight was increased at some of HS.	The fresh weight of total seedlings and per seedling increased with increasing concentrations for both plants without showing signs of growth inhibition up to 5000 mg L^{-1}. The results suggest that cell elongation was the only effect on lettuce seeds whereas an uptake of HS must have also occurred in the case of tomato seeds.	[127]
Dezhou, China	Column cultivation/Fluvo-aquic light loam	Maize	Leonardite HA-enhanced urea (HAU)/0.10 g of HA in 19.90 g molten ^{15}N urea per column (50 kg dry soil)	Grain yields were 5.58–18.67% higher than the control (urea treatment)	The uptake of fertilizer N under the HAU treatments was higher than that under the urea treatment by 11.49–29.46%. The aboveground dry biomass of plants grown with HAU was enhanced by 11.50–21.33% when compared to that of plants grown with urea. This is likely due to the abundance of the COO/C–N=O group in this HA component.	[128]
Islamabad, Pakistan	Pot experiment/alkaline calcareous soil	Maize	Brown coal HA/25 (HA_1) and 50 (HA_2) mg kg^{-1} soil in conjoint with N/150 (N_1) and 300 (N_2) mg kg^{-1} soil	Fresh biomass increased by 23% and 44% with HA_1 and HA_2 respectively, ~23% increase in dry biomass at both HA.	Cob weight and grain weight increased significantly (29% and 40%) with HA at 25 and 50 mg kg^{-1} respectively with regard to control (no HA), with N at 150 and 300 mg kg^{-1} the increase was 51% and 103%. The HA application increased plant N contents by 20% and 26%, P by 14% and 20% and K by 15% and 10% in HA_1 and in HA_2, respectively. The application of HA improved soil characteristic by playing its role in chelating nutrients that became available to plant.	[129]
Sanliurfa, Turkey	Field/Clay loam soil	Maize	Leonardite (750 kg ha^{-1}) + S (625 kg)	Grain yield was improved significantly under P deficiency and water stress.	The amendment mitigated the negative effects of stress factors (P deficiency and water deficit) and increased plant growth. Leaf total chlorophyll content, maximum fluorescence yield, leaf water potential, and leaf relative water content were improved. The addition of S-enriched leonardite increased the antioxidative defense system and photosynthetic machinery of maize under water stress and P deficiency, therefore, it can be recommended for field application under water limited calcareous soils.	[130]
Peshawar, Pakistan	Pot experiment/Silty clay loam	Maize	Brown coal HA/sprayed on the soil at 0, 50, 100, 150, 200, 250, and 300 mg kg^{-1} soil along with N-P-K (120–90−60 kg ha^{-1})	50 and 100 mg kg^{-1}, increased shoot and root yield by 14 to 23 and 7 to 39%, respectively	HA increased soil N concentration and plant N accumulation ($p < 0.05$) over control with no significant differences within the treatments. Soil P concentration improved ($p < 0.05$) by the addition of 200 mg kg^{-1} HA whereas plant P accumulation was not significantly affected by the application of different HA doses. The beneficial effect of HA on plant growth and nutrient uptake are mainly associated with the potential of HA to improve biochemical environments of the soil by improvement in soil microbial activity and soil CEC.	[30]
Faisalabad, Pakistan	Pot experiment/Sandy clay loam	Okra	Brown coal HA (10, 15 and 20 mg kg^{-1}) and NPK (60–50−30 mg kg^{-1})	Green pod yield (48 g $plant^{-1}$) at HA 20 mg kg^{-1} with NPK	The highest shoot fresh weight (112 g $plant^{-1}$) was recorded in HA at 20 mg kg^{-1} in combination with NPK. However, there was no effect of HA application on root fresh weight. Maximum N (1.28%), P (1.37%) and K (1.43%) in fruit was recorded when HA was applied at 20 mg kg^{-1} soil with NPK. The HA application alone had no significant effect on fruit N, P or K contents. The authors conclude that HA can be a supplement but not a substitute of fertilizers.	[131]
Córdoba, Spain	Greenhouse/River sand and peat (2:1) and field/Orchard	Olive	Leonardite HS (9% HA and 7% FA)/foliar application at 0.5, 1, 2, 4, 8 and 16%.	1% treatment 29.08 kg $tree^{-1}$ vs. control 24.61 kg $tree^{-1}$	Greenhouse: Shoot growth significantly increased at 0.5% or l%. Field: shoot growth stimulated and the accumulation of K, B, Mg, Ca, and Fe in leaves promoted. HS did not influence the nutritional status of the olive trees and, therefore, do not compensate for the lack of mineral nutrition.	[132]
Virudhunagar, India	Field/Sandy clay loam	Onion	Brown coal HA/soil application (10 and 20 kg ha^{-1}) foliar spray (0.1%)	Increased (11.31%) the bulb yield of control	At 20 kg ha^{-1} significantly increased the plant height (49.5 cm), the number of leaves per plant (47.2) and root length (11.2 cm). This might be due to the overall improvement of plant growth and allied increase in root biomass resulting in higher water and nutrient absorption.	[133]
Almaty, Kazakhstan	Greenhouse/Dark-chestnut soil	Potato	Leonardite—L (1.5 g kg^{-1}) and Leonardite HA—LHA (1 g kg^{-1})	Tuber yield 57.3 (L), 66.4% (LHA) and 49.3% (control)	Increase in the plant height, as well as the number of stems/plant (20.8% and 24% increases in plant height and the number of stems/plant relative to control, respectively) in LHA-treatment. The highest total number of potato tubers was obtained in LHA-group (88.5% more than in the control). Leonardite improves the physical properties of soil by increasing its sorption ability due to organic humified substances, subsequently improving the mineral nutrition of plants and their provision with microelements.	[36]
Isparta, Turkey	Research farm/Loam soil	Potato	Leonardite (200, 400, 600 kg ha^{-1})	Marketable tuber yield (38%) and total tuber yield (15%) increased over control.	There were no significant differences between the leonardite doses for plant height and specific gravity. Leonardite applications increased the number of tubers per plant (22%), improved protein and vitamin C contents, and specific gravity of tubers. Differences between 400 and 600 kg ha^{-1} leonardite doses were insignificant. However, the specific gravity was higher in tubers harvested from leonardite applied plots than the tubers harvested from control.	[134]

Table 4. *Cont.*

Location	Media/Soil Type	Crop	LRC Type/Applied Dose	Yield Change	Quality Effects and Inference	Reference
Tamil Nadu, India	Field/Non-acid kaolinitic soil	Rice	Brown coal HA/20 kg ha^{-1}	Grain yield 4253 kg ha^{-1} (control yield 3786 kg ha^{-1})	Straw yield increased for 6380 kg ha^{-1} over control (5679 kg ha^{-1}). N, P, and K uptake were increased from (control) 20.3, 5.82 and 28.97 kg ha^{-1} to 132.0, 20.75 and 86.98 kg ha^{-1}, respectively. Rice, being a monocot, could have taken up more amount of K by virtue of its high root CEC, which might be the reason for the marked increase in K uptake.	[135]
Varanasi, India	Pot experiment/Sandy loam soil	Rice	Brown coal K-humate (70% HA, 49.5% C and K 10%)/5.0 and 10.0 mg kg^{-1} + Zinc sulphate/12.5 mg kg^{-1}	48.35 g pot^{-1} at 10.0 mg kg^{-1} vs. control 29.32 g pot^{-1}	Application of 10 mg kg^{-1} brown coal K-humate along with zinc sulphate recorded highest N, P, K, S, and Zn uptake by straw and grain of rice. Increased nutrient content in soil due to HA application would have contributed to more K absorption by rice.	[136]
Gumushane, Turkey	Field/clay-loamy soil	Ryegrass	Leonardite/250, 500 and 750 kg ha^{-1}	Hay yield increased to 24% compared with the control	Amendment gave a higher crude protein yield than the control. The content of K, S, Ca, Mg, Fe, Mn, and B of ryegrass hay increased as compared with the control, whereas they had no significant effect on Cu and Zn content. Leonardite may have potential for use in organic agriculture as with these treatments hay production of annual ryegrass is improved in terms of yield, protein and mineral content.	[137]
Skierniewice, Poland	Greenhouse/nutrient solution	Tomato	Brown coal/fraction of brown coal with ⌀2.5, 10, and 20 mm	The total yield was obtained under ⌀2.5 mm.	The slightly acidic pH of brown coal positively influenced the availability of most nutrients (N, P, K, Mg, and Ca). Findings indicate that lignite is a good medium and could be used in greenhouse soilless cultivation.	[138]
Cottbus, Germany	Greenhouse/Quaternary sand	Wheat	N-modified brown coal granules (82% HS and 5% N)/5, 7.5, 11, 15, 28 t ha^{-1}	Grain and straw yields were 2-fold higher relative to control	N and water use efficiency even at low application rates and a better growth performance compared to control were observed. An application rate of 5 t ha^{-1} is looked upon as an adequate application rate for very poor substrates and soils. Long-term field studies for different soil and climatic conditions are needed to verify this concept.	[139]
Azad Kashmir, Pakistan	Greenhouse/Loam and silt loam	Wheat	Brown coal HA/soil application: 50, 100, 150 and 200 mg kg^{-1}; soil + foliar application: 100 mg kg^{-1} + 100 mg L^{-1}	1000-grain weight increased by <17%, grain yield by <58%	HA increased plant growth in terms of shoot length (18%), root length (29%), shoot dry weight (76%), root dry weight (100%) and chlorophyll content (96%). The relative increase in NPK uptake in plants was 57, 96, and 62%, respectively over the control. Long-term studies are recommended under field conditions to examine the HA benefits for increasing crop productivity.	[37]
Adana, Turkey	Field/Clay soil	Wheat	Brown coal HA/100 kg da^{-1}	564.3 kg da^{-1} and 442.9 kg da^{-1} (control)	The levels of SOM and available P significantly increased. The combined applications of HS with traditional chemical fertilizer seemed generally and quantitatively to provide better effects on soil characteristics and crop productivity over single chemical and humic applications.	[140]
Punjab, Pakistan	Field/Sandy clay loam	Wheat	Coal HA/10, 20, 30, 60, 90, 120, 150 kg ha^{-1}	3.24 t ha^{-1} at 90 kg ha^{-1} (9.86% more than control)	Coal HA improved the physical properties of soil (TOC, aggregate stability, saturated hydraulic conductivity, bulk density, and soil water contents). 120 kg ha^{-1} dose rate was an economical level. The authors concluded that coal HA improved soil health by stimulating microbial diversity and activity, thus increasing the wheat yield.	[141]

Note: HA—humic acid; N—nitrogen, P—phosphorus; S—sulfur; SOC—soil organic carbon; SOM—Soil organic matter; TOC—total organic carbon.

5. Application Forms of LRC for Soil Amendment and Fertility Management

5.1. Sole LRC and HS Application

As shown and discussed above, LRC has the ability to immobilize pollutants meanwhile making important nutrients and microelements more easily accessible for plants. Many commercial products are derived from coal matrices and mainly sold as humates and humalite [142]. LRC-based fertilizers, such as Rekulter, Actosol and others are successfully used in agricultural practice [18,21]. LRC or LRC-derived products can be formulated as soluble slow-release granules, powders, or even liquids that are applied either directly to the soil or as a foliar spray [33,143,144]. Products might vary in concentration of, for example, HS, specific HS extraction methods, and the composition of incorporated nutrients.

The mechanisms that lie behind the fertilizing activity of sole LRC and its derivatives can be (i) ion-exchange groups capable of complexing or adsorbing and (ii) high porosity that optimizes storage of water and nutrients, thus contributing to their adequate availability [145]. Moreover, considerable research efforts have been aimed to modify LRC using solid-phase activation techniques to improve its pore structure and nutrient uptake [26].

Chemical properties and structural characteristics of LRC facilitate biofunctionality with a wide variety of soil constituents. The extensive surface area, porous structure, and functional groups of coal are used to design slow- or controlled-release soil amendments. A popular solution to further increment the fertilizing value of LRC derivatives is supplementation with mineral fertilizers NPK (recommended doses are variable depending on soil type) [18,30,146]. Some other interesting types of LRC-modifications have been proposed as well and will be briefly reviewed in the following sections. Here it is important to note that careful characterization and assessment of parent LRC with respect to the potential synergistic amendments are key aspects of positive performance outcomes.

5.2. Amendments Used along with LRC

5.2.1. Coal-Urea Fertilizers

Nitrogen (N) is one of the most important nutrients for agricultural crop production. Annually large volumes of synthetic N fertilizers are applied to improve soil fertility and productivity. However, more than 50% of N fertilizers added to soil are commonly lost through volatilization and leaching, resulting in groundwater pollution, plant diseases, N_2O emissions, etc. [147,148]. Furthermore, long-term excessive fertilizer applications may deteriorate soil physical properties, reduce TOC and basic cation content, in consequence increasing soil acidification [149]. The depletion of SOC due to intensive agriculturalization can be a further cause for declining fertilizer N efficiency since SOC plays a pivotal role in the retention of soil N and limits its losses [150].

On the contrary, combining LRC rich in HS with N fertilizer would allow normalization of nutrient uptake by plants and would help to decrease N losses in the soil from excess N fertilizer application [21]. In recent years, various kinds of urea-N loaded brown coal have been developed and investigated for their potential to increase soil quality [24,27]. It was shown that LRC indeed reduces volatilization loss of N from urea-amended soil by inhibiting urease activity, thus increasing urea-N availability for plants [151,152]. Some studies already demonstrated that LRC can be also successfully employed in the synthesis of controlled-release fertilizers based on urea, formaldehyde, and KH_2PO_4 [107,153]. More recently, LRC-urea granulates came into focus since they offer good predictable performance, the vast availability of the substrate, and a standardized production process [112,139].

5.2.2. Combination with Coal Solubilizing Bacteria

The effect of added LRC on the chemical and biological properties of soil is expectedly greater if applied in conjunction with active strains of coal solubilizing bacteria. A number of species of *Escherichia*, *Pseudomonas*, *Streptomyces*, *Bacillus*, *Staphylococcus*, and *Rhodococcus* are capable to release humic organic matter through biotransformation of the coal [154–157]. Co-application of LRC and exogenous *Bacillus mycoides*, *Microbacterium* sp. and *Acinetobacter baumannii* increased the soil respiration and microbiological activity [25,31].

Experiments from Jeong et al. [126] by using a hydroponic lettuce culture showed that microorganisms were directly and actively participating in plant growth stimulation by HS.

5.2.3. LRC and Biochar

Another sustainable alternative to the application of conventional organics fertilizers is represented by LRC combined with biochar produced on a laboratory and industrial scale. Biochar is produced through the pyrolysis of biomass and like LRC possesses high stability against decay and a superior ability to retain nutrients in soil [158]. Biochar and LRC have many common physicochemical and biological characteristics, such as extensive surface area, porous structure, chemical functional groups, high water-holding, and CEC [159,160]. The synergetic employing these materials may provide a long-term positive effect on SOM, thereby building up a sustainable form of C [161]. The utilization of biochar and LRC alternatively or in the mixture to improve soil health and plant growth is well documented by many authors [8,34,48].

5.3. LRC in Composting Technologies

Composting is a cost-effective and sustainable process of conversion of various organic wastes into compost [162]. However, composting can be related to a high loss of N through NH_3 volatilization [163]. In turn, NH_3 poses negative effects on ecosystems and indigenous biodiversity [164]. Existing strategies to mitigate nitrous emissions and retain N during composting include using bulking agents, bedding materials, microbial and/or chemical amendments, and optimization of composting duration/conditions [165]. However, these approaches are not widely recognized due to the difficulties and limitations in commercial on-farm implementation [166]. Due to its excellent absorption properties, brown coal may suppress NH_3 volatilization in composted material [99]. This was confirmed in recent studies where NH_3 emissions were significantly reduced by brown coal additives (Table 5). Good N retention by brown coal can be also attributed to better N mineralization and inhibition of compost urease activity, resulting in a lower rate of NH_3 production and emission [167]. We argue that the LRC-based technologies can be especially effectively used in the processing of livestock and poultry manure to reduce environmental impacts and improve efficiency.

Table 5. Investigated effects of brown coal applications on NH_3 emission or N retention from various composting systems.

Experiment Type/Location	Composting Substrate	LRC Type (Mode of Application)/Applied Dose	Composting Period	Reported Effects	Reference
Commercial composting reactors (160 L)/Australia	Poultry litter	Brown coal (incorporation)/5, 10, and 15%	65 days	15% amendment significantly increased the temperature in the thermophilic stage of composting, leading to faster degradation of organic matter and accelerated detoxification than control. Brown coal in compost increased NH_4^+ content by 66% and decreased TN loss by 18%. The higher concentration of NH_4^+ corresponds with higher TN content (3.2% in 15% vs. 2.2% in control) and higher concentration of total acid groups in 15% (2.46 mmol g^{-1}) than those in control (2.17 mmol g^{-1}).	[167]
Commercial cattle feedlot/ Australia	Cattle manure	Brown coal (surface application)/ 4.5 kg m^{-2}	90 days	Adding brown coal reduced N losses by 54% during composting, but increased CH_4 and N_2O emissions (due to anaerobic conditions), as well as CO_2 emissions (due to additions of labile C). Total GHG emissions (CO_2-e) (N_2O and CH_4 and NH_3 as indirect N_2O) from the brown coal amended manure was 2.6 times greater than that of the non-coal treatment.	[168]
Commercial beef cattle feedlot/ Australia	Cattle manure	Brown coal (surface application)/20% *w*/*w* (45 t dry brown coal ha^{-1})	21 days	Brown coal treatments retained N by suppressing NH_3 loss by 35–54%, resulting in amended composts having 10–19% more total N than the unamended compost. Relative to manure only, brown coal reduced GHG emissions over the composting: N_2O (58–72%), CO_2 (12–23%) and CH_4 (52–59%).	[99]
Organic fertilizer factory/Greece	Solid swine manure enriched with rice husk and cotton residues	Brown coal (mixing)/1:1	85 days	Brown coal, due to its excellent odor- and moisture absorbing capacities allowed for the successful incorporation of the wet and malodorous swine manure into the compost process. The maximum temperature range was achieved between 45–55 °C for about 20 days and overall compost process of 85 days with ambient air values below 10 °C.	[169]
Open beef feedlot system/Australia	Cattle manure	Brown coal (surface application)/4.5 kg m^{-2}	40 days	Brown coal decreased NH_3 loss by approximately 66%. The cumulative NH_3 losses were 6.26 and 2.13 kg N $head^{-1}$ (steer) in the control and brown coal treatment, respectively.	[166]
Cattle feedlot pens/Australia	Cattle manure	Brown coal (surface application)/3 and 6 kg m^{-2}	Phase 1: 28 days; Phase 2: 38 days	Compared to the control, brown coal application decreased NH_3 emissions by approximately 30%. Brown coal application increased direct N_2O emissions by 40 and 57%, to 0.14 and 0.22 g N_2O-N $head^{-1}$ day^{-1}, for Phase 1 and Phase 2, respectively.	[152]
Respirometer/UK	Wheat straw	Brown coal HA (mixing)/1:1	31 days	Brown coal treatment significantly reduced both the rate of O_2 consumption and CO_2 evolution from the substrate, thus having a practical application as a means of increasing the microbial stability of composts.	[170]
Commercial tumbling composter/ Australia	Poultry litter	Brown coal (mixing)/5, 10 and 15% *w*/*w*	65 days	Brown coal addition effectively promoted the removal of manure borne antibiotic resistance genes (ARGs), mainly from Actinobacteria and Firmicutes. The relative abundances of ARGs decreased by 8.9% in control (no brown coal) and by 15.8, 27.7 and 41.5% in 5, 10 and 15% brown coal treatments, respectively.	[171]

Note: CH_4—methane; CO_2—carbon dioxide; GHG—greenhouse gas; N_2O—nitrous oxide; TN—total nitrogen.

6. Knowledge Gaps, Needs, and Concerns

LRC is an abundant and valuable resource but there are critical issues that must be addressed to facilitate its agronomic value:

- Consideration of the issues related to long-term soil behavior under LRC loading is important for interpreting lasting soil quality changes since most of the described applications of LRC have been derived from short- to medium-term studies [8,95].

- Only limited work has been performed so far to investigate the inherent chemical heterogeneity and functional diversity LRC as soil amendment and there are still uncertainties regarding the chemical mechanisms of LRC as flow-release fertilizer.
- Since the most common techniques for producing HS from coal based on alkaline extraction, it may be unable to achieve its purpose of separating humic from non-humic substances (i.e., from functional biomolecules, their partial decomposition products, and from microbial residues) [172]. There was an apparent lack of relationship between biological functioning of OM and its alkaline extractability [173].
- A weakly acidic nature may make LRC unsuitable for amending many contaminated soils [48]. Consequently, mitigation of soil acidification via liming should be considered. Researches have however demonstrated that LRC when used in heavy metal-polluted soils increment buffering capacity of soil [174,175].
- Most coal-derived HS are combined with Ca and Mg, which have poor solubility in water and weak decomposability in soil, thereby demanding activation processes to convert it into more suitable forms, e.g., brown coal is usually pretreated with strong oxidants [176,177].
- Depending on origin and quality, LRC may itself contain elevated levels of organically-bound chlorides and inorganic constituents, implying a conceivable risk of soil contamination by polycyclic aromatic hydrocarbons, and heavy metals [178,179].
- Causes of suboptimal outcomes applying LRC products can be attributed to the manufacturer's recommended rate with limited knowledge of optimal rates, timing, and methods of application for a given plant-soil combination [33].
- Systematic experimental evidence concerning the amendment dose/rate depending on the soil type, environmental conditions is still missing, resulting in a lack of theoretical models and full understanding regarding plant growth-response to LRC amendment.

The selection criteria for LRC amendment should integrate environmental and agronomic factors, such as soil quality, material availability, economic accessibility, cost, application needs, safety compliance, and sustainability [8].

7. Conclusions

Coal seams and spoils generated during mining and processing operations, are generally considered economically non-viable and deteriorating public and environmental health. However, according to numerous existing studies and interdisciplinary evaluations, low-rank coals could be successfully used for the production of soil amendments/conditioners and reclamation of disturbed lands. Different types and combinations of LRC applied to the soil at specific rates can provide various short- to medium-term benefits, i.e., ameliorate soil structure, improve nutrient mobility, stimulate microbial and enzymatic activity; enhance soil productivity and crop yield. Potentially in a long-term perspective, LRC can serve as a stable source of SOM. However, thorough consideration and careful matching of the involved components and specific factors (soil, crop, LRC, location, etc.) is of paramount importance.

Author Contributions: Conceptualization, N.S.A. and I.D.; software, K.T.T.; validation, N.S.A., I.D. and D.B.J.; formal analysis, D.K.S.; investigation, N.S.A.; resources, K.T.T.; data curation, D.K.S. and N.P.A.; writing—original draft preparation, N.S.A.; writing—review and editing, I.D.; visualization, D.B.J.; supervision, I.D.; project administration, K.T.T.; funding acquisition, N.S.A. All authors have read and agreed to the published version of the manuscript.

Funding: This research has been funded by the Science Committee of the Ministry of Education and Science of the Republic of Kazakhstan (Grant No. AP08855394).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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