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Views & Comments

A Roadmap for Sustainable Agricultural Soil Remediation Under China's Carbon Neutrality Vision

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1. Introduction

Agricultural soil pollution is a major threat affecting soil health and the ability of soil to yield safe and sufficient food; thus, it is a barrier to the goal of zero hunger worldwide [1]. The food deficiency problem continues to grow, particularly under the current atmosphere of global tension. United Nations (UN) organizations, including the UN Environment Program (UNEP) and the Food Agricultural Organization (FAO), previously reached a consensus to "Be the solution to soil pollution," which has contributed to raising global awareness of the threats posed by soil pollution. According to a national soil survey, millions of hectares of agricultural lands in China are affected by heavy metal(loid) pollution (e.g., cadmium (Cd), nickel (Ni), copper (Cu), and arsenic (As)), resulting in a loss of rice productivity exceeding 10 million tons per year [2]. China has already achieved a miracle in sustaining one quarter of the world's population with only 9% of the world's land in the past decades; however, soil pollution is becoming a new challenge in meeting the increasing demand for food in the 21st century. China's central government released an ambitious soil pollution prevention and control action plan in 2016 aimed at reversing the exacerbation of soil degradation. In contrast to the finely developed framework used in restoring industrial brownfield sites, agricultural soil remediation on such a large scale is unprecedented. During the period of China's 13th Five-Year Plan, pilot-scale demonstrations have been extensively implemented across the country, particularly in southern China, based on the national soil survey. Although a great deal of experience has been gained after a massive injection of funds, many fundamental engineering problems remain unresolved. Since China's new "double carbon" goal (i.e., achieving a carbon peak and carbon neutrality) was proposed to cope with climate change, there has been a strong demand for integrating carbon neutrality with other targets in order to achieve harmony between state strategies [3]. In particular, a massive injection of energy and resources into the large area of agricultural soil remediation can significantly contribute to carbon emissions, which is unsustainable and unlikely. Agricultural land is considered to be an important component of carbon pools in the terrestrial system; thus, under proper regulation, it can contribute to carbon sequestration and fixation, thereby reversing the carbon flux.

In this article, we briefly touch upon the fundamental principles of heavy metal(loid) regulation that underlie agricultural soil remediation, from the perspective of the biogeochemical processes of key elements cycling in the soil–plant system (Fig. 1). We then provide an analysis of potential state-of-the-art technologies to meet the multiple objectives of food production safety, soil quality improvement, and carbon reduction/sequestration (Fig. 1). Based on the theory of the whole life cycle, we ultimately introduce a comprehensively sustainable assessment and carbon footprint analysis that can be used as a reliable reference for policymakers and practitioners (Fig. 2).

2. Unlocking the biogeochemical driving forces of pollutants in the soil–plant system

The traditional understanding of heavy metal(loid)s in soils is mainly based on their physicochemical interactions with minerals, against which chemical stabilization approaches have been directly adopted from the remediation of brownfield sites. For example, lime and its derivative agents—which are very low-cost amendments—have been extensively used to remedy Cd-contaminated agricultural lands with the aim of mitigating the acidity of China's red soil region in the demonstration engineering of the national action. Although it can effectively alleviate the Cd bioavailability in soil for a short period of time, this chemical-based approach cannot be considered a genuine success due to its consequences, which include acidity rebound, negative impacts on soil quality, and the potential activation of coexisting As. This lack of long-term success can be ascribed to the large gaps that exist in the knowledge transition from the fundamental biogeochemical processes of these heavy metal(loid)s to their control principles in soil as exemplified in Fig. 1, for which the driving forces of these heavy metal(loid)s in soil are largely overlooked.

From a biogeochemical perspective, in addition to its role of carrying/immobilizing pollutants in the form of iron (Fe) minerals in

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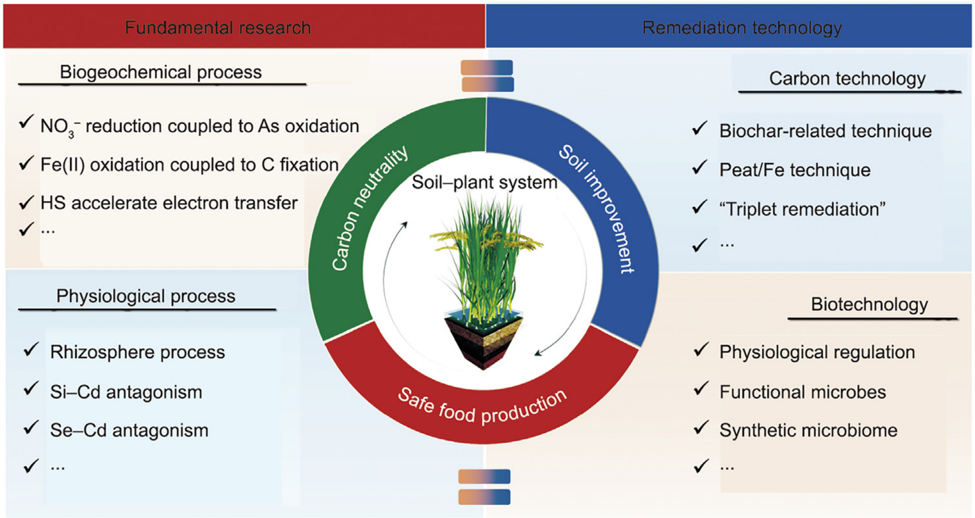

Fig. 1. Representative fundamental research and green technology for agricultural soil remediation. NO_3^-: nitrate; Hs: humic substances; C: carbon; Si: silicon; Se: selenium; Fe: iron; Fe(II): ferrous.

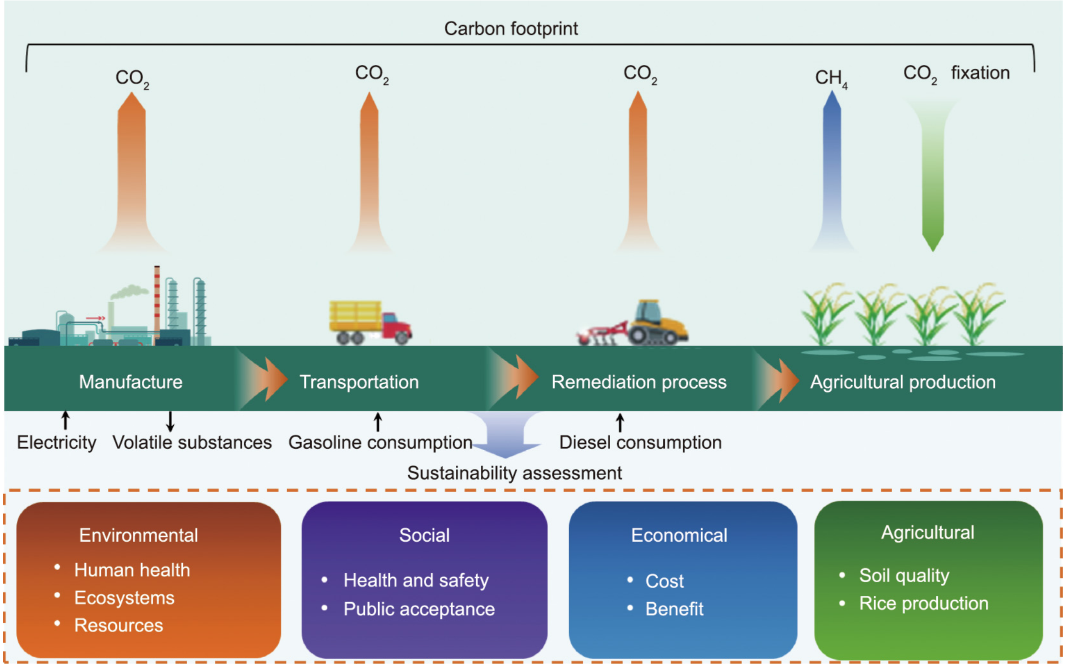

Fig. 2. A conceptual framework for sustainability assessment and carbon footprint analysis.

the soil, Fe is considered to be an iconic element of red soil, in which the microbially driven redox of Fe cycling plays a pivotal role in many environmental processes involving nutrients and toxic metal(loid)s [4,5]. As a representative example, our group has demonstrated that the abiotic oxidation of biogenic ferrous (Fe(II)) by oxygen (O_2) in paddy soil during the drainage stage is a major contributor in proton generation and therefore in increasing the acidity of the soil, which is difficult to overcome through chemical neutralization with lime [6]. An alternative strategy is to take advantage of the coupling process between microbially anaerobic Fe(II) oxidation and nitrate reduction, with which excess protons from Fe(II) oxidation can be effectively scavenged by nitrate reduction [4,7]. Importantly, microbial nitrate reduction can also trigger significant oxidation of arsenite (As(III)) as a result of activating As-oxidizing bacteria (e.g., *Pseudogulbenkiania*), limiting the mobility of As under anaerobic conditions [8]. Humic substances (HSs) appear to participate in these biogeochemical processes as well by accelerating electron transfer, thereby significantly determining the thermodynamics and kinetics of these coupling processes. These are just a few examples to illustrate the benefits of taking advantage of biogeochemical processes to address problems that cannot be solved with pure chemistry alone. In fact, microbial-induced Fe cycling has also been found to determine carbon fixation or mineralization in soil, in processes such as dissimilatory Fe reduction coupled with organic carbon mineralization [9] and Fe(II) oxidation coupled with carbon assimilation (i.e., carbon dioxide (CO_2) fixation) [10]. These processes hold great potential for simultaneously achieving carbon sequestration and heavy metal(loid) regulation in future engineering practices.

Another critical knowledge gap exists regarding the uptake of heavy metal(loid)s from the rhizosphere zone to the edible parts of crops (e.g., rice grains), which involves multiple microbial, chemical, and physiological processes. Such a feature involving complex interfaces and matrices is in contrast to the remediation of brownfield sites. It should be remembered that the ultimate goal of agricultural soil remediation is to achieve safe food production, so it is essential to prevent the absorption and transportation of pollutants from the roots to the edible parts of a plant. In the process of pollutants passing from the soil into the root, Fe plaques have been revealed to be particularly important as a means of obstructing heavy metal(loid)s entering root cells. The formation of Fe plaques and their interception of heavy metal(loid)s, such as through nitrate oxidation-induced Fe plaque formation, have garnered significant research attention [11].

The uptake and translocation of heavy metal(loid) pollutants from the root to the edible parts of a plant also involve complex physiological processes, in which element antagonistic effects can play a crucial part. Cumulative evidence shows that silicon (Si) and selenium (Se) can effectively downregulate the expression of *OsLCT1*, the gene responsible for Cd transport, and significantly upregulate *OsHMA3*, the gene responsible for detaining Cd in the vacuoles of the root [12]. Researchers have recently proposed that complexes of Fe with organic carbon can target-regulate the gene expression in rice tissues in order to preferentially accumulate Cd into the leaves, further advancing the progress of developing new strategies for heavy metal(loid) regulation in crops [13]. This advance is a landmark of success in formulating the control principles of agricultural soil remediation; nevertheless, there is still a high demand for multidisciplinary knowledge exchange in this area.

3. Looking out for "green technology for multi-objective achievement

As stated in Section 1, the goal of agricultural soil remediation is to achieve the multiple objectives of soil quality improvement, safe food production and, recently, greenhouse gas (GHG) mitigation. Therefore, it is necessary to ensure that the whole remediation process will not have secondary negative impacts on soil health. Increasing evidence shows that carbon-based technology holds great potential for simultaneously enhancing soil quality and immobilizing heavy metal(loid)s in soils. In fact, the deficiency of organic carbon in the red soil region of China (e.g., about 4.4 $g\cdot kg^{-1}$ in Guangdong Province) is a major cause of soil degradation that can also increase the risk of heavy metal(loid)s, according to the national standard of soil environmental quality for agricultural land (GB15618-2018) [14]. Biomass-derived carbon materials have emerged as popular materials for agricultural soil remediation, and may well be the most studied by far. Although debate on their cost efficiency is still ongoing, biochar-based materials such as Fe-incorporated biochar, which was developed based on the biogeochemical theory of the Fe/C coupling process, have already exhibited their effectiveness in national engineering demonstration projects in southern China [15]. Nitrate-modified peat—a humic-like organic carbon—has been developed in parallel, based on the nitrate/Fe coupling theory, and has been commercialized and successfully applied for simultaneous As and Cd passivation in field trials. We have estimated that approximately 9 million tons of carbon can be sequestrated and stored in the soil if all agricultural biomass can be carbonized and returned to the agricultural land in Guangdong Province, which would be a huge contribution toward reducing carbon emissions. The application of these persistent organic carbons in agricultural soil can also beneficial for carbon fixation and GHG (e.g., methane (CH_4)) mitigation.

The rapid development of nanotechnology for the physiological regulation of heavy metal(loid)s in crops has also begun to flourish. Si nanoparticles, or quantum dots (DQs), have been found to be useful for regulating the Cd uptake in rice plants, which is particularly useful for realizing safe production in slightly Cd-contaminated agricultural lands [16]. The attractive superiority of Si-based nutrient regulators can be attributed to the eco-friendly nature of silicon, which is an important biological element of crops. However, the disadvantage of these foliar application technologies is their limited efficiency, due to which they are generally required to be combined with soil amendments such as biochar. To address this issue, our group has proposed a new strategy called "triplet remediation" to tackle the complicated processes of heavy metal (loid)s (mainly Cd and As) in the soil–plant system, which has demonstrated significant success in paddy field remediation. However, there is still a long way to go in conquering the challenges of agricultural soil pollution in China. In particular, the debate on the longevity and stability of these technologies continues, as these limitations have not yet been resolved once and for all. More importantly, green technology has already become a new proposition under China's vision of the "double carbon" goal. To further advance biochar technology, technological developments such as mobile equipment can realize *in situ* biomass carbonization and carbon sequestration, thus minimizing energy consumption from transportation. More effort is still required to enhance the cost efficiency of such technologies, since the added value of agricultural production is considerably lower than that of other industries.

As a strategic emerging industry, microbial technology is becoming extremely appealing in diverse fields [17], recently including environmental remediation, even though its overwhelming chemical approach is as yet unrealistic for agricultural soil remediation. In fact, autotrophic microorganisms such as cyanobacteria and photoautotrophic bacteria that fix CO_2 based on the Calvin–Benson–Bassham cycle play a central role in the soil carbon balance [18]. Soil nitrogen fixation by means of rhizobium and *Geobacter* has also drawn a great deal of attention for soil quality improvement. These bacteria may have potential to establish electron-transferring networks with redox metals such as chromium (Cr(V)) and antimony (Sb(V)); for example, a novel *Desulfurivibrio* spp. that can mediate sulfur oxidation coupled to Sb(V) reduction was recently identified. Moreover, functional bacteria that can directly immobilize or detoxify heavy metal(loids) could be considered as another approach for heavy metal(loid)s remediation. For example, the isolation and application of an arsenite respiration strain, such as *Thermus* or *Rhodospirillaceae*, are considered to be a likely approach. When using these single strains for soil remediation, the challenge lies in battling for ecological niches against indigenous species; here, synthetic biotechnology may be an option and a powerful tool. Other potential biotechnologies offering different strategies to acclimate complex soil ecosystems include periphyton-based regulation in paddy fields, arbuscular mycorrhizal symbiosis in dryland, and phytoremediation allowing carbon sequestration and heavy metal reduction in soil.

4. Decision-making based on sustainability assessment and carbon footprint analysis

Among the UN 17 Sustainable Development Goals (SDGs), interlinkage among different goals—particularly between SDG 2, "End hunger, achieve food security and improved nutrition, and promote sustainable agriculture," and SDG 11, "Tackling climate change"—exists naturally and should be seriously considered in order to achieve synergies and trade-offs among agriculture, energy, and climate change. Hence, there is an unambiguous demand in integrated decision-making processes for agricultural soil remediation at the national level. To provide a framework based on a set of indicators for government officials and other stakeholders, the FAO developed a protocol for the assessment of sustainable soil management (SSM) in 2020, which constitutes a fundamental tool to assess any intervention implemented in the field as well as practical terms providing key indicators to assess soil functions [19]. This protocol is representative of a new shift to maximize the environmental, societal, agricultural, and economic benefits throughout the duration of the entire remediation process. Based on this principle, it can be seen that the sustainability of biochar for paddy soil remediation is significantly better than that of lime for achieving the double objectives of soil quality improvement and safe rice production [15]. However, an appraisal merely based on either cost or efficiency tends to oppose this conclusion, largely due to ignorance of the significant negative impacts of production/exploitation and transportation. In addition, as an important supplement, a carbon footprint analysis of remediation activity must be incorporated with a sustainability assessment (Fig. 2), in order to synergize carbon reduction (SDG 11) with food production (SDG 2). Therefore, a holistic assessment of agricultural soil remediation is of critical importance and can provide an accurate reference for policymakers and practitioners who will have a significant influence on regulatory affairs and the market.

In summary, the agricultural soil remediation of China, as an important component of China's third national action plan (i.e., the Action Plan on the Prevention and Control of Soil Pollution), is a unprecedented piece of engineering in human history and one that presents numerous challenges. The successful experience gained from exploratory research and practices during the 13th Five-Year Plan is paving the way to sustainably curb the deterioration of agricultural land. It is imperative to avoid repeating the same mistakes and loss of national funds. The direct adoption of remediation technologies from other contexts cannot be accepted without transformation, and innovations in remediation technology should take the characteristic features of the relevant agricultural land into account. Fundamental research must be performed to determine the control principles of pollutants in agricultural land, based on the nature of the multiple mediums, interfaces, and processes in the soil–plant system. It is also necessary to maintain an awareness of the multi-objective synergy of safe food production, soil quality improvement, and carbon neutrality. To assist in policymaking, an appropriate and holistic assessment of potential technologies is necessary prior to the extensive practice of such technologies.

Aside from the abovementioned issues, many other issues remain to be solved, including the need to customize suitable strategies (e.g., classifying and grading-based remediation) for pollutant control from site to site, due to the heterogeneity of agricultural soil on a large scale, and the need for new technical standards for guiding practical implementation. The foundations of agricultural soil remediation have already been laid, while the extent to which it will grow remains to be explored. Collaboration among scientists from multiple disciplines, such as environmental chemistry, microbiology, and geochemistry, is vital in this endeavor. Our aim with this article is to attempt to lay out a meaningful reference for success in the green and sustainable remediation of contaminated agricultural land, in order to support China's unprecedent national action plan on soil.

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