

## Long-Term Effects of Biochar on Mitigating Methane Emissions from Paddy Soil

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### 1. Abstract

Biochar has been reported to mitigate short-term methane emissions from paddy soil. At present, methane mitigation by biochar has primarily focused on the abundance and variations of methanogens and methanotrophs, and changes in their activity during methane production and consumption. However, long-term effects of biochar on methane mitigation from paddy soil remain controversial. In this review, we highlighted two existing opinions on the long-term methane mitigation effect upon biochar application. Combining the already explored mechanism of fresh biochar on methane mitigation from paddy soil and a novel discovery, that chemical reactivity of biochar can also stimulate anaerobic oxidation of methane, we analyzed the possible influences of biochar on methane production and consumption during its aging.

Global warming is a major problem for humankind. Methane (CH<sub>4</sub>) is an important greenhouse gas, contributing up to 20% to global warming [1, 2, 3, 4]. Wetlands, including paddy fields, are an important source of CH<sub>4</sub> emissions [5, 6, 7]. Flooded paddy soil contributed to 25% of the CH<sub>4</sub> emissions from agricultural soil in 2017 [8]. Thus, reducing CH<sub>4</sub> emissions from paddy fields is a major concern for alleviating global warming [9].

Biochar has been reported as a promising material for mitigating CH<sub>4</sub> emissions from paddy soil [10, 11, 12]. Biochar is a black-colored product produced as a result of biomass pyrolysis under limited oxygen [13]. It has been explored as an avenue for carbon sequestration, crop yield increase, and mitigation of CH<sub>4</sub> emis-

sions [14-19]. Rice husk and maize straw have been reported to significantly reduce cumulative CH<sub>4</sub> emissions as per meta-analysis [11]. Ji (2020) [20] reported significantly reduced CH<sub>4</sub> emissions using biochar in an incubation experiment conducted for 77 days. Annual biochar application at a low rate (2.8 t ha<sup>-1</sup>) has also been reported to reduce CH<sub>4</sub> emissions by 41% from paddy, on an average, over a span of five years [21]. Ecological benefits of biochar application have been widely recognized internationally. The '2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventory' newly added biochar to the calculation method of the annual change in the organic carbon storage of mineral soil [22]. This suggests an international recognition of biochar for its ecological benefits.

The mechanism by which biochar facilitates the mitigation of CH<sub>4</sub> emissions from paddy soil was examined in terms of the effect of biochar attributes on soil physicochemical and microbial structure. Generally, most of the explored mechanisms focus on the influence of biochar on the activity changes in methanogens and methanotrophs during CH<sub>4</sub> mitigation [23, 10, 12, 24, 19]. It is generally believed that biochar application in soil mainly promotes CH<sub>4</sub> oxidation activity [23, 21, 24]. Biochar is porous and alkaline [25, 26] the former attribute provides a satisfactory habitat for methanotrophs to increase CH<sub>4</sub>-capturing ability and decrease Al<sup>3+</sup> toxicity [20, 27]. The alkaline attribute of biochar usually increases soil pH to the optimum range for methanogens and methanotrophs [28]. Methanotrophs are more sensitive to soil pH, and therefore, CH<sub>4</sub> oxidation activity substantially increases compared

to methanogenic activity [29]. Furthermore, the use of large particle size biochar in soil increased soil aeration, and enhanced CH<sub>4</sub> oxidation activity. These findings revealed that biochar promoted aerobic CH<sub>4</sub> oxidation activity.

Recently, positive effects of biochar on anaerobic CH<sub>4</sub> oxidation activity induced by the electronic accepting capacities of biochar have also been observed. Biochar can act as an electron acceptor as well as a donator, facilitating CH<sub>4</sub> production and oxidation [30-32]. O-containing functional groups are the main entities responsible for the Electron Accepting Capacities (EAC) and Electron Donating Capacities (EDC) of biochar. Carbonyl and quinone determine the EAC properties of biochar. Zhang et al. (2019b) [32] reported that biochar stimulates the anaerobic oxidation CH<sub>4</sub> due to the presence of C=O. Phenolic groups, quinone/hydroquinone moieties, and condensed aromatic (sub-) structures of biochar that allow electrons to be transferred across the conjugated p-electron systems are the main sources of EDC. Biochar with a high EDC value is beneficial for promoting methanogenic activity [33, 34]. Hence, EDC and EAC are also important factors in assessing the effect of biochar on CH<sub>4</sub> mitigation in paddy soil.

However, most studies that have demonstrated satisfactory results on CH<sub>4</sub> mitigation from paddies, to date, have mainly focused on the CH<sub>4</sub> mitigating effect in pot experiments and short-term experiments. In contrast, observations from field studies and long-term effects, especially after years of biochar aging, are lacking; in other words, the efficacy of biochar in mitigating CH<sub>4</sub> emissions after years of aging remains to be explored.

Opinions on the effect of aged biochar in reducing CH<sub>4</sub> emissions from rice paddies differ. One opinion is that biochar, after four or six years of aging, still effectively mitigates CH<sub>4</sub> emissions, as suggested by pot/incubation experiments [28, 12]. Some other studies demonstrated that fresh biochar significantly decreased cumulative CH<sub>4</sub> emissions from paddy fields but showed no significant difference from the control group after one year of aging [21, 35]. The main reason for this is that fresh biochar increased CH<sub>4</sub> oxidation potential in the first year, while this effect decreased with its aging [21]. Spokas (2013) [36] also reported that biochar aging decreased the ability to promote methanotroph activity. To maximize the long-term CH<sub>4</sub> mitigation effect of biochar from paddies, the mechanism underlying the mitigation effect of aged biochar needs to be further explored.

Theoretically, the exploration of the mechanism should first focus on changes in biochar attributes. Agricultural activities, such as plowing and tillage, usually occur during the fallow period [17, 37]. After years of rice growth, farming, and fallow cycles, biochar particles become smaller resulting in an increase in their exposed

surface area [17]. This facilitates the formation of smaller-sized soil aggregates compared to those produced when fresh biochar is applied [38]. These smaller soil aggregates hamper effective soil aeration. Hence, when large soil aggregates are fractured into smaller ones, the soil electric potential (Eh) would decrease, which may promote methanogenic activity. In addition, small soil aggregates present a much weaker barrier between microorganisms and organic matter [39], indirectly increasing substrate availability for methanogens. During rice tillering and jointing growth stage, the ash content of biochar dissolves and leaches, leading to the gradual disappearance of the liming effect [40, 41]. Smaller biochar particles accelerate the biochar oxidation rate by exposing more biochar surfaces to oxidation by root secretions and oxides. Aged biochar is characterized by a higher number of O-containing functional groups such as carboxyl C, carbonyl C, and phenolic OH [13]. It has been reported that after four months of soil incubation, biochar quinone content decreased [42]. As quinone mainly contributes to EAC, and phenolic OH is the main EDC source, changes in O-containing functional group changes inevitably affect methanogenic and CH<sub>4</sub> oxidation processes [30, 32]. Therefore, to reveal the mechanism of the effect of aged biochar on CH<sub>4</sub> emissions, the key step is to explore the metabolic differences between methanogens and methanotrophs based on changes in the physicochemical and electrochemical properties of the biochar.

Hence, further research should focus on specifying biochar-specific properties that greatly impact methanogenic and CH<sub>4</sub> oxidation processes. Studies have reported the effect of biochar particle size on CH<sub>4</sub> emission [43, 44], and the changes in biochar particle size during biochar aging are evident. However, few studies have focused on changes in CH<sub>4</sub> emissions from paddy soil based on biochar EAC/EDC and soil aggregate distribution; these may be proved to be other important factors that affect CH<sub>4</sub> emissions in paddy soil. In addition, although pot and short-term field experiments are usually feasible operationally and economically results may not adequately reflect the scenario is under actual field conditions. Hence, long-term field positioning experiments, based on soil-plant-atmosphere ecosystems, must be conducted to obtain the most reliable results and CH<sub>4</sub> mitigation mechanisms in paddy fields upon biochar application.

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