Responses to the comments from Reviewer

Reviewer #1

Comment 1: The references cited in the MS are relatively dated. It is recommended that the authors incorporate more recently published relevant literature to strengthen the timeliness of the review.

Response: Thank you for this insightful comment. We have added recently published literature. We believe that these revisions improve both the timeliness and theoretical grounding of the manuscript.

Comment 2: It is recommended that the detailed calculations pertaining to the electrochemical performance and soil internal forces be provided in the Supporting Information.

Response: Thank you for this valuable comment. We have included detailed calculations related to the electrochemical properties of the soil surface and soil internal forces to enhance the transparency and reproducibility of the methodology.

1. Calculation of soil surface electrochemical properties

The calculation formula for parameters related to the electrochemical properties of the soil surface is as follows:

a. Surface potential φ_0:

$$\varphi_0 = \frac{2RT}{2(\beta_{Ca} - \beta_{Na})F} ln \frac{a^0_{Ca} N_{Na}}{a^0_{Na} N_{Ca}} \quad (1)$$

where φ_0 (V) is the surface potential; R (J K^{-1} mol^{-1}) is the ideal gas constant; T (K) is the absolute temperature; F (C mol^{-1}) is the Faraday constant; β_{Ca} and β_{Na} are the effective charge coefficients of Ca^{2+} and Na^{+}, respectively, with $\beta_{Ca} = -0.0213\ln(I^{0.5})$

+ 1.2331 and β_{Na} = 0.0213ln($I^{0.5}$) + 0.766; I (mol L^{-1}) is the ionic strength; a^0_{Na} (mol L^{-1}) and a^0_{Ca} (mol L^{-1}) are the activities of Na^+ and Ca^{2+} in solution; and N_{Na} (mol g^{-1}) and N_{Ca} (mol g^{-1}) are the amounts of Na^+ and Ca^{2+} adsorbed by the soil.

b. Surface charge density σ_0:

$$\sigma_0 = \mathrm{sgn}(\varphi_0)\sqrt{\frac{\varepsilon RT}{2\pi}\left(a^0_{Na}\, e^{\frac{\beta_{Na}F\varphi_0}{RT}} + a^0_{Ca}\, e^{\frac{2\beta_{Ca}F\varphi_0}{RT}}\right)} \tag{2}$$

In the equation: σ_0 (C m^{-2}) is the surface charge density of soil particles; ε is the dielectric constant of water, with a value of 8.9×10^{-10} C^2 J^{-1} dm^{-1}.

c. Surface electric field strength E_0:

$$E_0 = \frac{4\pi}{\varepsilon}\sigma_0 \tag{3}$$

In the equation: E_0 (V m^{-1}) is the surface electric field strength.

d. Specific surface area S:

$$S = \frac{N_{Na}\kappa}{ma^0_{Na}} e^{\frac{\beta_{Na}F\varphi_0}{2RT}} = \frac{N_{Ca}\kappa}{ma^0_{Ca}} e^{\frac{\beta_{Ca}F\varphi_0}{RT}} \tag{4}$$

In the equation: S (m^2 g^{-1}) is the specific surface area; m = 0.5259ln($c_{Na}{}^0/c_{Ca}{}^0$) + 1.992; κ (dm^{-1}) is the Debye parameter, and its reciprocal κ^{-1} represents the thickness of the diffusion double layer of colloidal particles, calculated as follows:

$$\kappa = \sqrt{\frac{8\pi F^2 C_0}{\varepsilon RT}} \tag{5}$$

e. Surface charge number (*SCN*):

$$SCN = 10^5 \frac{S\sigma_0}{F} \tag{6}$$

In the equation: *SCN* (cmol(c) kg^{-1}) is the surface charge number.

2. Calculation of soil internal forces

The SIF interactions between soil particles at the mesoscopic scale mainly include electrostatic repulsive pressure (P_E), surface hydration repulsive pressure (P_h), and van der Waals attractive pressure (P_{vdw}). The net inter-particle force (P_{net}) is the combined force of the three, which can be calculated from the soil surface property parameter to obtain the SIFs, calculated as follows.

Electrostatic repulsive pressure (P_E)

$$P_E = \frac{2}{101} RTc_0 \left\{ \cosh\left[\frac{ZF\phi(d/2)}{RT} \right] - 1 \right\} \tag{7}$$

Where P_E (atm) is the electrostatic repulsive pressure; c_0 (mol L^{-1}) denotes the concentration of ions in the native solution; d (dm) is the distance between neighboring particles; φ (d/2) (V) is the midpoint potential at the overlap of the particle bilayers; and Z denotes the valence of the cation.

Surface hydration repulsive pressure (P_h)

$$P_h = 3.33 \times 10^4 e^{-5.76 \times 10^9 d} \tag{8}$$

Where P_h (atm) is the surface hydration repulsive pressure; d (dm) denotes the distance from the particle surface.

Van der Waals attractive pressure (P_{vdw})

$$P_{vdw} = -\frac{A_{eff}}{0.6\pi} (10d)^{-3} \tag{9}$$

Where P_{vdw} (atm) is van der Waals attractive pressure; A_{eff} (J) is Hamaker constant.

Hamaker constant was determined using the method proposed by Tuller and Or. (2005). Weigh 10 g of soil samples sieved through a 2 mm mesh and place them in an aluminum box. Add varying volumes of water to the samples, mix thoroughly, seal

with plastic film, and allow them to equilibrate for one week. Soil water potential at different moisture contents was measured using a dew point water potential meter (WP4-T). The samples were then weighed, dried at 105 ℃, and re-weighed to calculate the soil water content. Hamaker constant was obtained by fitting the soil water potential to the water content through the following model.

$$\theta_m = -\rho_w S\left(\sqrt[3]{\frac{A_{eff}}{6\pi\rho_w g\psi}}\right) \quad (10)$$

Where θ_m (kg kg^{-1}) is the soil water content; ρ_w (kg m^{-3}) is the density of water; S (m^2 g^{-1}) is the SSA of the soil; A_{eff} (J) is the Hamaker constant; and g (m s^{-2}) is the acceleration of gravity; Ψ (m H_2O) is the soil water potential.

Net force (P_{net}):

$$P_{net} = P_E + P_h + P_{vdw} \quad (11)$$

Comment 3: Soil electrochemical properties under biochar treatment for 365 days?

Response: Thank you very much for this helpful comment. The soil electrochemical properties under biochar treatment for 365 days have been provided in the Supporting Information as Supplementary Table S1.

Comment 4: The data in Table 5 should be used the same order of magnitude.

Response: Thank you very much for this helpful suggestion. We have standardized all Hamaker constants in Table 5 to the same order of magnitude, namely $\times 10^{-20}$ J.

Comment 5: Fig. 9, the differences between a and b, c and d?

Response: Thank you very much for this helpful comment. In the revised manuscript, we have clarified the differences among Fig. 9a–d in the figure caption. Specifically, Fig. 9a shows the relationship between aggregate breaking strength for particles < 5

μm and net interparticle pressure at a separation distance of 2 nm under different application rates of maize straw biochar after 180 days. Fig. 9b shows the relationship between aggregate breaking strength for particles < 10 μm and net interparticle pressure under different application rates of wheat straw biochar after 180 days. Fig. 9c and Fig. 9d show the corresponding relationships after 365 days for maize straw biochar with particles < 5 μm and wheat straw biochar with particles < 10 μm, respectively. We have revised the caption accordingly to make the figure easier to understand.