Biochar Addition Affects the Performance of Portland Cement Composites: Meta-analysis and Laboratory Experiments

by

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Abstract

Portland cement is the most widely used binder in construction. However, the production of Portland cement emits a large amount of CO_2, and one strategy to decrease such anthropogenic CO_2 emissions is to reduce the amount of Portland cement produced by partially replacing it with supplementary cementitious materials (SCMs). Biochar is a potential SCM that is a stable porous pyrolytic material, which may improve the performance of Portland cement composites and increase carbon sequestration simultaneously. However, due to limited research, the effects of biochar addition on the performances of Portland cement composites are not fully understood. The performance of different forms of biochar, when mixed with Portland cement composites, has also not been fully synthesized. Such knowledge gaps require a quantitative assessment of the effects of biochar addition, particularly the addition of different biochar types.

Meta-analysis was used to investigate the impact of biochar addition on the 7- and 28-day compressive strength of Portland cement composites based on 606 paired observations. Biochar feedstock type, pyrolysis condition, pre-treatment and modifications, biochar dosage, and curing type all influenced the compressive strength of Portland cement composites. Biochars obtained from plant-based feedstocks (except rice and hardwood) improved the 28-day compressive strength of Portland cement composites by 3-13%. Biochars produced at pyrolysis temperatures higher than 450 °C, with a heating rate of around 10 °C min^{-1}, increased the 28-day compressive strength more effectively. Furthermore, adding biochar with small particle sizes (D_{90} is around 40 μm) increased the compressive strength of Portland cement composites by 2-7% compared to those without biochar addition. Biochar dosage $< 2.5\%$ of the binder weight enhanced both the 7- and

28-day compressive strengths, and common curing methods maintained the effect of biochar addition. However, when mixing the cement, adding fine and coarse aggregates such as sand and gravel affects the concrete and mortar's compressive strength, diminishing the effect of biochar addition and making the biochar effect nonsignificant.

Laboratory experiments were conducted to explore the effects of the addition of different forms of biochar on the performances of Portland cement composites. This study compared oat hull biochar with sawdust biochar, with different particle sizes, to explore their effects on concrete's mechanical performances and durability, including fresh properties, compressive strength, and water sorptivity. The quality of cement paste under the same batching design was also assessed to exclude the aggregate effects. This study found that biochar addition decreased slump by 29% and increased fresh air content by 18% on average. Although it did not affect the overall compressive strength, the smaller particle size and lower biochar dosage significantly increased 28-day compressive strength by more than 20%. In addition, adding biochar decreased initial sorptivity by 39% on average and increased secondary sorptivity by 31% on average but did not affect total water absorption. Moreover, biochar addition altered the quality of cement paste, including setting time, compressive strength, and hardened density. However, since there were only one or two significant relationships among performances of cement paste and concrete and their effect sizes, based on Pearson correlation analysis, the changes in the quality of cement paste did not directly reflect changes in concrete performance.

I conclude that the appropriate addition of biochar could partially improve the mechanical performance and durability of concrete and alter the quality of cement paste. However, it is important to note that not all changes in the quality of cement paste were correlated with performance changes in concrete. Future research should explore and fully understand the mechanisms of biochar effects on the performance of Portland cement composites and how biochar works in the cementitious matrix, especially considering how biochar cooperates with aggregates, and should also consider expanding the selection of different biochars to optimize biochar application.

Preface

This thesis is my original work, aiming to assess the effects of biochar addition on the performance of Portland cement composites. This would improve the understanding of how biochar works on Portland cement composites and provide data for future research. This thesis is a part of a biochar-concrete research project supported by a Natural Sciences and Engineering Research Council of Canada (NSERC) Alliance grant (ALLRP 571094-21), with support from Innovative Reduction Strategies Inc. (IRSI).

The data analysis, interpretation and conclusions presented in Chapters 2 and 3, along with the introduction and literature review in Chapter 1 and synthesis in Chapter 4, are all my original work. Additional information, symbols and abbreviations are listed in the Appendix. I was responsible for laboratory work, data collection, data analysis, and manuscript composition. Dr. Scott X. Chang supervised this work. Dr. Ali El-Naggar, Johnson Kau, Hafiz Asher Muhammad, Chris Olson and Dr. Douglas Tomlinson assisted me. Chapter 2 has been published as:

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“*‘Creativity’ is the principle of novelty.*”

– Alfred North Whitehead

“*Doch alle Lust will Ewigkeit—,*

—will tiefe, tiefe Ewigkeit!”

– Friedrich Wilhelm Nietzsche

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Chapter 1: Introduction and Literature Review

1.1 Overview

The increasing emission of CO_2 from anthropogenic sources causes the most devastating environmental problem in the 21st century, leading to global climate change (Lamb et al., 2021; Olivier, 2022). As one of the most important industries worldwide, construction demand has rapidly increased in the last several decades with the increasing global population, and the demand is expected to increase in the future (Cantero et al., 2020; Uratani and Griffiths, 2023). Concrete is one of the critical construction materials, and Portland cement is the most widely used binder for concrete. The total annual consumption of Portland cement in 2016 was 4.13 Gt, which would grow to 4.68 Gt annually by 2050 (Schneider, 2019). However, a large amount of CO_2 is emitted due to the combustion of fossil fuels and the calcination of carbonaceous matter in raw materials during the production of Portland cement. The CO_2 emission from Portland cement production emits around 5-8% of global anthropogenic carbon emissions (Andrew, 2019; Friedlingstein et al., 2022; Kurda et al., 2018; Miller et al., 2017). Therefore, developing strategies to reduce such emissions is necessary for mitigating the global climate effects of carbon emissions.

Strategies to mitigate global CO_2 emissions from the cement industry include developing novel binders and partially replacing Portland cement to reduce the demand, aiming to increase the sustainability of the concrete industry (Tayebani et al., 2023). For example, calcium aluminate cement and reactive belite-rich Portland cement have been used to substitute conventional Portland cement, and such substitutions have increased the concrete strength and durability, as well as decreasing water demand and carbon emission (Gartner and Sui, 2018; Juenger et al., 2011).

Partially replacing Portland cement with limestone could also help to reduce carbon footprint and maintain concrete performance (Sharma et al., 2021). In addition, partially replacing Portland cement with supplementary cementitious materials (SCMs), including fly ash, silica fume and other such traditional industrial wastes, can be a more practical approach to reduce the demand for Portland cement and maintain concrete performances, which would also help recycle the industrial waste (Al-Yousuf et al., 2021; Juenger et al., 2019). However, considering the current construction structure and the immature development of novel binders, replacing Portland cement would be the optimal approach to mitigate carbon emissions and control costs. Furthermore, traditional SCMs do not contribute to carbon sequestration (Miller, 2018). Such situations indicate the demand for novel SCMs to partially replace Portland cement to maintain concrete performances and improve the carbon neutrality of the cement industry.

Biochar has become a novel material that can be used as a potential SCM. It is a stable porous pyrolytic material produced from waste organic matter, including waste plant materials and industrial sludge, and the main composition is aromatic forms with various organic functional groups and metals (Amalina et al., 2022; Chen et al., 2019; Liu et al., 2015). The production of biochar recycles waste and pollutants, as well as produces bio-oil and syngas. Biochar application has been reported to increase carbon sequestration compared to conventional waste management, such as landfills and incineration (Chen et al., 2024; Chen et al., 2021; You et al., 2018). Researchers have dabbled the biochar potential in environmental remediation and soil science. For example, biochar was found to improve soil physical and chemical properties, improve seed germination, and retard soil nutrient leaching in the short term due to its alkalic properties and high ion exchange capacity (Aller, 2016; Joseph et al., 2021). It was also reported that biochar could immobilize heavy metals and degrade organic contaminants in the environment through absorption and activating degradation with redox reactions through its inherent reactive-active moieties (Ji et al., 2022; Wan et al., 2020). Such research indicated that biochar is expected to achieve more potential, for example, in construction.

Preliminary results reported that the use of biochar at low dosage would help to improve Portland cement composites' mechanical performance, including compressive strength and flexural strength (Gupta and Mahmood, 2022; Qin et al., 2021; Zhang et al., 2022a), and durability, including freeze-thaw and chloride resistances (Gupta et al., 2021b; Sikora et al., 2022; Yang and Wang, 2021a), with decreases in Portland cement composites' water permeability (Gupta et al., 2020b, 2018a) and shrinkage (Dixit et al., 2021; Mobili et al., 2022). Such improvements were driven by accelerated cement hydration as the porous structure of biochar could provide nucleation sites for cement hydration and space for mitigating water loss (Khan et al., 2022; Restuccia and Ferro, 2016; Tan et al., 2022). Some life cycle assessments also reported that adding biochar into concrete would decrease the total carbon emissions compared to conventional concrete usage (Chen et al., 2022b, 2022a). These results indicated that biochar could be used as an SCM for construction to partially replace Portland cement and mitigate carbon emissions.

On the other hand, however, researchers found that biochar addition did not consistently improve the performance of Portland cement composites. For example, adding biochar derived from poultry litter decreased the 28-day compressive strength of concrete by 25% compared to the control (Akhtar and Sarmah, 2018). In addition, different feedstock and production conditions would significantly affect the performance of Portland cement composites; for example, biochars produced from barley straw performed better than those made from manure in improving the 28-day compressive strength of concrete, with a 9% increase in adding biochar produced at 500 °C compared to that of the biochar produced at 350 °C (Zhang et al., 2022b). Researchers also found that the effects of biochar addition would be affected by the batching design of Portland cement composites, with the mechanical improvement decreased with increasing biochar dosage and varied results of water/cement ratio (Gupta et al., 2018a; Laili et al., 2017). Such results suggested that biochar had heterogeneous effects on the cementitious matrix, which required explanations based on the biochar addition mechanisms.

Researchers have concluded several potential mechanisms of how biochar performs in cementitious matrices. For example, biochar could act as a water maintainer and nucleation sites for cement hydration in cementitious matrix, which could accelerate and maintain cement hydration in temporal series to have higher degrees of hydration and densified cementitious matrix (Senadheera et al., 2023; Zhang et al., 2022). However, no research has assessed the correlations between biochar properties and performances of Portland cement composites. In addition, no research collected all research articles relating to particular topics. For example, researchers once concluded that the biochar dosage should be around 2% of binder weight to optimize the mechanical improvement by biochar addition (Maljaee et al., 2021a); however, this conclusion was only based on results of around 15 research articles. Furthermore, many biochar types have yet to be explored, requiring researchers to expand the research scope and improve the understanding of this field. Therefore, assessing how biochar addition alters the performances of Portland cement composites and identifying the knowledge gaps in this field is necessary.

Meta-analysis could be used to quantify the effects of biochar addition, and it has been widely used in ecology and medicine (Arnqvist and Wooster, 1995; Hartung et al., 2008; Hernandez et al., 2020). Meta-analysis is a big data analysis that collects extensive data from individual studies on a particular research topic to assess the overall effect numerically and generalize the specific topic. Such a method could avoid the small sample size problem, as all papers meeting particular filtering criteria will be included in the analysis. A recent meta-analysis focused on the effects of cement and aggregate replacement on the mechanical performance of Portland cement composites (Anwar et al., 2022), but it did not provide details on the properties of the mixtures, especially for biochar, so their meta-analysis was undesirable for providing suggestions to future research or practices. Therefore, it is necessary to conduct further research to explore and provide quantitative evidence on how biochar affects the performances of Portland cement composites and provide guidance for biochar selection.

Mechanical performances of Portland cement composites could be assessed through selected parameters for a meta-analysis. Compressive strength is the capacity for Portland cement composites to resist loads tending to reduce their size, which could represent the mechanical performances since this parameter is empirically significantly correlated with other mechanical performances, including flexural strength and split tensile strength, and this parameter is also reported to indirectly relate to other performances parameters, including hardened density, porosity and durability (Kosmatka and Wilson, 2011; Neville, 1981). Therefore, this parameter could represent the overall performance of Portland cement composites.

1.2 Research objectives

Overall, although biochar has potential benefits as an SCM, there is no assessment to synthesize how biochar production conditions and properties correlate to the performances of Portland cement composites. Also, a limited selection of forms of biochar used for research restricts the expansion of the understanding of biochar addition and its application in practice. Based on the existing knowledge gaps, an assessment of how biochar addition affects the mechanical performances of

Portland cement composites and an expansion of selected biochar types is required to improve the understanding of mechanisms of how biochar addition alters the performances of Portland cement composites. Therefore, this thesis has the following objectives on how biochar addition affects the performance of Portland cement composites:

(1) To perform a meta-analysis to explore how biochar addition affects the compressive strength of Portland cement composites, considering biochar production conditions and properties with the batching design of Portland cement composites.

(2) To test the effect of adding oat hull and sawdust biochars on concrete's compressive strength and water sorptivity.

(3) To measure fresh properties and the compressive strength of cement paste in the same water/binder ratio with concrete to explore how biochars affect binder properties without considering the aggregate effects.

(4) To conduct correlation analyses to explore the relationships between concrete and cement paste performances and between their effect sizes and to check if biochar addition has consistent effects among performance parameters of concrete and cement paste.

1.3 Thesis structure

This thesis contains two data chapters: a meta-analysis chapter and a chapter based on laboratory experiments. The research was performed progressively, with the design of the laboratory experiments dependent on the meta-analysis results.

Chapter 1 (this chapter) is a brief introduction and literature review of this thesis and the research topics covered in this thesis. The research objectives and structure of this thesis are also described in this chapter.

Chapter 2 presents the meta-analysis work, which evaluated how biochar addition affects the compressive strength of Portland cement composites. The meta-analysis also explored the effects of factors such as biochar pyrolysis condition, biochar pre-treatment and modification, and the batching design of Portland cement composites, including biochar dosage, curing type, and forms of composites, on the compressive strength of Portland cement composites.

Chapter 3 reports the results and discussion of the laboratory experiments, including exploring the effect of the addition of oat hull biochars and sawdust biochars on concrete's 7- and 28-day compressive strength, initial and secondary sorptivity, as well as the cement paste's compressive strength, flow and setting time. The relationships among these performances and their effect sizes are also presented to check if biochar addition has consistent effects on concrete and cement paste.

Chapter 4 summarizes key findings from Chapters 2 and 3, highlights the conclusions of the thesis research, describes the originality of this thesis, and discusses the research limitations. It also outlines perspectives on future research opportunities to explore mechanisms of biochar effects on concrete performances and environmental and economic assessments.

Appendix A presents additional information, including the references used in the meta-analysis and supplementary results for Chapter 2. Appendix B presents the list of recurrent symbols and abbreviations.

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Chapter 2: Biochar Affects Compressive Strength of Portland Cement Composites: A Meta-analysis

2.1 Introduction

Anthropogenic emissions of CO_2 have resulted in one of the most devastating environmental problems in the 21st century (Lamb et al., 2021; Olivier, 2022). Efforts are being taken to enhance carbon sinks in terrestrial ecosystems, including urban and other highly human-influenced environments, to attain carbon neutrality and mitigate climate change (Wang et al., 2021). After fossil fuel usage and land-use change, cement production is one of the most significant anthropogenic carbon emissions, accounting for almost 5% (4000 Mt) of global anthropogenic CO_2 emissions in 2019 (Andrew, 2019; Friedlingstein et al., 2022). Replacing cement with supplementary cementitious materials (SCMs), such as fly ash, silica fume and waste glass, could help reduce anthropogenic CO_2 emissions and improve concrete performance (Li et al., 2022; Mehta and Ashish, 2020; Miller et al., 2021). However, traditional SCMs are industrial by-products that will be less available in the future, and these materials do not contribute to carbon removal. Therefore, novel SCMs need to be designed to fill the potential gap.

Adding biochar to Portland cement composites (a general term for cement paste, mortar, and concrete in this article) has emerged as a potential solution to providing the needed SCMs and removing excessive carbon from the atmosphere. Biochar is a stable porous pyrolytic material produced from feedstocks such as waste plant materials and industrial sludge, reducing the release of waste and pollutants (Amalina et al., 2022; Chen et al., 2019). Biochar can improve soil physical and chemical properties, mitigate greenhouse gas emissions, and remediate soil pollution, among other benefits (He et al., 2022; Osman et al., 2022; Singh et al., 2022). Using biochar as SCMs in civil engineering is an emerging field supported by promising results (Danish et al., 2021; Singhal, 2023; Tan et al., 2021). For instance, biochar addition to Portland cement composites has been shown to promote heat evolution of hydration (Sikora et al., 2022; Zhang et al., 2022). Adding wood-derived biochars at 0.5 and 2% (by weight) of cement improved 28-day compressive strength by 16 and 9%, respectively, and decreased the water permeability of concrete by 40% at a dosage of 2% (Gupta et al., 2020b). These results were consistent with the observed dense interfacial transition zone (ITZ) between biochar and cement paste, suggesting that biochar promoted the hydration process (Dixit et al., 2019). The internal curing of biochar assisted the cement hydration process by absorbing and releasing water (Gupta and Mahmood, 2022). Biochar addition may also improve concrete thermal conductivity and electromagnetic shielding capacity (di Summa et al., 2023; Ryms et al., 2022). Biochar addition increased the carbon sequestration of Portland cement composites without significantly increasing the composite production cost (Gupta et al., 2018c; Praneeth et al., 2020). Overall, adding biochar to Portland cement composites could improve their performance and increase the carbon sink to mitigate climate change.

Some studies, however, reported adverse effects of biochar addition on the performances of Portland cement composites. For example, Akhtar and Sarmah (2018) found that adding litter-derived biochar decreased 28-day compressive strength, which can be attributed to reduced hydration products due to the dilution effect of biochar addition. The biochar type and production condition also affect the potential of biochar to enhance the performance of Portland cement composites. For instance, biochars produced from barley straw performed better than manure biochars in improving the 28-day compressive strength of concrete (Zhang et al., 2022b), and adding biochars produced at high pyrolysis temperatures resulted in higher compressive strength of cement mortar than biochars produced at low pyrolysis temperatures (Gupta and Kua, 2018), demonstrating that biochar properties would significantly affect the performance of Portland cement composites.

Several review papers have discussed biochar effects on Portland cement composites' mechanical properties (Maljaee et al., 2021a; Senadheera et al., 2023; Zhang et al., 2022), but these papers did not review all related literature on this topic, which may cause sampling error and imprecise conclusions. In addition, comprehensive evaluations of biochar addition effects are needed to reconcile contradictory results of biochar effects on the mechanical properties of Portland cement composites. Meta-analysis collects an extensive data set from individual studies on a particular research topic to assess the overall effect numerically and boost the generalization of the collected data; meta-analysis has been widely used in ecology and medicine (Arnqvist and Wooster, 1995; Hartung et al., 2008; Hernandez et al., 2020). A recent meta-analysis focused on the effects of cement and aggregate replacement on the mechanical performance of Portland cement composites (Anwar et al., 2022); however, it did not provide details on the properties of the mixtures, especially for biochar, making it difficult to optimize the selection of the mixtures. Therefore, it is necessary to conduct further research to explore and provide quantitative evidence on how biochar properties affect the mechanical performance of Portland cement composites and provide guidance for biochar selection for further research and industrial applications.

Meta-analysis was used to quantify the effect of biochar addition on the mechanical performance of Portland cement composites based on 606 paired observations from 51 peer-reviewed papers. The effect size was calculated using 7- and 28-day compressive strengths, which are crucial quality indices for the performance of Portland cement composites and correlated with other performance indicators, such as flexural strength, hardened density, and water permeability

(Kosmatka and Wilson, 2011). This study considered biochar pyrolysis conditions, including pyrolysis temperature, heating rate and residence time, biochar pre-treatment and modification, biochar dosage, concrete curing type, and cementitious matrices. I hypothesized that: (1) the effect of biochar addition on the performance of Portland cement composites is influenced by biochar pyrolysis condition; (2) biochar pre-treatment and modification impact the effects of biochar addition; (3) the Portland cement composite batch design, including biochar dosage, curing type and forms of composite, influences the effect of biochar addition. This study aims to provide quantitative evidence of the effects of biochar production conditions, biochar properties and Portland cement composite batching designs on compressive strength with illustrations of the potential mechanisms of biochar addition effects.

2.2 Methods

2.2.1 Literature search

The data of 7- and 28-day compressive strengths of Portland cement composites for this study were collected from peer-reviewed research papers via Web of Science and Scopus using the following search terms: “biochar” AND “cement” AND “compressive strength.” Papers related to Portland cement composites used as building materials were shortlisted by reviewing the titles and abstracts, with papers on soil and environmental remediation excluded from further data collection and analysis. A total of 387 papers were initially screened; they were filtered to include those that measured 7- and 28-day compressive strengths by scanning abstracts and figures in each paper. This filtration excluded some papers where: 1) biochar was not the only SCMs; 2) microbes were introduced to biochars; 3) biochars were not produced through pyrolysis; 4) incomplete statistical data; 5) Portland cement used in the study was not ordinary Portland cement. Finally, 51 papers were included for the meta-analysis based on papers published before December 1st, 2023, with 41 and 48 papers including 7- and 28-day compressive strength, with 254 and 352 paired observations, respectively (Table S1). Data from collected papers were organized as paired-observation datasets. Each paired observation was treated as one record. Pyrolysis conditions, biochar properties, and Portland cement composite batching design information were extracted from the literature.

2.2.2 Data compilation

The means, standard errors/deviations (SE/SD), and the number of replicates (n) for 7- and 28-day compressive strength were extracted from each reference. The compressive strength units were megapascal (MPa), and both control and treatment groups were recorded. All SE were converted into SD via the equation: $SD = SE * \sqrt{n}$. Data with missing SE/SD were less than 15% of the total data. They were estimated using an imputation method, where the weighted average of SD from the other records was used to estimate the imputed SD (Bracken, 1992). Data presented in figures in the literature were extracted through the OriginPro software.

Biochar feedstocks were categorized into plant and organic waste groups. The plant group includes agricultural and forestry plant materials, and the organic waste group includes sludge and manure. Main food crop residues and wood waste are often selected as they are primary sources of biochar. Some feedstocks (including wood materials, which were not indicated as hardwood or softwood) were unknown or had a small sample size (for example, bamboo, bagasse, and peanut), and they were categorized as "Other plant materials." In addition, several feedstock materials from the same origin (including corn, rice, and wheat) were combined as one feedstock source to satisfy the sample size requirement for meta-analysis. Pyrolysis temperature was divided into four groups: “<350,” “350-450,” “450-550,” and “>550” °C. The pyrolysis heating rate was divided into three groups: “0-5,” “5-10,” and “>10” °C min^{-1}. For pyrolysis residence time (min), this analysis used three groups of “<60,” “60-180,” and “>180” to represent short, medium and long residence time, respectively. Biochar pre-treatments were categorized into only physical and chemical treatments. Reducing the particle size was the primary physical modification, including using ball milling, sieving and manual grinding, which was treated as grinding. Chemical treatments, as detailed in the papers selected for the meta-analysis, were recorded.

The factors considered for mixture design were biochar dosage, concrete curing type and cementitious matrices. Biochar dosage was calculated as the ratio of the biochar weight and the binder (cement + biochar) weight, shown as “% of binder weight" or “to binder weight.” The curing type was divided into “carbonization,” “seal,” “dry,” and “wet.” “Carbonization” represented curing the composites in an environment with a high concentration of carbon dioxide; “seal” represented blocking the composites away from the external environment during curing; “dry” represented curing the composites under an ambient environment; and “wet” represented curing the composites under high humidity or submerged environment. Cementitious matrices represented the forms of Portland cement composites, divided into “cement paste,” “mortar,” and “concrete.”

2.2.3 Data analysis

The meta-analysis used a log-transformed ratio to analyze the effect size of performance parameters by biochar and batching variables (Chen et al., 2022; Hedges et al., 1999). Each collected paper was treated as having homogeneous experiment conditions. The individual effect size was calculated according to Eq. (2.1):

$$L = lnRR = ln\,(\frac{\bar{X}_t}{\bar{X}_c}) \tag{2.1}$$

$\bar{X}_t$ is the mean of the treatment group, which is the group that added biochar into cementitious matrices; $\bar{X}_c$ is for the control group without biochar addition. Positive values of L or $lnRR$ represent an increase in compressive strength compared to the control group and vice versa. Then, the variance of individual effect size was calculated using Eq. (2.2):

$$v = \frac{{S_t}^2}{n_t{\bar{X}_t}^2} + \frac{{S_c}^2}{n_c{\bar{X}_c}^2} \tag{2.2}$$

S_t and S_c are standard deviations of the treatment and control group; n_t and n_c are sample sizes of the treatment and control group. Considering the effect of sample size and variance, a weighted mean of effect size for each categorized parameter was calculated to obtain an overall effect response of each factor (Eq. (2.3)):

$$L_w = \frac{\sum_{i=1}^{k} w_i L_i}{\sum_{i=1}^{k} w_i} \tag{2.3}$$

k is the number of paired data points; w_i is the weighting factor, which is sensitive to v_i (Hedges et al., 1999). After the weight effect size was calculated, a 95% confidential interval (95CI) was calculated using Eq. (2.4):

$$95CI = L_w \pm 1.96se_{L_w} \tag{2.4}$$

se_{L_w} is the standard error of L_w. When 95CI does not overlap 0, the effect is significant. To intuitively illustrate the effect, the log response ratio was back-transformed to a natural response ratio in percentage using Eq. (2.5), and it is called an effect index:

$$Effect\ index\ (\%) = (e^{lnRR} - 1) \times 100 \quad (2.5)$$

The meta-analysis was processed using the metagear package in R, and all correlation analyses in this study were simple linear regressions conducted using ggplot2 in R, with the following equation (Eq. (2.6)):

$$L = lnRR = \beta_0 + \beta_1 x + \varepsilon \quad (2.6)$$

β_1 is a coefficient; x is the factor; ε is the sampling error. Based on the database size in this study, the regression would only be used for factors with at least 100 paired comparisons.

2.3 Results and discussion

Overall, biochar addition to Portland cement composites did not reduce the 7- and 28-day compressive strengths (Figure A1). Moreover, the effect sizes of 7- and 28-day compressive strengths were positively correlated ($R^2 = 0.72$, $p < 0.01$) (Figure A2), indicating that biochar addition would maintain its effect on compressive strength throughout the curing process. However, as the overall effect sizes for 7- and 28-day compressive strengths had high heterogeneities ($p < 0.01$ for both parameters), the potential of biochar to maintain the compressive strength of Portland cement composites varied significantly with biochar type and pyrolysis condition, as well as the batching design.

2.3.1 Effects of biochar characteristics

2.3.1.1 Feedstocks

The effect of biochar produced from different feedstocks on the compressive strength of Portland cement composites was inconsistent. In particular, biochar produced from corn significantly increased the 7-day compressive strength by 17% (Figure 2.1, Table 2.1), whereas most plant-based biochar types increased the 28-day compressive strength by 3-13% (Figure 2.2, Table 2.2). However, biochars produced from rice residues did not affect the compressive strength after 7 or 28 days. Meanwhile, biochars produced from forestry materials exhibited contradictory effects, as softwood biochars increased the 7- and 28-day compressive strength by 12 and 7%, respectively, while hardwood biochars decreased them by 22 and 23%, respectively (Figures 2.1 and 2.2; Tables 2.1 and 2.2). Biochars produced from manure decreased the 7-day compressive strength by 26%; however, this effect was minimal after 28 days. Finally, biochars produced from sludge did not increase or decrease the compressive strengths of Portland cement composites.

The superiority of biochars produced from agricultural feedstock over other feedstock types in increasing the compressive strength of Portland cement composites could be attributed to their low molar oxygen/carbon (O/C) ratio, primarily caused by the low carbon contents of biochars (Tables 2.1 and 2.2), as lower molar O/C ratio is associated with higher biochar hydrophobicity due to its low content of oxygen-containing functional groups (Hassan et al., 2020; Xing et al., 2019; Zhao et al., 2013). Other researchers found that hydrophobic silica fume could accelerate cement hydration due to more available water surrounding cement particles, offsetting the negative effect of the larger particle size of the hydrophobic silica fume, indicating accelerated cement hydration under high hydrophobicity (Jeong et al., 2020). Biochars produced from plant wastes had lower molar O/C ratios than manure and sludge biochars (Tables 2.1 and 2.2), contributing to their high hydrophobicity and potential for enhancing cement hydration. However, agriculture-sourced biochars increased 7- and 28-day compressive strengths, but forestry-sourced biochars did not, even though forestry-sourced biochars had higher carbon contents (76 and 76%, respectively in 7- and 28-day compressive strength) than agriculture-sourced biochars (43 and 56%, respectively) (Tables 2.1 and 2.2). As forestry-sourced biochars had a more macroporous structure than agriculture-sourced biochars due to their high lignin content (El-Naggar et al., 2022), agriculture-sourced biochars have a highly mesoporous structure, which may contribute to considerable water-absorption-release capacity, where biochars absorb water in the early curing stage to densify the cementitious matrix and then desorb water in response to a humidity gradient to maintain cement hydration (Khan et al., 2022), increasing the compressive strength. However, due to the lack of data, it is not possible to conclude the different effects between hardwood and softwood biochar addition. More research is needed to better understand the mechanisms involved.

Ash content, which includes oxides, could also affect cement hydration. Amorphous silica oxide (SiO_2) was the most critical oxide for cement hydration, contributing to the pozzolanic reaction, in which SiO_2 would consume $Ca(OH)_2$ to form calcium silicate hydrate (C-S-H) to enhance the growth of strength (Thomas, 2011; Zhang et al., 2020; Zhou et al., 2020). Other oxides, such as Fe_2O_3 and Al_2O_3, can negatively and positively, respectively, affect cement hydration (Stephan et al., 2008). In this study, biochars produced from plant sources had a relatively high Si concentration (0.4% on average). The SiO_2 can make up most of the oxides (19% on average) in the 28-day compressive strength measurement. In contrast, manure biochars contained the least Si, causing the least positive effect from the pozzolanic reaction (Table 2.2). These results illustrate that the positive effect of Si on 28-day compressive strength was better than other types of biochar. Similarly, Si and SiO_2 contents of biochars produced from plant sources for 7-day compressive strength measurement were the highest compared to other biochars (1 and 22% on average, respectively; Table 2.1), indicating relatively intense pozzolanic reactions during early cement hydration.

Overall feedstock effects on compressive strength demonstrated that biochars produced from plant wastes (except rice) had significant positive effects, and manure biochars had significant adverse effects on 7-day compressive strength, as manure biochars had a higher molar O/C ratio than the plant-based biochars. Meanwhile, elements and oxide content, especially for Si, which contributed to the pozzolanic reaction, could promote cement hydration. However, more data was required to analyze the effect of feedstock type and oxides on cement hydration to validate the above findings.

2.3.1.2 Pyrolysis condition

Adding biochars produced at pyrolysis temperatures between 450 and 550 °C significantly improved 7-day compressive strengths by 5%, but biochars produced between 350 and 450 °C decreased this parameter by 8% (Figure 2.1, Table 2.1). Biochars produced at pyrolysis heating rates between 5 and 10 °C min^{-1} improved 7-day compressive strength by 6%. On the other hand, adding biochars produced at a temperature higher than 450 °C significantly improved 28-day compressive strength by more than 4%, and biochars produced at higher heating rates more significantly increased 28-day compressive strength compared to lower rates (Figure 2.2, Table 2.2). However, pyrolysis residence time did not affect the compressive strengths of Portland cement composites.

Pyrolysis temperature and heating rate highly affected biochar properties, including molar O/C ratios and specific surface areas. In this study, both compressive strengths negatively correlated to the molar O/C ratio, while positively correlated to the specific surface area of biochars (Figure 2.5). Biochars produced at temperatures between 450 and 550 °C had a relatively lower molar O/C ratio (Tables 2.1 and 2.2), as higher temperature conditions would decompose organic substances, increasing C content and decreasing O content, resulting in a decreased molar O/C ratio (Ghodake et al., 2021). However, the molar O/C ratio of biochars produced at 350 °C was similar to biochars produced between 450 and 550 °C with different effects. In this study, most of the biochars produced below 350 °C were forestry-sourced biochars with high lignin contents, leading to a relatively low molar O/C ratio, while other temperature categories comprising other feedstocks with relatively low C content (Tables 2.1 and 2.2). Meanwhile, biochars with high specific surface areas, such as biochars produced at temperatures < 350 °C and between 450 and 550 °C in this study (Tables 2.1 and 2.2), could provide more nucleation sites for cement hydration, contributing to more hydration products (including C-S-H) to increase compressive strength (Restuccia and Ferro, 2016; Zhang et al., 2022). However, organic matter would be left in biochars produced at low temperatures due to uncompleted decomposition, such as fatty acids and residual saccharides (Chen et al., 2022; Das et al., 2021; Gupta et al., 2020a, 2020c; Muthukrishnan et al., 2019). These organic matters would retard cement hydration (Choi and Choi, 2021; Kochova et al., 2017), counteracting the benefits of the high specific surface area. In this case, biochars produced at a lower temperature did not improve compressive strength. However, excessive pyrolysis temperature (> 500 °C) could damage the biochar pore structure to break water absorption and release capacity, which was a counterproductive effect (Fu et al., 2012), and this finding could indicate the insignificant effects of high-temperature (> 550 °C) biochar addition on

7-day compressive strength. As for the pyrolysis heating rate, rates between 5 and 10 °C min^{-1} had a relatively high specific surface area and low molar O/C ratio in 7- and 28-day compressive strength measurements (Tables 2.1 and 2.2). Slow pyrolysis (heating rate < 50 °C min^{-1}) would result in a higher biochar yield. In contrast, fast pyrolysis would produce more oil and gas phases due to secondary reactions of decomposed polysaccharides, reducing the yield of the solid phase (Al-Rumaihi et al., 2022; Chen et al., 2021; Ghodake et al., 2021). This information indicated that the mild pyrolysis heating conditions would retard biomass gasification and liquefication, where biochars could maintain their structure of carbon skeleton to benefit cement hydration.

The overall effect of pyrolysis condition on 7- and 28-day compressive strengths illustrated that high pyrolysis temperature would improve 7- and 28-day compressive strengths. Additionally, medium heating rates could significantly improve the 28-day compressive strength, which was highly negatively correlated to biochar molar O/C ratio and positively correlated with specific surface area. It is necessary to enlarge the research range to include biochars produced under different conditions to select optimal biochars for altering the performance of Portland cement composites.

2.3.1.3 Biochar modification and pre-treatment

Grinding was the primary physical modification of reducing particle size; most grinding was done through ball milling. Biochar grinding did not reduce the 7-day compressive strength but increased the 28-day compressive strength by 7% (Figures 2.1 and 2.2). However, biochars without physical modification will decrease the 28-day compressive strength. Biochar grinding could reduce the biochar particle sizes (D_{90} is around 45 μm), similar to or smaller than the cement particles (D_{90} is around 40 μm), compared to biochars without this modification (D_{90} is around 200 μm) (Tables A2 and A3). Such tiny particles could fill the ITZ between cement particles and aggregates as a filler and improve the Portland cement composites' compressive strengths as nucleation sites due to the enlarged specific surface area (Dixit et al., 2019; Gupta et al., 2020; Yang and Wang, 2021). In the early curing stage, the reduced water/binder ratio, by water absorption of biochars, had a higher effect than the filler effect, and water would be desorbed later due to the humidity gradient (Gupta, 2021), maintaining the cement hydration. In the later curing process, most cement particles reacted, and the filler effect was superior to the water absorption and release effect. Research on carbon nanotubes, which were nano-size carbon materials that could considerably increase concrete compressive strength, could also provide valid evidence that tiny particle size would be beneficial to increase compressive strength (Silvestro and Gleize, 2020; Zhang et al., 2023). However, as grinding could destroy the original pore structure of biochars, this modification would retard the function of nucleation at the early curing stage. As grinding could destroy macropores with less water absorption and release capacity, biochar's water-holding capacity and filler effect would increase compressive strength later, offsetting the retardance in the early curing stage.

Presoaking biochars with water and other pre-treatment methods did not affect either compressive strength (Figures 2.1 and 2.2). However, only two studies in the database of this paper reported the effect of pre-treating biochar with water on its potential to enhance compressive strength, which made it hard to evaluate the effect of presoaking biochar. For instance, Gupta and Kua (2018) presoaked biochar with water to provide additional water to mortar, and they found an improvement in 28-day compressive strength. However, Jafari et al. (2023) reported that water-presoaking treatment could not counteract the negative effect of high biochar dosage, but such a decrease could be mitigated by combining the high biochar dosage with other SCMs, such as MgO expansive additives (Mo et al., 2019). Another paper, not included in this meta-analysis, indicated that presoaking biochar could maintain concrete strength in the long term, showing the potential benefit of presoaking biochar (Sirico et al., 2022). Haque et al. (2021) also mixed biochars with stearic acid when grinding to obtain super-hydrophobic surface characteristics. However, the high biochar dosage did not affect the 28-day compressive strength, as further discussed in Section 2.3.2.1. Other biochar pre-treatments were also applied, such as carbon dioxide pre-dosage (Gupta et al., 2018b), melamine pre-treatment (Jeong et al., 2022), alkaline electromagnetic pre-treatment (Beskopylny et al., 2022) and acid pre-treatment (Zeidabadi et al., 2018), which introduce additional substances or oxygen-contained functional groups to biochar surfaces to alter its physical and chemical properties, but they were not included in this article due to the small sample size. The lack of data limited further analysis of pre-treatment effects on the compressive strength of Portland cement composites.

Overall, the effects of biochar modification and pre-treatment methods on compressive strength demonstrated that modifying biochar with an appropriate method could enhance their potential to improve the compressive strength of Portland cement composites. Grinding could improve the 28-day compressive strength due to the filler effect. However, as data on biochar modification and pre-treatments are scarce, future studies need to explore this field to better understand the effects of biochar addition on the performance of Portland cement composites.

2.3.2 Effects of batching design and curing

2.3.2.1 Batching dosage of biochars

Low biochar dosages (< 2.5% of binder weight) increased 7- and 28-day compressive strengths by 6 and 7%, respectively (Figures 2.3 and 2.4). However, higher biochar dosages negatively impacted compressive strength (Figure 2.6) due to its porous structure, which could not strengthen Portland cement composites (Mohan et al., 2014). In addition, higher biochar doses may dilute cement hydration products and cause agglomeration (Mota-Panizio et al., 2023). In particular, biochar would agglomerate through van der Waal's forces when its dosage was more than 5% of cement weight (Gupta et al., 2018a; Maljaee et al., 2021b), indicating that biochar may be poorly dispersed in the cementitious matrix when applied at a higher dosage, leading to a heterogeneous composition and structure of cementitious matrices. Furthermore, excessive biochar dosage would compete with cement to absorb water, which would retard the cement hydration process and strength growth (Tan et al., 2022). However, the adverse effects of biochar at different dosages on compressive strength may also be due to the damaging effects of the pyrolysis process on the biochar pore structure (Zhang et al., 2022b). As mentioned in previous sections, the undecomposed matter left in biochar after pyrolysis would also negatively affect the cement hydration process. Forestry-sourced biochar, containing less Si, might not trade off the dilution effect (Akhtar and Sarmah, 2018; Ghodake et al., 2021). Therefore, although a low biochar dosage would increase the compressive strength of Portland cement composites, other factors, including those mentioned above, might offset the low-dosage benefits.

Overall, the results suggest that the optimal biochar dosage to improve the compressive strengths of Portland cement composites is less than 2.5% of binder weight due to the filler effect, nucleation effect and potential pozzolanic reaction. However, other factors might counteract the benefits of low biochar dosage, indicating that careful consideration of biochar addition to Portland cement composites is needed.

2.3.2.2 Cement curing and cementitious matrices

Wet and dry curing, the most common curing method, did not affect both compressive strengths. Carbonization curing was the most effective method for improving compressive strength, leading to 30 and 21% increases in 7- and 28-day compressive strengths, respectively (Figures 2.3 and 2.4). Carbonation curing might convert cement composites and hydration products into densified carbonatized composites and silica chains; however, it would consume water, which requires subsequent wet curing to continue the cement hydration process (Chen et al., 2022; Liu and Meng, 2021). However, this curing method needs to be carefully considered as it requires the use of CO_2, especially in steel-reinforced concrete; it would destroy the passive layer around steel reinforcement to worsen corrosion and decrease mechanical performance (Kua and Tan, 2023; Marques et al., 2013; Tapan and Aboutaha, 2011). Sealed curing, on the other hand, had adverse effects, with a 22% decrease in both 7- and 28-day compressive strengths. Sealed curing would physically cover the surfaces of the Portland cement composites to prevent water loss, and the internal curing of biochar would improve compressive strength (Maljaee et al., 2021; Wang et al., 2019). However, the decreased effect of biochar addition in this study indicated that other factors would cooperate with curing, including biochar particle size (Yang and Wang, 2021) and dosage (Haque et al., 2021), where the negative effects of particle size and dosage counteracted the positive effect of sealed curing in this study.

Cementitious matrices did not affect compressive strengths (Figures 2.3 and 2.4). It was expected that biochar addition would increase the compressive strength of cement paste, based on the degree of cement hydration, as discussed in previous sections. However, the presence of salts, such as sodium salt and sylvite, in biochar would cover biochar or interact with C-S-H, retarding cement hydration or destroying the C-S-H structure (Gupta et al., 2021; Maljaee et al., 2021; Restuccia and Ferro, 2016). These mechanisms counteracted the benefits of biochar addition to cement paste. When coexisting with aggregates, biochar, especially ground biochar with tiny particle sizes close to cement particles, could accelerate hydration and work as fillers to fill the pores of the ITZ between aggregates and cement paste (Park et al., 2021; Scrivener et al., 2004). However, one hypothesis suggested that filling pores might make other tiny particles gather around fine aggregates, providing a convenient route for cracking and decreasing the compressive strength (Aziz et al., 2023). Although requiring validation, such a viewpoint indicated more complex mechanisms of biochar effects, requiring complex models to describe it. In addition, various factors affected mortar and concrete's compressive strength, including water/binder ratio, coarse aggregate properties, coarse aggregate amount and ITZ properties; effects of these factors indicated alteration of cement quality would not significantly affect concrete and mortar's strength (Maso, 1996; Scrivener et al., 2004; Sims et al., 2019). Therefore, studies on the effects of biochar addition to mortar and concrete should address specific composite types, such as lightweight concrete and ultra-high-performance concrete.

Overall curing effects on compressive strength illustrated that carbonation curing would promote the effects of biochar addition. On the other hand, the composition of cementitious matrices did not impact the compressive strength of Portland cement composites, indicating that specializing in matrices in future research is necessary to provide more detailed and precise information on biochar effects on Portland cement composites.

2.4 Conclusions

Overall, I conclude that adding pyrolytic biochars did not decrease the compressive strength of Portland cement composites. Plant-based biochars, rather than organic-waste biochars, are ideal for addition to Portland cement composites. I recommend that biochars should be produced at high temperatures (> 450 °C) with a slow pyrolysis rate (around 10 °C min^{-1}) to optimize the positive effects of biochars on Portland cement composites. The reduced particle size of biochars, at least similar to cement particle size (D_{90} is around 40 μm), is recommended to accelerate the cement hydration process; water-presoaking of biochars provide more available water for cement hydration, but more research is required to valid the benefits. The low molar O/C ratio and high specific surface area of biochars were highly correlated to the improvement effects of biochar addition, which were substantially affected by biochar feedstock type, pyrolysis condition and pre-treatment. On the other hand, low biochar dosages (< 2.5% of binder weight) improved compressive strength. Biochars also cooperated with aggregates to affect compressive strength, but the performance of biochar addition on concrete and mortar is highly context-specific due to the complexity of compositions and properties of aggregates.

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Table 2.1 Effect indexes and critical properties of biochar used for 7-day compressive strength measurements, including means and numbers of records.

Biochar variables	Effect index (%)		C content (%)		Molar O/C ratio		Specific surface area (m^2 g^{-1})		Si concentration (%)		SiO_2 concentration (%)	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Feedstock												
Corn	17	35	-	-	-	-	483	24	-	-	4.6	24
Rice	-4	16	43	11	1.02	7	17	9	5.67	7	66.4	9
Hardwood	-22	16	66.7	9	0.24	1	60	8	-	-	-	-
Softwood	12	39	76	29	0.17	29	147	9	0.42	29	15.4	3
O.P.	-2	115	68	72	0.29	50	62	49	0.36	28	23.2	23
Manure	-26	5	19	5	3.06	5	-	-	0.03	5	-	-
Sludge	-4	28	47	12	0.89	12	250	5	0.34	6	-	-
Pyrolysis temperature (°C)												
< 350	6	31	60	17	0.33	17	403	9	0.40	16	4.5	6
350-450	-8	46	56	24	1.19	16	221	15	0.15	6	19.8	12
450-550	5	113	69	71	0.35	59	164	61	1.17	46	22.7	30
> 550	-5	48	67	17	0.69	11	27	18	0.21	6	34.6	9
Pyrolysis heating rate (°C min^{-1})												
5-10	6	146	70	85	0.28	79	217	75	0.37	58	13.2	32
> 10	2	29	51	6	0.94	6	118	4	9.70	4	44.2	6
Pyrolysis residence time (min)												
< 60	1	122	64	94	0.55	74	84	47	0.95	62	34.1	26
60-180	1	69	65	22	0.37	22	288	45	0.40	5	4.6	24
> 180	-16	6	-	-	-	-	142	2	-	-	-	-

Note: The Si concentration represents the Si content to the total weight of biochar, with the Si content typically determined through inductively coupled plasma spectroscopy. The SiO_2 concentration represents the SiO_2 content to the total weight of oxides, with the SiO_2 content typically determined through X-ray fluorescence. O.P.: other plant materials. n: numbers of records. The term "Effect index" is defined by Eq. (2.5).

Table 2.2 Effect indexes and critical properties of biochar used for 28-day compressive strength measurements, including means and numbers of records.

Biochar variables	Effect index (%)		C content (%)		Molar O/C ratio		Specific surface area (m^2 g^{-1})		Si concentration (%)		SiO_2 concentration (%)	
	Mean	**n**	**Mean**	**n**	**Mean**	**n**	**Mean**	**n**	**Mean**	**n**	**Mean**	**n**
Feedstock												
Corn	9	35	-	-	-	-	483	24	-	-	4.6	24
Rice	-6	19	47	14	0.81	10	19	7	0.25	5	49.3	14
Wheat	13	17	67	11	0.34	5	103	12	-	-	10.7	5
Hardwood	-23	16	66.7	9	0.24	1	60	8	-	-	-	-
Softwood	7	39	76	29	0.17	29	147	9	0.42	29	15.4	3
O.P.	3	178	73	112	0.25	68	82	73	0.40	39	18.5	30
Manure	-8	17	19	5	3.06	5	-	-	0.03	5	-	-
Sludge	-5	31	47	12	0.89	12	250	5	0.34	6	-	-
Pyrolysis temperature (°C)												
< 350	0	43	60	17	0.33	17	403	9	0.40	16	4.5	6
350-450	-13	43	51.7	18	1.73	10	279	11	0.15	6	4.7	6
450-550	4	191	70	124	0.30	91	150	88	0.38	55	19.9	53
> 550	6	59	70	28	0.69	11	61	29	0.21	6	34.6	9
Pyrolysis heating rate (°C min^{-1})												
5-10	5	186	70	124	0.29	102	221	89	0.37	58	14.6	55
> 10	16	56	71	18	1.62	2	54	28	-	-	44.2	6
Pyrolysis residence time (min)												
< 60	1	175	66	132	0.48	96	102	69	0.36	61	25.8	49
60-180	4	89	69	42	0.31	32	240	55	0.40	5	4.6	24
> 180	-1	12	69	6	-	-	172	8	-	-	-	-

Note: The Si concentration represents the Si content to the total weight of biochar, with the Si content typically determined through inductively coupled plasma spectroscopy. The SiO_2 concentration represents the SiO_2 content to the total weight of oxides, with the SiO_2 content typically determined through X-ray fluorescence. O.P.: other plant materials. n: numbers of records. The term "Effect index" is defined by Eq. (2.5).

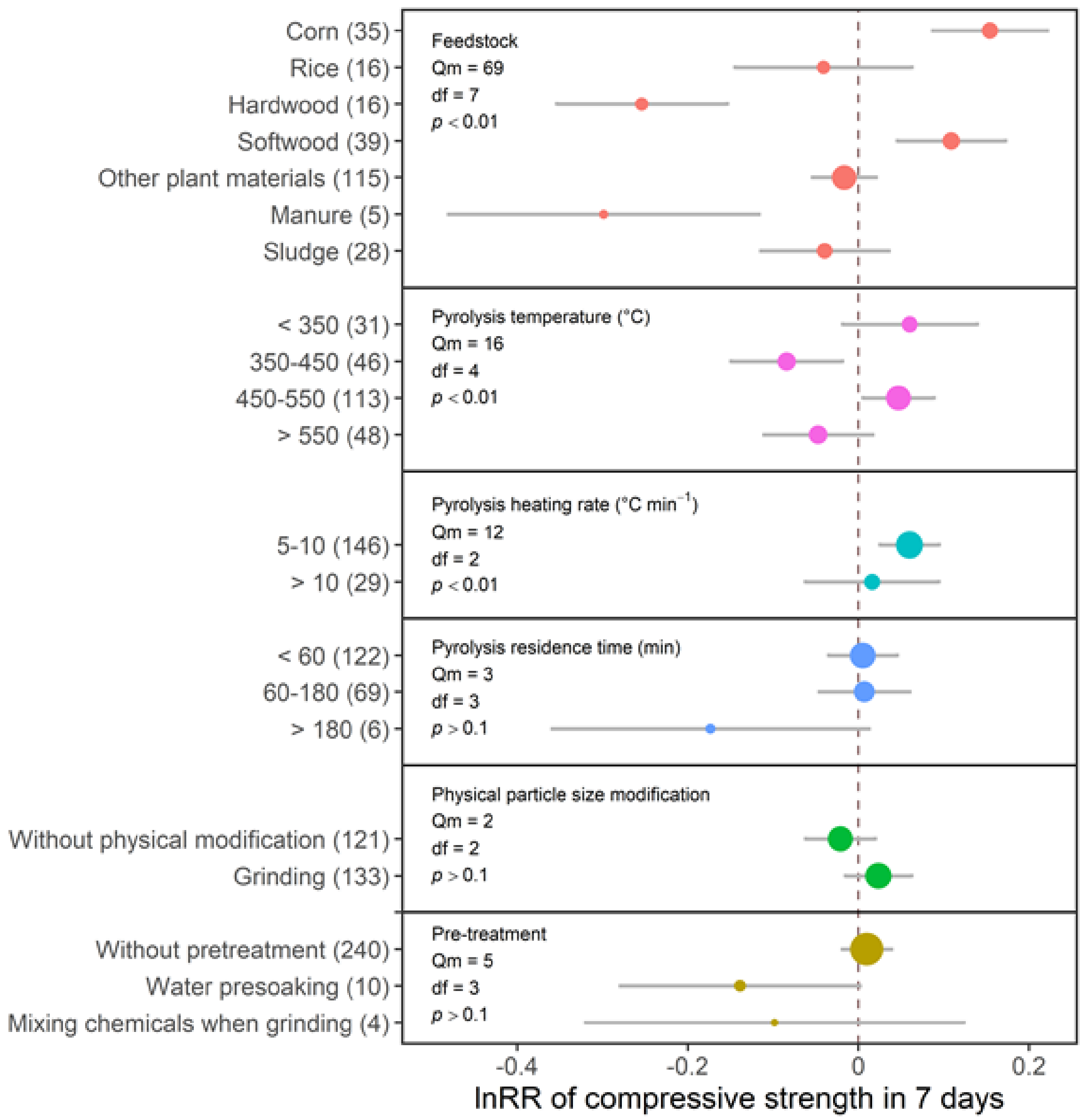

Figure 2.1 The effect sizes of biochar addition on the 7-day compressive strength of Portland cement composites, as affected by the feedstock used for biochar production, pyrolysis temperature, pyrolysis residence time, pyrolysis heating rate, biochar modification and pre-treatment. Each point represents effect sizes, and the size of the point represents the relative number of records compared to the total records. Grey bars represent 95CI. The vertical dash line represents the value of 0. The numbers of records are indicated in the brackets.

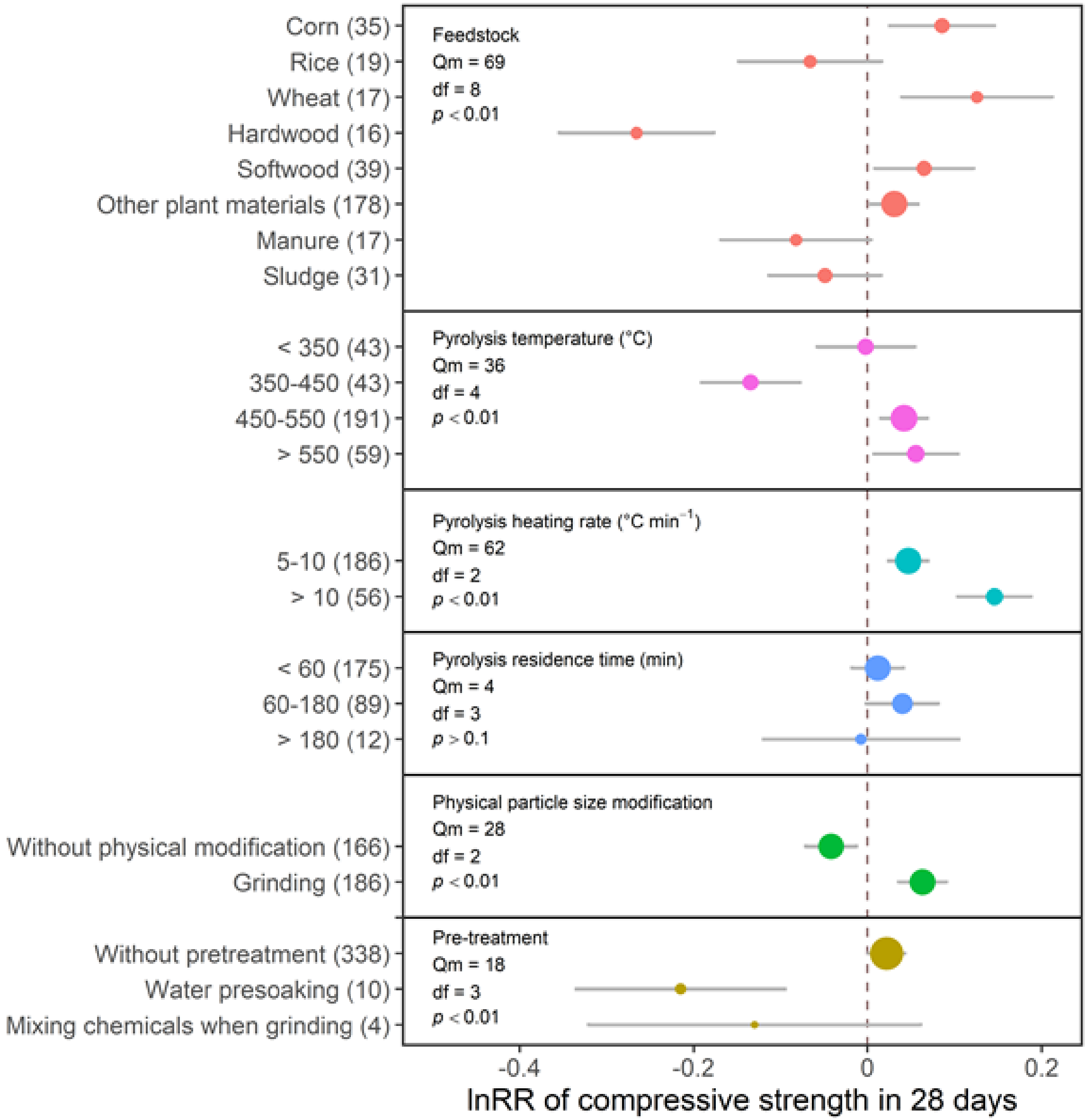

Figure 2.2 The effect sizes of biochar addition on the 28-day compressive strength of Portland cement composites, as affected by the feedstock used for biochar production, pyrolysis temperature, pyrolysis residence time, pyrolysis heating rate, and biochar modification and pre-treatment. Each point represents effect sizes, and the size of the point represents the relative number of records compared to the total records. Grey bars represent 95CI. The vertical dash line represents the value of 0. The numbers of records are indicated in the brackets.

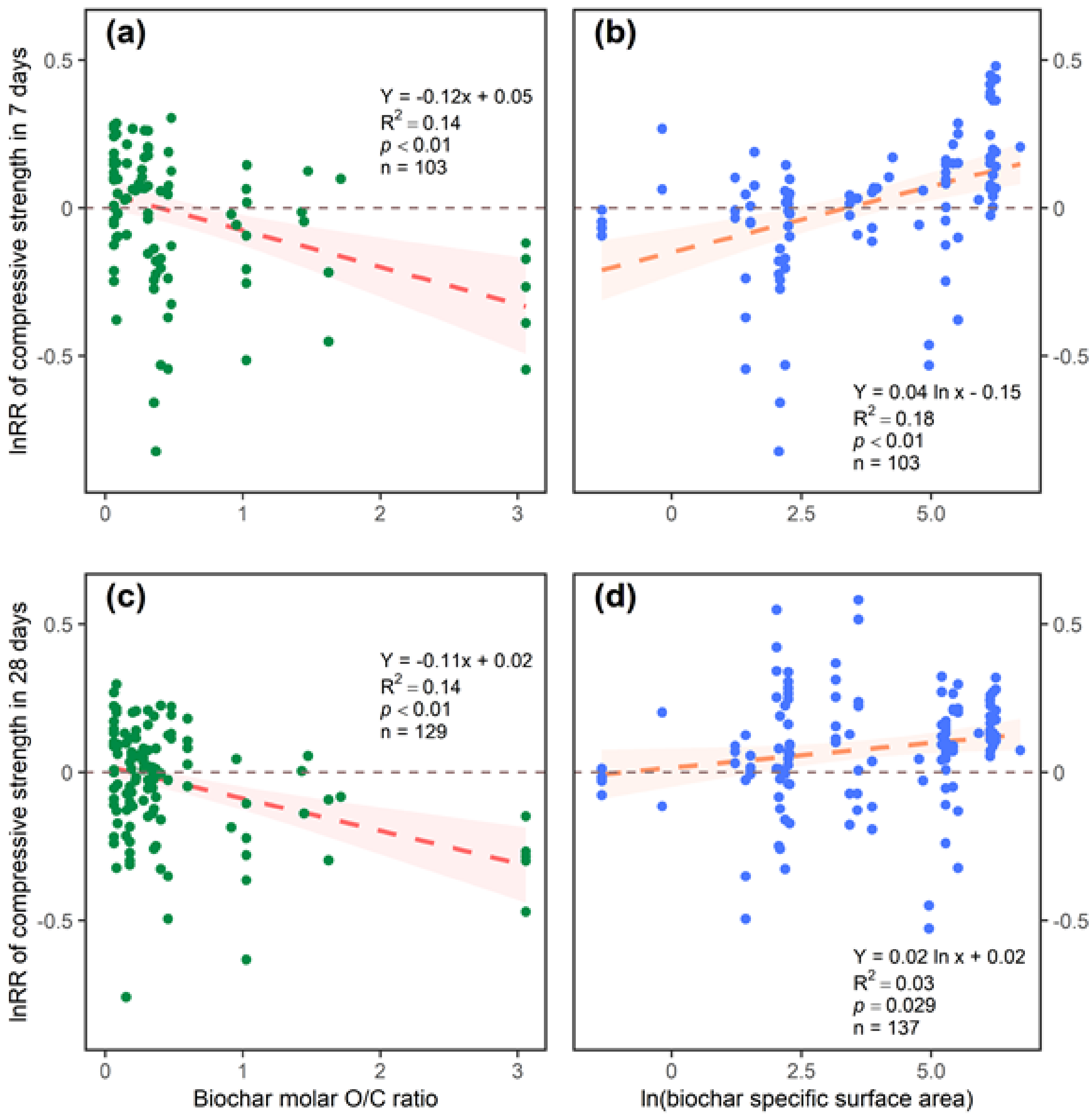

Figure 2.3 Linear correlations between (a) the molar O/C (oxygen/carbon) ratio of biochar and effect size of 7-day compressive strength, (b) the natural log-transformed specific surface area of biochar and effect size of 7-day compressive strength, (c) the molar O/C ratio of biochar and effect size of 28-day compressive strength, and (d) the natural log-transformed specific surface area of biochar and effect size of 28-day compressive strength. Points in each figure represent paired records. The simple linear regression lines with 95% confidential intervals are shown, with the number of records (n) presented. The horizontal dash lines represent the value of 0.

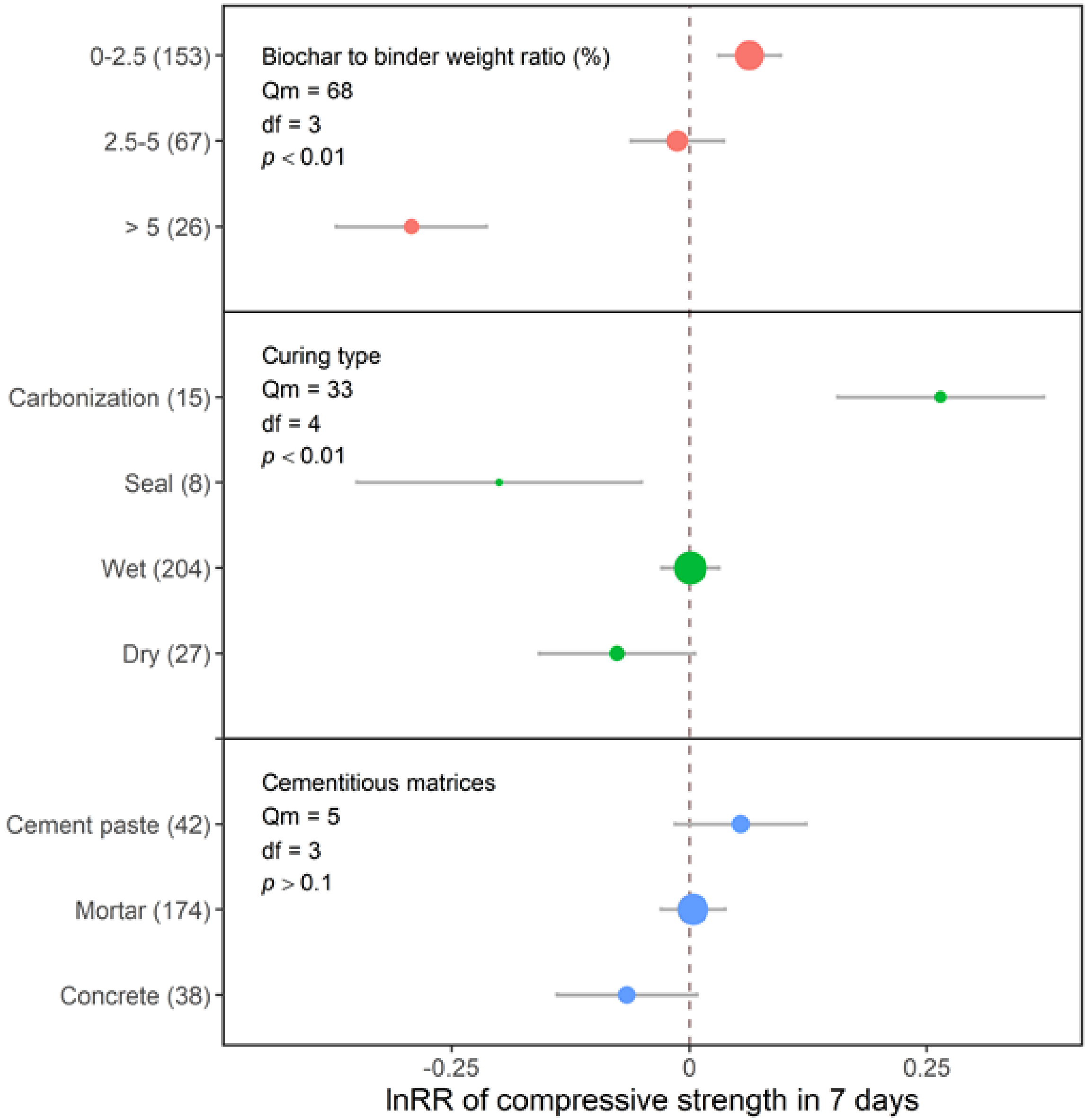

Figure 2.4 The effect sizes of biochar addition on the 7-day compressive strength of Portland cement composites, as affected by the dosage of biochar application, curing method, and cementitious matrix. Each point represents effect sizes, and the size of the point represents the relative number of records compared to the total records. Grey bars represent 95CI. The vertical dash line represents the value of 0. The numbers of records are indicated in the brackets.

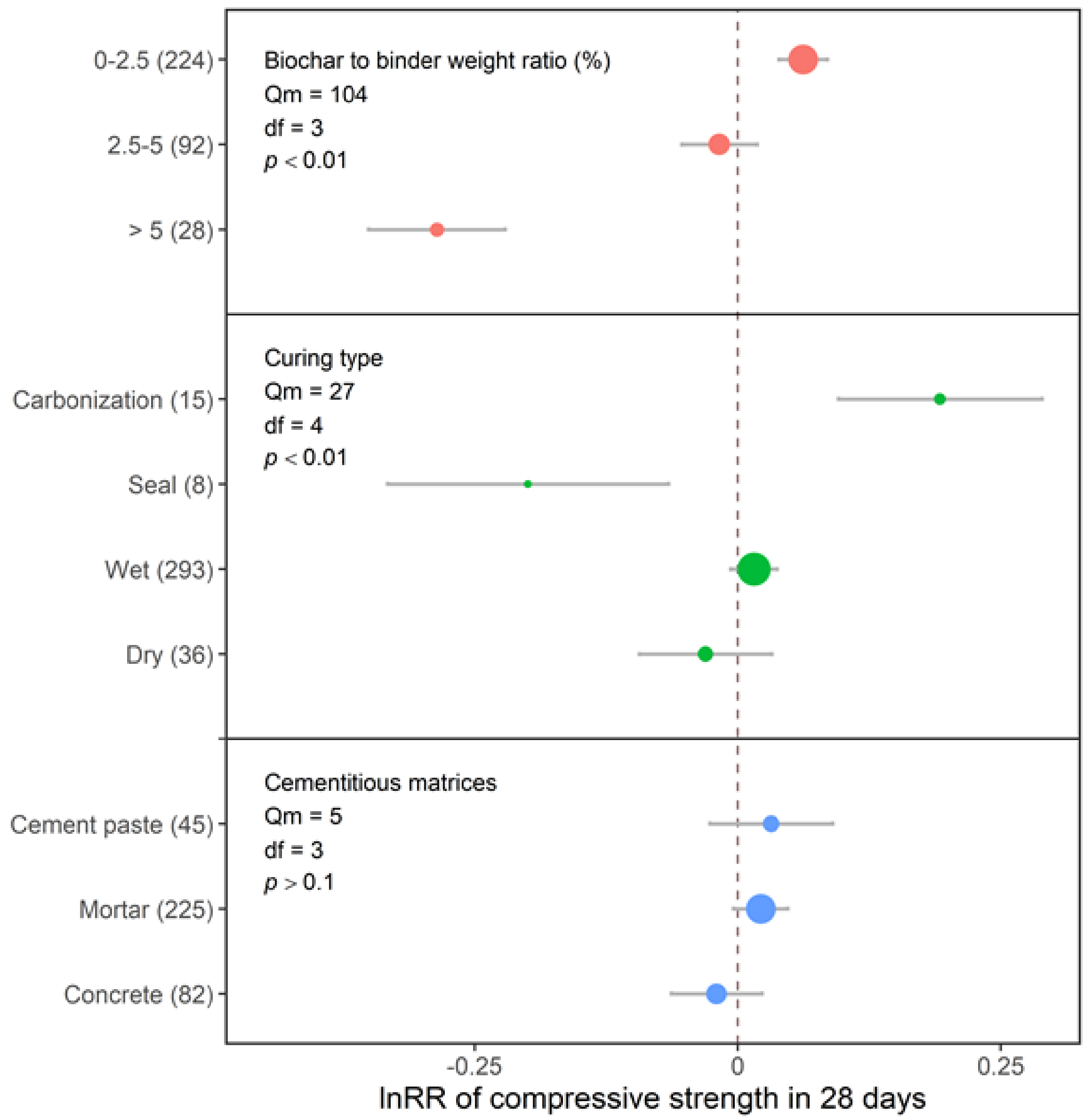

Figure 2.5 The effect sizes of biochar addition on 28-day compressive strength of Portland cement composites, as affected by the dosage of biochar application, curing method, and cementitious matrix. Each point represents effect sizes, and the size of the point represents the relative number of records compared to the total records. Grey bars represent 95CI. The vertical dash line represents the value of 0. The numbers of records are indicated in the brackets.

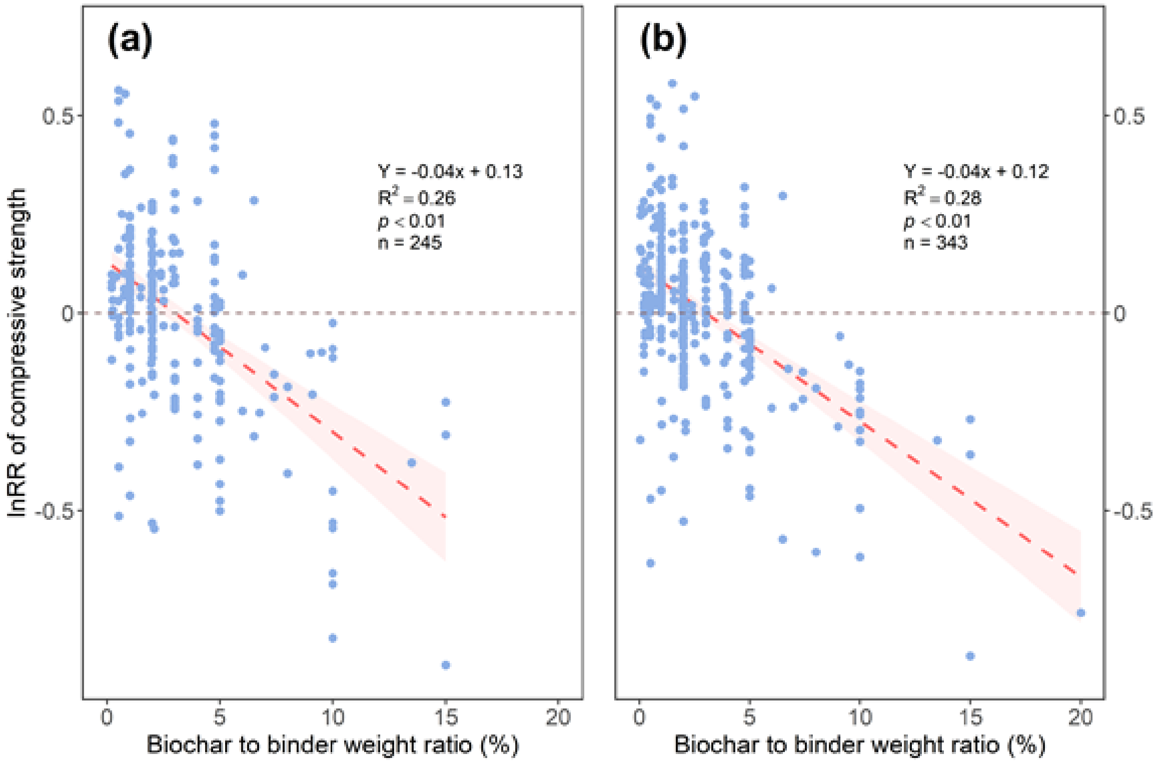

Figure 2.6 Linear correlations between biochar dosage and the compressive strength of Portland cement composites: (a) biochar to binder weight ratio and effect size of 7-day compressive strength, (b) biochar to binder weight ratio and effect size of 28-day compressive strength. Points in each figure represent paired records. The simple linear regression lines with 95% confidential intervals are shown, with the number of records (n) presented. The horizontal dash lines represent the value of 0.

Chapter 3: Biochar Affects the Performance and Quality of Concrete and Cement Paste: Laboratory Experiments

3.1 Introduction

Construction is one of the most important industries worldwide, and concrete is one of the critical construction materials (Monteiro et al., 2017). Portland cement is a vital material in concrete and acts as a binder. With the increased economic growth worldwide, the demand for Portland cement is expected to increase by 12-23% above 2020 levels by 2050 (Cheng et al., 2023). However, the production of Portland cement emits a large amount of CO_2 due to the calcination of limestone and the combustion of fossil fuels, composing around 5-8% of global anthropogenic CO_2 emissions (Andrew, 2019; Friedlingstein et al., 2022; Miller et al., 2017). Therefore, developing a strategy to balance the increased Portland cement production and carbon neutrality requirements is vital (Wei et al., 2022). One strategy to improve the balance is to replace Portland cement with supplementary cementitious materials (SCMs), including fly ash, silica fume and waste glass (Li et al., 2022; Mehta and Ashish, 2020; Miller et al., 2021). These SCMs have positive effects when partially replacing Portland cement in concrete construction and can maintain the concrete performance requirements (Kosmatka and Wilson, 2011). However, with the demand for the green industry, the number of traditional factories will decrease, causing a decreased availability of traditional SCMs such as fly ash. On the other hand, one study found that the reduction of CO_2 emissions was poorly related to the amount of traditional SCM used in concrete when concrete compressive strength is maintained due to the SCM availability and the variation of SCM transportation's distance and mode (Miller, 2018), indicating that traditional SCMs may not meet the carbon neutrality requirements. Therefore, novel SCMs need to be developed to maintain concrete performances and decrease CO_2 emissions simultaneously.

Biochar, a recalcitrant pyrolytic material derived from biomass and other organic matter, is an efficient environment ameliorant widely applied in environmental restoration and soil amendment. Biochar production and applications have been reported to contribute to negative carbon emissions (Chen et al., 2019; Mishra et al., 2023; Werner et al., 2018). In addition, past research has concluded that biochar addition increased Portland cement composite's mechanical performances (Gupta and Mahmood, 2022; Qin et al., 2021; Zhang et al., 2022a) and durability (Gupta et al., 2021b; Sikora et al., 2022; Yang and Wang, 2021a), while decreasing Portland cement composites' water permeability (Gupta et al., 2020b, 2018a) and shrinkage (Dixit et al., 2021; Mobili et al., 2022). These progressive results indicated that biochar could be used in concrete construction to mitigate carbon emissions and maintain or improve concrete performance. Although a high dosage of biochar (> 5% of binder by weight) acting as SCMs is expected to decrease the compression strength of Portland cement composites due to biochar agglomeration (Maljaee et al., 2021b), this shortcoming could be mitigated by cooperating with other SCMs or mixtures, such as fly ash and silica fume, which has also been reported to decrease greenhouse gas emissions, depending on life cycle assessment (Chen et al., 2022b, 2022a; Gupta and Kua, 2020; Praneeth et al., 2020). These results indicate that biochar addition has a high potential to be used as an SCM for carbon sequestration.

Although biochar addition achieved impressive improvements in the performance of Portland cement composites, the use of biochar as an SCM in Portland cement composites for building construction has a short history, as the first research paper was published no more than 10 years ago (Restuccia and Ferro, 2016). There are still many research gaps in this field. One of the knowledge gaps is that only some biochar types have been tested for their suitability as an SCM. Sawdust was the most popular biochar feedstock in the study, composing around 50% of the total peer-reviewed studies in my meta-analysis, followed by corn, rice, and other agricultural wastes (Zhao et al., 2024). However, as the sixth cereal production worldwide, oat hulls can be a significant source of feedstock for biochar production whose biochar has different properties from other cereal straw-based biochars (Kaur et al., 2019), but no researcher has used this biochar for such a study. In addition, the effects of biochar addition on Portland cement composite's compressive strength have heterogeneous results based on my meta-analysis (Zhao et al., 2024), indicating a complex correlation between biochar properties and concrete performances. Although several review papers conclude the mechanism of the effects of biochar addition (Maljaee et al., 2021a; Senadheera et al., 2023; Zhang et al., 2022), it is hard to precisely predict how biochar addition will affect the performances of Portland cement composites due to lack of experimental data. Moreover, testing the performances of Portland cement composites in different forms should have been an optimal way to consider the effects caused by aggregate to detail how biochar works, which can also provide clues of correlation among different performances among forms of Portland cement composites, but only a few researchers had done it (Sirico et al., 2020; Suarez-Riera et al., 2020). Testing only one form of Portland cement composites will mix primary and secondary factors of biochar addition effects, making it difficult to draw conclusions. Therefore, it is necessary to expand the research scope to improve the understanding of the effects of biochar addition.

In this study, I used oat hull and sawdust biochars to test the effect of biochar addition on concrete's compressive strength and water sorptivity, properties representing concrete's mechanical performance for construction resistance to fracture and durability for construction resistance to chemical erosion, respectively. To exclude effects caused by aggregates, I also tested cement paste’s fresh properties and compressive strength using the same batching design as in the concrete experiments to see how these biochars affect the binder quality. Finally, I checked the correlation between concrete and cement paste performances and the relationships between their effect sizes to check if biochar addition has consistent effects among the performances of concrete and cement paste. I hypothesize that: (1) amendments of oat hull and sawdust biochars have significantly different effects on the alteration of concrete performance and cement paste quality; (2) the alteration of cement paste quality cannot explain that of concrete performance due to the nonsignificant correlations among their performance parameters. This study aims to provide information on whether the effects of oat biochar addition on concrete differ from that of sawdust biochar and whether biochar-amended concrete resistances to fracture and chemical erosion differ from the normal concrete, as well as the information on whether the alteration of concrete performance can be explained by the alteration of cement paste quality with the biochar amendment, which can provide clues of modelling the biochar amendment mechanisms and applying biochar to construction in the fieldwork.

3.2 Methods

3.2.1 Materials and batching design

Portland cement Type GU produced by Lafarge Canada Inc., compliant with CSA A3001, was used for this study. Sand (5 mm) and gravel (limestone, 20-14 mm), supplied by SiteOne Landscape Supply Ltd., were used as fine and coarse aggregates. Tap water was used for mixing. Compliant with ASTM C128, sand's moisture content and water absorption were 0.20% and 1.54%, respectively. Compliant with ASTM C127, gravel's moisture content and water absorption were 0.09% and 1.13%, respectively.

Raw biochars produced from two feedstocks were used (ARTi, Des Moines, USA) in this study: oat hull (OH) and sawdust (SD) biochars, produced under a pyrolysis temperature of 550 ℃. The pyrolysis residence time of oat hull biochar was 20 minutes, while that of sawdust biochar was 25 minutes. Each type of biochar was ground using two methods: one was produced by grinding the raw biochar using a blender in the laboratory at the University of Alberta and labelled as 1OH and 1SD, and the other was processed through an automatic mill at ARTi and labelled as 2OH and 2SD. Fourier-transform infrared spectroscopy (FTIR) for detecting chemical bonds of biochars, X-ray diffraction analysis (XRD) for detecting crystal substances of biochars, inductively coupled plasma-optical emission spectrometry (ICP-OES) for measuring biochar mental contents, scanning electron microscope (SEM) for creating microscopic surface images and measurements for biochar macroelement contents (carbon, nitrogen, sulphur) were conducted, which provide information of biochar physical features and chemical properties. SEM was also used to compare the relative particle size between biochars and cement particles.

The batching designs for the concrete and cement paste studies are shown in Table 1. The weight ratio of cement, water, sand, and gravel was 1: 0.45: 1.39: 2.12, and this design is for normal strength concrete with the expected 28-day compressive strength of around 30 MPa to represent the most widely utilized concrete type worldwide. Biochar was used to replace 2% and 4% of the cement by weight. Cement paste was mixed to test fresh and hardened performances to check how biochar addition altered its quality. Subtracting the absorbed water by aggregates based on the trail batching and aggregate water absorptions, the proportion of cement and water was 1: 0.41. An FX-700 compression testing machine (Forney, Zelienople, USA) was used to test compressive strength.

An RLX-6 Type B mechanical mixer (Red Lion, Bolton, Canada) was used for concrete mixing, and a mixing machine (Hobart, Toronto, Canada) was used for cement paste mixing.

3.2.2 Mixing and testing

Concrete mixing was carried out in the RLX-6 Type B mechanical mixer at ambient temperature (around 23 °C), compliant with ASTM C192/C192M. Concrete materials (sand, biochar, cement, and gravel) were dry mixed first for 20 seconds, followed by the addition of water, and then they were mixed for 1 minute until all materials were mixed well. The fresh concrete was then cast into 100×200 mm (diameter×height) cylinder moulds. The cast samples were covered with plastic sheets for the next 24 h till demolding. After demolding, all samples were transferred to a fog room (with 95% relative humidity) for curing at ambient temperature. The samples were cured for 7 or 28 days.

The slump and air content of the fresh concrete were measured before casting, and they were compliant with ASTM C143/C143M and ASTM C231/C231M, respectively. These two performance parameters provide information on concrete workability and freeze-thaw resistance. For the slump measurement, a slump cone was used, with the base diameter, top diameter, and height set at 200 mm, 100 mm, and 300 mm. During the slump experiment, the mould was filled with fresh concrete, and then the mould was removed over 5 s. The differences in vertical height between the concrete after the mould removal and mould were measured, with the results rounded to the nearest 5 mm. For the fresh air content measurement, a pressure method was used through a Type-B meter (Humboldt Mfg. Co., Elgin, USA), and the testing procedure strictly followed the standard. Each batching treatment was not replicated for individual mixing.

Compressive strengths at 7 and 28 days were measured and calculated, and they were compliant with ASTM C39/C39M. Each treatment had 3 replicates. The two performance parameters provide information on the concrete early strength growth rate and the resistance to fracture. Samples were ground flat at both ends at 6 and 27 days for the 7- and 28-day treatments, respectively, to ensure flat ends of cylinders. After being removed from the fog room, each sample was measured within 30 min. For each sample, the maximum pressing force (kN) was recorded as P, and the average diameter (mm) of the ends of the sample was recorded as D. The compressive strength (MPa) was calculated based on Eq. (3.1) in ASTM C39/C39M:

$$f_c' \ (MPa) = \frac{4000P}{\pi D^2} \tag{3.6}$$

Water sorptivity tests and calculations were conducted, representing the water penetration rate of concrete, including initial and secondary sorptivity, compliant with ASTM C1585. These two performance parameters provide information on the status of concrete pore structure, which is explained in Section 3.3.2.2. All samples were cut from the 100×200 mm (diameter×height) 28-day cylinders with 50 mm height, and each treatment had 3 replicates. Samples were sealed in closed containers in 80% relative humidity and placed in a 50 °C oven for 3 days to adjust the effects of the initial water content of samples on initial sorptivity (Hall, 1989). After water content correction, samples were sealed in Ziploc bags (SC Johnson, Racine, USA) for at least 15 days. Each sample's side surface and the upper end were sealed with duct tape and plastic wraps before the experiments to control the water absorption route. Samples were put on supports in a pan, and the water level was 1 to 3 mm above the top of the supports, aiming to exclude the effect of hydraulic pressure. The time 0 was set as the time when samples were put into the pan, and the weights of samples were measured at time 0, 1 min, 5 min, 10 min, 20 min, 30 min, 1 h, 2 h, 3 h, 4 h, 5 h, 6 h, 1 day, 2 day, 3 day, 5 day, 6 day, 7 day and 8 day. The equivalent water absorption heights (EWHs) (mm) were calculated based on Eq. (3.2):

$$EWH\ (mm) = \frac{(W_t - W_0)}{A \times \rho_0} \times 10 \quad (3.2)$$

W_t is the weight of samples at time t, W_0 is the weight of samples at time 0; A is the area of sample ends; ρ_0 is the water density: 0.9975 g cm^{-3}. Two regression lines between EWH and time were made: time from 1 minute to 6 h, and the other one for time from 1 day to 8 days. The 1 min to 6 h regression slope represents the initial sorptivity, and the 1-day to 8-day regression slope represents secondary sorptivity.

Hardened density and water absorption were measured and calculated using the immersed weighting method compliant with ASTM C642. These two performance parameters supplement compressive strength and water sorptivity results and represent the status of concrete's macroscopical structure in this study. The 28-day concrete samples were immersed in water for at least 48 h until the weight of the cylinders did not change, and the weight (kg) was recorded as A. Then, samples were immersed in boiling water for 5 h, and the weight (kg) was recorded as B. After boiling water immersion, samples were immersed and weighed in water, and the weight (kg) was recorded as C. Finally, samples were dried in an oven under 105 °C. The weight (kg) was recorded as D. Each treatment had 3 replicates. Water density ρ is 997.5 kg m^{-3} under ambient temperature (around 23 °C). Therefore, hardened density and water absorption were calculated based on Eq. (3.3) and (3.4) in ASTM C642:

$$Hardened\ density\ (kg\ m^{-3}) = \frac{D}{B - C} \times \rho \quad (3.3)$$

$$Water\ absorption\ (\%) = \frac{A - D}{D} \times 100 \quad (3.4)$$

The cement paste was mixed in an epicyclic mechanical mixer at ambient temperature, compliant with ASTM C305. Water was first added into the mixing bowl, followed by the addition of biochar and cement, and the mixtures were left to sit for 30 s. Then, the paddle was put in the mixing machine. After the machine was turned on, the mixing machine was set to the low-speed mode (~140 r min^{-1}), and the mixtures were mixed for 30 s, and then the mixer was stopped for 15 s. Then, the machine was set to medium speed mode (~285 r min^{-1}), mixed for 1 minute, and finished the mixing. The fresh paste was then cast into 50×50×50 mm cubic moulds. The cast samples were covered with polythene sheets for the next 24 h till demolding. After demolding, all the samples were transferred to a fog room for curing at ambient temperature. The samples were cured till the pre-determined age for testing.

The compressive strengths of cement paste at 7 and 28 days, as well as critical indicators of the quality of cement pastes, were measured and calculated and were compliant with ASTM C109. The two performance parameters provide information on the bond strength of the cement paste. Each type of biochar-contained concrete has 3 replicates to ensure the reliability of the results. For each sample, the maximum pressing force (kN) was recorded as P, and the average length and width (mm) of ends of the sample were recorded as L and W, and the compressive strength (MPa) was calculated based on Eq. (3.5):

$$f_c{}' \ (MPa) = \frac{1000P}{L \times W} \tag{3.5}$$

The flow of fresh cement paste (or paste flow) was measured and calculated, representing the flowability of cement paste, compliant with ASTM C1437. This performance parameter indirectly reflects the cement paste’s viscosity and yield stress. Each type of biochar-contained paste and the control group had 2 replicates. A flow table with a flow mould was used. When testing the paste flow, one layer of cement paste was first placed into the mould, the cement paste was tamped 20 times with a tamper, and then the mould was filled with the cement paste and tamped. After the mould was fulfilled, the cement paste was cut off to a flat top surface. Then, the mixture was left for 1 minute after the mixing operation, and the mould was lifted away from the cement paste. The original base diameter of the cement paste was immediately measured and recorded as L_1. Then, the table was dropped 25 times in 15 s. The diameter of the base of the cement paste after dropping was recorded as L_2. Paste flow is calculated through Eq. (3.6) in ASTM C1437:

$$Flow\ (\%) = \frac{L_2 - L_1}{L_1} \times 100 \quad (3.6)$$

The cement paste's initial and final setting times were adjusted, measured and calculated based on ASTM C191. Each type of biochar-contained concrete has 2 replicates. These two performance parameters represent when the cement paste loses plasticity and when it is to be hardened, indirectly reflecting its viscosity and yield stress. A manual 1-mm Vicat needle apparatus (Humboldt Mfg. Co., Elgin, USA) was used. The paste was placed into the conical mould with tamping, and the mould was placed in a fog room for 30 min. When testing the setting time, the Vicat needle was put on the paste's surface and dropped by gravity. The penetration of the Vicat needle at this time was determined every 10 min after that until a penetration depth of 25 mm or less was obtained. The time was recorded as H and E (min) before and after the 25-mm penetration, with the penetration depth as P_1 and P_2. The Eq. (3.7) from ASTM C191 was used to calculate the initial setting time. After this procedure, the time when there was no significant penetration on the surface of the paste was recorded as the final setting time.

$$IST\ (min) = \left(\frac{H - E}{P_1 - P_2}\right) \times (P_1 - 25) + E \quad (3.7)$$

This study used Eq. (3.8) to calculate effect sizes to illustrate the alteration of concrete and cement paste performances compared to the control:

$$lnRR = ln\,(\frac{\bar{X}_t}{\bar{X}_c}) \tag{3.8}$$

$\bar{X}_t$ is the mean of the treatment group, which is the group that biochar is added into cementitious matrices; $\bar{X}_c$ is for the control group without biochar addition.

3.2.3 Data analysis

All statistical analyses were performed in RStudio (version 4.3.2). The normality of data distribution was performed using the Shapiro-Wilk test. Levene's test was used to detect data homogeneity of variance under the normal distribution. Data will be transformed by Box-Cox transformation if data does not meet the normality of distribution and homogeneity, and transformed data will be mentioned in Section 3.3. One-way analysis of variance (ANOVA) was used to determine significant differences among treatments, and the Tukey test was used to process the post hoc analysis. This study also used the simple linear model (SLM) represented by Eq. (3.9) to describe their relationships. Data used for SLM was also tested for the normality of distribution and homogeneity of variance, with the same solution as data for ANOVA. Eq. (3.9) could be used to generate the Pearson correlation coefficients:

$$Y = \beta_0 + \beta_1 x + \varepsilon \tag{3.9}$$

β_1 is the coefficient; x is the treatment factor; ε is the sampling error. Pearson coefficient is calculated based on the results of SLM. Statistical significance was set as $p < 0.05$ ($\alpha = 0.05$) since all experiments were strictly controlled.

3.3 Results and discussion

3.3.1 Biochar characterization

Biochar with blender grinding had similar chemical properties between the two feedstocks, except for total sulphur content (Table 3.1). In addition, biochar addition could reduce the content of metals in the binder, including calcium, potassium, and aluminum, and those metals are mainly replaced by carbon, which would act as the filler. The XRD spectrum of all four biochars (Figure 3.1) showed 2θ peaks at 26.71° and 29.40°, which represent quartz and calcite, and the tiny 2θ peaks at 23.43° represent amorphous silica. In addition, 2θ peaks at 27.11°, 29.90° and 38.67° for the 2OH biochar represent calcium oxide. The FTIR spectrum (Figure 3.2) indicated that these biochars contain alkane C-H, $-COO^{-}$, C-O, and O=S=O. The presence of these oxygen-containing functional groups suggests the hydrophilicity of all biochars, and the presence of O=S=O indicates that these biochars contain exchangeable sulphate. Rich metals have complex effects on the hydration of Portland cement. For instance, aluminum could accelerate early cement hydration, but iron would retard it, while magnesium could replace calcium in calcium silicate hydrate (C-S-H) and would not significantly affect paste quality (Stephan et al., 2008). The presence of calcite with low content could also prevent the formation of monosulfate, which did not affect cement paste strength (Taylor, 1997). Potassium could attract water into the C-S-H intralayer to increase the stiffness of C-S-H, increasing compressive strength, and sodium could react as a catalyst as soluble sodium would form sodium silica to generate C-S-H (Gupta et al., 2021a; Jawed and Skalny, 1978). However, since the content of calcite and metals was low in biochar, their effects might not be significant under low biochar dosages. Detectable quartz and amorphous silica indicate a complex silica phase in the biochar (Nguyen, 2021), which might work differently from silica fume or other silica materials. On the other hand, biochar after blender grinding (1OH and 1SD) had a smaller particle size than mill-ground biochar (2OH and 2SD). Blender-ground biochar has particle sizes similar to cement particles based on SEM images (Figure 3.3), and it is spherical compared to biochar with automatic mill grinding, indicating that biochars after blender-grinding would perform better as fillers than mill-ground biochars.

3.3.2 Performances of biochar-amended Portland cement composites

3.3.2.1 Fresh concrete properties

Biochar addition generally reduced slump and increased the air content of fresh concrete (Table 3.1), but it did not affect the paste flow and initial setting time of cement paste (Table 3.5). In addition, treatment 1OH-4 significantly decreased the final setting time. Air content generally increased with biochar addition, except for treatments 1OH-2, 1OH-4, and 1SD-2. Since plant-based biochars are generally hydrophilic and can absorb water during mixing, the addition of such biochars will reduce free water, decrease the workability and increase the cohesiveness of paste due to the densification of the cementitious matrix (Gupta, 2021; Gupta and Kashani, 2021), but the spherical shape of biochar would offset the effect of decreasing workability (Neville and Brooks, 2010). In addition, porous structures would increase the porosity of the cementitious matrix, causing an increase in fresh air content, especially for unsaturated materials (Gupta et al., 2018b, 2018a), while the decrease in free water would reduce fresh air content (Mindess, 2019). However, grinding breaks the pore structure of biochar (Figure 3.3), which can diminish biochar's water absorption capacity and decrease biochar's adverse effect on the paste flow. Biochar could act as nucleation sites for cement hydration after mixing with cement, accelerating the cement hydration and paste hardening (Javed et al., 2022). On the other hand, sulphate might be the main sulphur species in biochars in this study and acts as secondary gypsum (Cheah et al., 2014; Leng et al., 2022; Taylor, 1997), possibly with higher sulphate content in biochars produced from agricultural waste than from wood waste (Table 3.2). The addition of oat hull biochar might increase the local concentration of sulphate in the cementitious matrix and cause false set in these areas (Chung et al., 2017), while the high water/binder (w/b) ratio used in this study might lengthen the setting time and dilute the sulphate concentration (Bentz et al., 2009), and these two effects offset each other and made the biochar addition effects nonsignificant in the initial setting.

Overall, the fresh properties of concrete and cement paste illustrated that the biochar amendment reduced the concrete slump and slightly increased the fresh air content due to introducing biochar pores, indicating poor workability but slightly improved freeze-thaw resistance compared to normal concrete. In addition, the flow and initial setting time of cement paste were not affected, which might be related to the dilution of sulphate under the high w/b ratio. It is necessary to consider adding other admixtures to maintain the desired slump and ensure proper mixing and placing processes in practice.

3.3.2.2 Hardened properties

Biochar addition did not significantly affect 7-day compressive strength; although biochar addition did not significantly affect the overall 28-day compressive strength, 1OH-4 biochar significantly increased 28-day compressive strength by 26% (Figure 3.4, Table 3.3), and all treatments, except the treatment 2OH-2, had higher 28-day compressive strength than the targeted value (30 MPa). However, the biochar treatments did not affect hardened densities (Table 3.4). Blender grinding made biochar smaller and has similar particle sizes to cement particles, where biochar could act as nucleation sites for cement hydration and a filler to enhance the bonding strength of the binder (Gupta et al., 2020a; Restuccia and Ferro, 2016), leading to a 14% increase in the 28-day compressive strength compared to the control group (Table 3.3). In addition, amorphous silica in the biochar might be involved in the pozzolanic reaction, increasing the compressive strength (Liu et al., 2022).

On the other hand, cement paste's 7- and 28-day compressive strengths were significantly altered (Figure 3.5). For the 7-day compressive strength, biochar addition did not significantly alter paste strength except for the 1OH-2 treatment. For the 28-day compressive strength, although the overall effects of biochar addition on the paste quality were not significant compared to the control, the higher biochar dosage had a more significant impact on reducing the strength, which was correlated to changes in the hardened density of the paste (Table 3.5). In addition, although not statistically significant, biochar with mill grinding decreased paste strength more than those with blender grinding, supported by SEM images that show cracks around biochar with large particle sizes in the concrete after 28-day curing (Figure 3.6). The results indicated that biochar addition did not alter paste quality, consistent with the results of my meta-analysis (Zhao et al., 2024). Since biochar is a fragile material, which is mainly made up of aromatic compounds (Liu et al., 2015), it would provide weak ITZ between biochar and paste, making the ITZ convenient routes for cracking. Such biochar properties would offset the benefit of biochar being a filler and nucleation site, particularly if the biochar application rate is high and the biochar particle size is large.

All biochar addition treatments have lower initial sorptivity than the CK (Figure 3.6, Table 3.3). Since ground biochar can fill the pores in the cementitious matrix, and the initial sorptivity of treatments 2OH-2, 1SD-4 and 2SD-4 was much lower than that in other treatments; however, the biochar feedstock type and dosage did not significantly affect this performance. Although biochar addition did not significantly alter secondary sorptivity, specific biochar type and modification affected this performance, resulting in a 72% increase for treatment 1OH-2 (Figure 3.6, Table 3.3). The initial sorptivity represents the amount of permeable capillary pores, and its decrease by biochar addition was due to the densification of cementitious matrices by reduced free water and enhanced cement hydration (Gupta et al., 2018c; Khan et al., 2022; Tan et al., 2020;), with small biochar particles acting as fillers to fill the pores to reduce capillary activities (Gupta et al., 2020a). The secondary sorptivity represents the permeable macropores in the concrete, which was primarily correlated to the porosity of aggregates (Gupta et al., 2018c). One research reported that biochar would capture air to form discrete macro-voids in concrete (Gupta and Kua, 2018), cooperating with the macropores of biochar to enhance the pore connectivity in concrete (Muthukrishnan et al., 2019), indicating that biochar addition may increase the volume of macropores and increase secondary sorptivity. However, since the biochar dosage applied in this study was low, biochar addition did not significantly affect total water absorption (Tables 3.4), indicating that biochar addition with low dosage did not significantly alter concrete's whole pore structure.

Overall hardened properties measurements demonstrated that biochar amendment maintained or improved the compressive strength of concrete, but it had more significant effects on the compressive strength of cement paste. Biochar addition also decreased initial sorptivity and increased the secondary sorptivity of concrete. However, the total water absorption and hardened density of concrete were not altered, indicating that biochar addition in this study could alter the microscopic structure but not the macrostructure of concrete. These results implied that biochar-amended could be used to enhance concrete mechanical performance and be designed to resist penetrative chemical attacks. However, such practices require more research to enrich the understanding of the biochar amendment in different mechanical scenarios and various chemical attacks.

3.3.3 Relationships among different performance parameters under biochar addition

The hardened density and water absorption of concrete had significant negative relationships. Although the relationships among performance parameters of cement paste were similar to those of concrete, the paste flow was positively correlated to the final setting time. The hardened density of cement paste was positively correlated to paste flow and final setting time (Table 3.6).

Furthermore, the effect size of the hardened density of concrete was positively correlated with the effect size of water absorption, and the effect size of paste flow was positively correlated to the effect size of the hardened density of cement paste. In addition, the effect size in concrete's secondary sorptivity was positively correlated with the effect size of initial sorptivity (Table 3.7).

Additionally, this study's 28-day concrete compressive strength was negatively correlated with paste flow and initial setting time (Table 3.6). The effect size of concrete's secondary sorptivity was positively correlated with the effect size of the 28-day cement paste compressive strength (Table 3.7).

Since the water absorption of concrete represents the permeable porosity of concrete, such a significant negative relationship between absorption and hardened density and the negative relationship of their effect sizes indicated that biochar addition did not significantly alter the quality of the solid phase of the concrete in this study. Since paste bonding strength could be affected by biochar addition, biochar addition could alter the cracking route in the concrete (Sisman et al., 2024), correlating to parameters relating to the quality of cement paste when under compression, including paste flow and setting time.

In addition, in this study, paste flow, hardened density and setting time of cement paste were correlated to each other, which could be explained by the absorption of water by biochar, where decreasing free water would reduce the paste flow and accelerate the hardening of cement paste (Yu et al., 2023), and the replacement of cement with biochar that has a lower hardened density than cement paste decreased the hardened density of the biochar-cement mixed paste.

On the other hand, although the performance of concrete was mainly affected by the use of aggregates (Maso, 1996; Scrivener et al., 2004; Sims et al., 2019), paste quality would alter the binding quality of the binder, which may also have a significant effect on the performance of concrete. Since water sorptivity was correlated with concrete porosity, which was highly affected by biochar addition, alterations in the capillary pores would align with alterations in the macropores. On the other hand, with the same aggregate compositions, alterations of macropores will be correlated to changes in paste quality, which was shown by the relationship between the effect sizes of secondary sorptivity and the 28-day cement paste compressive strength. However, since most of the effect sizes of initial sorptivity and 28-day cement paste compressive strength are negative, such relationships can be hypothesized as the compensation that the addition of porous biochar could partially increase the local porosity to retard the densification of the cementitious matrix and fill the pores to counteract the effects of weakening ITZ. Such a hypothesis should be explored through more correlation analyses in the future, based on experiments adding one type of biochar into different forms of Portland cement composites with different dosages.

Overall, the results of correlation analyses indicated that the performance parameters of concrete and cement paste and their effect sizes did not strongly correlate with those of each other after biochar amendment, although there were a few significant Pearson coefficients among them. Such results implied that the presence of aggregates plays a critical role in these performances, indicating the importance of considering the specific batching design of concrete during the biochar application.

3.4 Conclusions

Overall, this study found that adding biochar did alter the cement paste's quality and concrete performance parameters, with nonsignificant effects on the biochar feedstock type. I recommend applying biochar in the future with more profound explorations of biochar amendment mechanisms. Biochar addition decreased fresh concrete workability and increased fresh air content for the freeze-thaw resistance, with the maintenance or improvement in the 28-day compressive strength of concrete. Biochar amendment reduced the volume of capillary pores but increased that of macropores. However, the microstructure of concrete was altered, and the macroscopical structure of concrete was not significantly altered due to the low biochar dosage used in this study. In addition, biochar application significantly altered the quality of cement paste, including compressive strength, final setting time and hardened density, indicating that biochar addition can alter the viscosity, yield stress and bonding strength of the binder in concrete. It was apparent that cement paste quality and concrete performance parameters with their effect sizes were partially correlated, indicating that changing binder quality after biochar addition can alter concrete performances. However, since aggregates are the main factors affecting concrete performance, the significance of the effects of binder quality alteration will be partially obscured, and the mechanisms should be explored in the future.

3.5 References

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Table 3.1 Batching design of concrete and cement paste, as well as concrete slumps and fresh air contents.

Treatment	Water (kg)	Cement (kg)	Sand (kg)	Gravel (kg)	Biochar (kg)	Slump (mm)	Fresh air content (%)
Concrete							
CK	6.33	14.07	19.56	29.76	0	75	1.2
1OH-2	6.33	13.79	19.56	29.76	0.28	50	1
1OH-4	6.33	13.51	19.56	29.76	0.56	75	1
2OH-2	6.33	13.79	19.56	29.76	0.28	70	1.5
2OH-4	6.33	13.51	19.56	29.76	0.56	30	2
1SD-2	6.33	13.79	19.56	29.76	0.28	70	1
1SD-4	6.33	13.51	19.56	29.76	0.56	55	1.4
2SD-2	6.33	13.79	19.56	29.76	0.28	45	1.6
2SD-4	6.33	13.51	19.56	29.76	0.56	30	1.8
Cement paste							
CK	0.82	2.00	-	-	0	-	-
1OH-2	0.82	1.96	-	-	0.04	-	-
1OH-4	0.82	1.92	-	-	0.08	-	-
2OH-2	0.82	1.96	-	-	0.04	-	-
2OH-4	0.82	1.92	-	-	0.08	-	-
1SD-2	0.82	1.96	-	-	0.04	-	-
1SD-4	0.82	1.92	-	-	0.08	-	-
2SD-2	0.82	1.96	-	-	0.04	-	-
2SD-4	0.82	1.92	-	-	0.08	-	-

Note: 1OH-2: batching in adding oat hull biochar with blender grinding with the dosage of 2wt% of binder, for concrete and cement paste; 1OH-4: batching in adding oat hull biochar with blender grinding with the dosage of 4wt% of binder, for concrete and cement paste; 1SD-2: batching in adding sawdust biochar with blender grinding with the dosage of 2wt% of binder, for concrete and cement paste; 1SD-4: batching in adding sawdust biochar with blender grinding with the dosage of 4wt% of binder, for concrete and cement paste; 2OH-2: batching in adding oat hull biochar with automatic mill grinding with the dosage of 2wt% of binder, for concrete and cement paste; 2OH-4: batching in adding oat hull biochar with automatic mill grinding with the dosage of 4wt% of binder, for concrete and cement paste; 2SD-2: batching in adding sawdust biochar with automatic mill grinding with the dosage of 2wt% of binder, for concrete and cement paste; 2SD-4: batching in adding sawdust biochar with automatic mill grinding with the dosage of 4wt% of binder, for concrete and cement paste. Detailed meaning of treatment codes and material weight ratios are explained in Section 3.2.1 and Appendix. B. Replication of slump and fresh air content is one.

Table 3.2 Biochar element contents and other chemical properties.

	Biochar type			
Parameter	**1OH**	**2OH**	**1SD**	**2SD**
N (%)	0.55	0.53	0.51	0.19
C (%)	72.70	66.77	72.20	77.38
S (%)	0.22	0.04	0.02	-
Ca (%)	1.09	0.36	1.14	0.45
K (%)	1.73	2.55	1.73	0.26
Mg (%)	0.30	0.27	0.32	0.10
Na (%)	0.05	0.22	0.05	0.04
Al (%)	0.03	0.03	0.03	0.02
Fe (%)	0.17	0.19	0.20	0.11
O (%)	23.16	29.03	23.80	21.44
Molar O/C	0.24	0.33	0.25	0.21
pH	10.39	10.13	10.43	7.67

Note: Oxygen content is calculated by the difference between 100% and the total content of other elements. Abbreviations of biochar types and elements are explained in the footnote of Table 3.1.

Table 3.3 The 28-day compressive strength and the initial and secondary sorptivity of concrete for each treatment. Data are means ± standard deviations (n = 3). Different superscript letters of each column per treatment represent significant significances among the treatments within biochar type, biochar modification methods, and between biochar addition treatments.

Treatment	28-day compressive strength (MPa)	Initial sorptivity (10^{-3} mm $s^{-0.5}$)	Secondary sorptivity (10^{-3} mm $s^{-0.5}$)
Biochar type			
CK	30.65 ± 2.12	6.77 ± 0.03^{a}	1.60 ± 0.20^{a}
1OH	35.41 ± 4.20	4.77 ± 0.42^{b}	2.52 ± 0.37^{b}
2OH	31.74 ± 1.53	3.93 ± 0.75^{b}	1.93 ± 0.28^{a}
1SD	34.72 ± 3.04	4.01 ± 1.05^{b}	2.09 ± 0.18^{a}
2SD	33.67 ± 2.66	3.76 ± 1.04^{b}	1.85 ± 0.40^{a}
Biochar modification			
CK	30.65 ± 2.12^{a}	6.77 ± 0.03^{a}	1.60 ± 0.20^{a}
1 (blender grinding)	35.06 ± 3.51^{b}	4.42 ± 0.83^{b}	2.33 ± 0.36^{b}
2 (automatic mill grinding)	32.70 ± 2.30^{ab}	3.85 ± 0.87^{b}	1.89 ± 0.33^{a}
Biochar addition			
CK	30.65 ± 2.12	6.77 ± 0.03^{a}	1.60 ± 0.20^{a}
Y	33.88 ± 3.15	4.12 ± 0.88^{b}	2.11 ± 0.41^{b}

Note: The meanings of treatment codes are explained in the footnote in Table 3.1. "Y" represents batching with biochar addition.

Table 3.4 The water absorption by and hardened density of concrete for each treatment. Data are means ± standard deviations ($n = 3$). There are no significant differences among treatments.

Treatment	Water absorption (%)	Hardened density (kg m^{-3})
CK	6.28 ± 0.49	2270 ± 31
1OH-2	6.78 ± 0.51	2236 ± 28
1OH-4	6.13 ± 0.35	2269 ± 21
2OH-2	5.92 ± 0.35	2285 ± 21
2OH-4	6.14 ± 0.50	2263 ± 27
1SD-2	6.17 ± 0.53	2274 ± 31
1SD-4	6.54 ± 0.13	2249 ± 11
2SD-2	6.23 ± 0.32	2266 ± 21
2SD-4	6.50 ± 0.33	2237 ± 15

Note: The meanings of treatment codes are explained in the footnote in Table 3.1.

Table 3.5 The 28-day compressive strength, paste flow, setting time, and hardened density of cement paste. Data are means ± standard deviations (n = 3; for Flow: n = 2). Different superscript letters in each column represent significant significances among the treatments per treatment group.

Treatment	28-day compressive strength (MPa)	Flow (%)	Initial setting time (min)	Final setting time (min)	Hardened density ($kg\ m^{-3}$)
Batching					
CK	55.13 ± 4.82[ab]	108 ± 6	238 ± 14	386 ± 5[a]	1611 ± 4[a]
1OH-2	56.83 ± 3.89[a]	98 ± 9	223 ± 20	358 ± 4[a]	1577 ± 13[bc]
1OH-4	48.70 ± 2.41[ab]	94 ± 2	196 ± 7	355 ± 7[b]	1543 ± 2[d]
2OH-2	50.54 ± 1.22[ab]	104 ± 4	253 ± 14	365 ± 14[a]	1587 ± 8[b]
2OH-4	49.19 ± 0.95[ab]	104 ± 9	231 ± 26	372 ± 17[a]	1576 ± 8[bc]
1SD-2	52.85 ± 2.79[ab]	103 ± 2	216 ± 31	382 ± 5[a]	1598 ± 2[ab]
1SD-4	45.39 ± 4.18[b]	101 ± 7	249 ± 4	383 ± 4[a]	1574 ± 2[bc]
2SD-2	52.77 ± 5.09[ab]	105 ± 4	212 ± 2	369 ± 6[a]	1587 ± 1[b]
2SD-4	46.16 ± 4.52[b]	98 ± 2	207 ± 18	369 ± 12[a]	1561 ± 2[c]
Biochar dosage					
CK	55.13 ± 4.82[a]	108 ± 6	238 ± 14	386 ± 5	1611 ± 4[a]
2%	53.47 ± 4.07[a]	103 ± 5	226 ± 23	368 ± 11	1587 ± 10[b]
4%	47.27 ± 3.45[b]	99 ± 6	221 ± 25	370 ± 14	1565 ± 14[c]
Biochar with grinding type					
CK	55.13 ± 4.82	108 ± 6	238 ± 14	386 ± 5[a]	1611 ± 4[a]
1OH	53.35 ± 5.33	96 ± 6	209 ± 20	356 ± 5[b]	1560 ± 21[b]
2OH	49.87 ± 1.23	104 ± 6	242 ± 21	369 ± 13[ab]	1581 ± 10[ab]
1SD	49.12 ± 5.18	102 ± 4	233 ± 26	382 ± 4[a]	1586 ± 14[ab]
2SD	49.47 ± 5.69	101 ± 6	209 ± 11	369 ± 8[ab]	1574 ± 15[ab]
Feedstock type					
CK	55.13 ± 4.82	108 ± 6	238 ± 14	386 ± 5[a]	1611 ± 4[a]
OH	51.74 ± 4.25	100 ± 7	226 ± 26	362 ± 4[b]	1573 ± 18[b]
SD	49.32 ± 5.27	102 ± 4	221 ± 22	376 ± 9[a]	1580 ± 15[ab]
Biochar modification					
CK	55.13 ± 4.82	108 ± 6	238 ± 14	386 ± 5	1611 ± 4[a]
1 (blender grinding)	51.40 ± 5.49	99 ± 6	221 ± 25	369 ± 14	1573 ± 21[b]
2 (automatic mill grinding)	49.64 ± 4.25	103 ± 5	226 ± 23	369 ± 10	1579 ± 12[b]
Biochar addition					
CK	55.13 ± 4.82	108 ± 6	238 ± 14	386 ± 5[a]	1611 ± 4[a]
Y	50.48 ± 4.87	101 ± 6	223 ± 23	369 ± 12[b]	1576 ± 16[b]

Note: The meanings of treatment codes are explained in the footnote in Table 3.1. "Y" represents batching with biochar addition.

Table 3.6 Pearson correlation between performance parameters of concrete and cement paste.

Parameter	FC7	FC28	SS	IS	HD	AbP	cFC7	cFC28	Flow	IST	FST
FC28	0.08										
SS	-0.31	0.21									
IS	-0.37	-0.16	0.18								
HD	-0.43	0.08	-0.20	0.30							
AbP	0.42	-0.15	0.30	-0.09	-0.93**						
cFC7	0.45	0.22	0.31	0.14	-0.13	0.19					
cFC28	-0.23	-0.33	0.44	0.73*	0.14	0.11	0.38				
Flow	-0.35	-0.70*	-0.37	0.43	0.51	-0.38	-0.10	0.36			
IST	0.18	-0.69*	-0.28	-0.12	0.21	-0.09	-0.21	-0.01	0.56		
FST	0.10	-0.35	-0.47	0.28	0.17	-0.04	0.06	-0.05	0.67*	0.46	
cHD	-0.09	-0.61	-0.21	0.53	0.38	-0.14	0.24	0.59	0.89**	0.52	0.69*

Note: "FC7" represents concrete 7-day compressive strength; "FC28" represents concrete 28-day compressive strength; "SS" represents concrete secondary sorptivity; "IS" represents concrete initial sorptivity; "HD" represents concrete hardened density; "AbP" represents concrete water absorption; "cFC7" represents cement paste 7-day compressive strength; "cFC28" represents cement paste 7-day compressive strength; "IST" represents cement paste initial setting time; "FST" represents cement paste final setting time; "cHD" represents cement paste hardened density.

"*" represents p values between 0.01 and 0.05, "**" represents p values lower than 0.01, and blank represents p values larger than 0.05.

Table 3.7 Pearson correlation between different effect sizes (lnRR) of performance parameters of concrete and cement paste.

Parameter	FC7	FC28	SS	IS	HD	AbP	cFC7	cFC28	Flow	IST	FST
FC28	0.13										
SS	-0.37	0.03									
IS	-0.60	0.19	0.86**								
HD	-0.44	0.18	-0.05	0.25							
AbP	0.40	-0.16	0.25	-0.11	-0.95**						
cFC7	0.48	0.19	0.24	0.28	-0.11	0.19					
cFC28	-0.29	-0.18	0.74*	0.70	0.09	0.09	0.48				
Flow	-0.44	-0.61	-0.11	0.07	0.48	-0.43	-0.04	0.19			
IST	0.18	-0.69	-0.17	-0.39	0.16	-0.08	-0.17	-0.11	0.54		
FST	0.09	-0.16	-0.28	-0.18	0.07	-0.02	0.13	-0.35	0.56	0.41	
cHD	-0.15	-0.48	0.10	0.17	0.32	-0.16	0.38	0.48	0.85**	0.50	0.56

Note: "FC7" represents concrete 7-day compressive strength; "FC28" represents concrete 28-day compressive strength; "SS" represents concrete secondary sorptivity; "IS" represents concrete initial sorptivity; "HD" represents concrete hardened density; "AbP" represents concrete water absorption; "cFC7" represents cement paste 7-day compressive strength; "cFC28" represents cement paste 7-day compressive strength; "IST" represents cement paste initial setting time; "FST" represents cement paste final setting time; "cHD" represents cement paste hardened density.

"*" represents *p* values between 0.01 and 0.05, "**" represents *p* values lower than 0.01, and blank represents *p* values larger than 0.05.

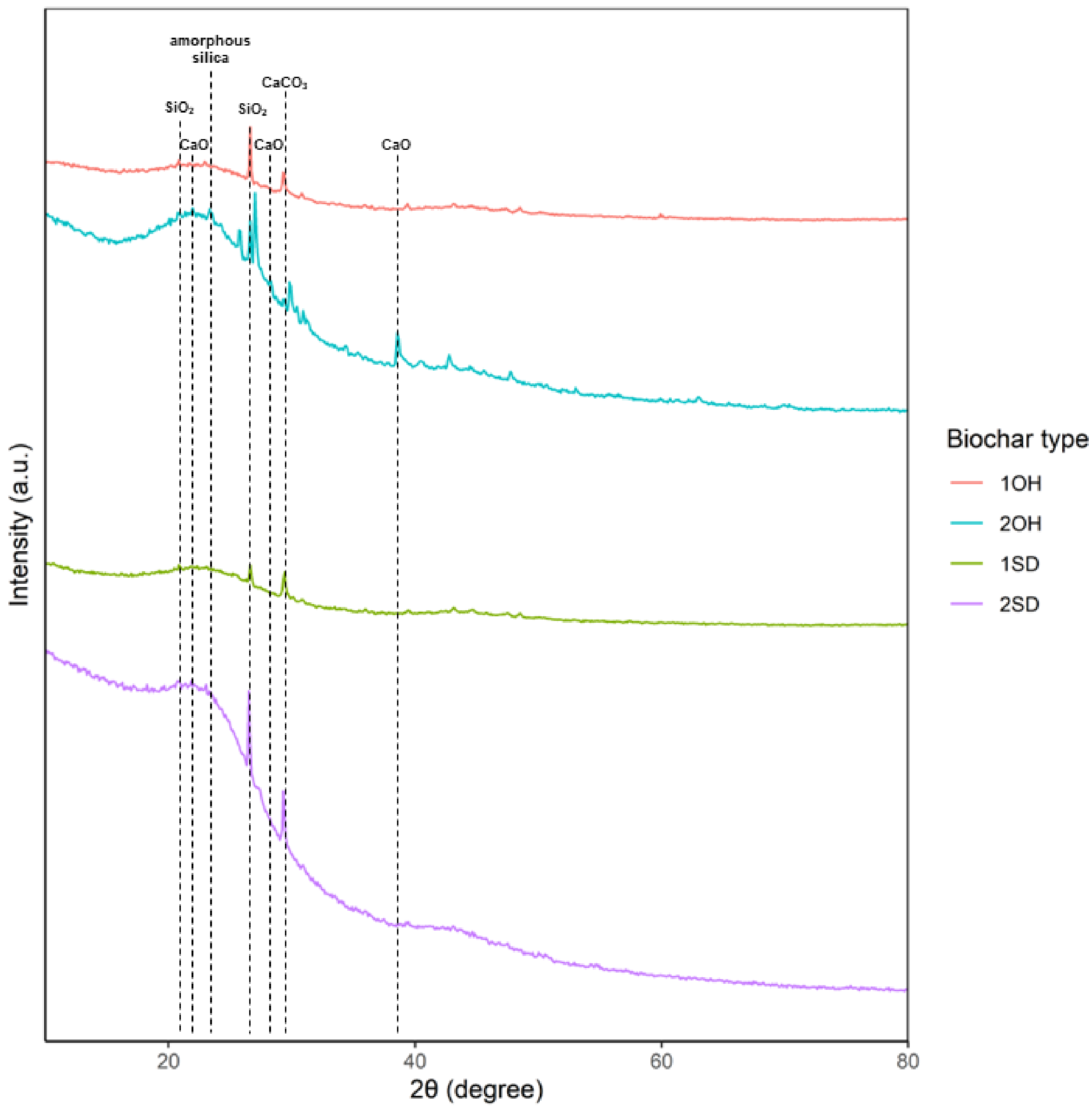

Figure 3.1 XRD spectra of different biochars. Abbreviations of biochar types are explained in the footnote of Table 3.1.

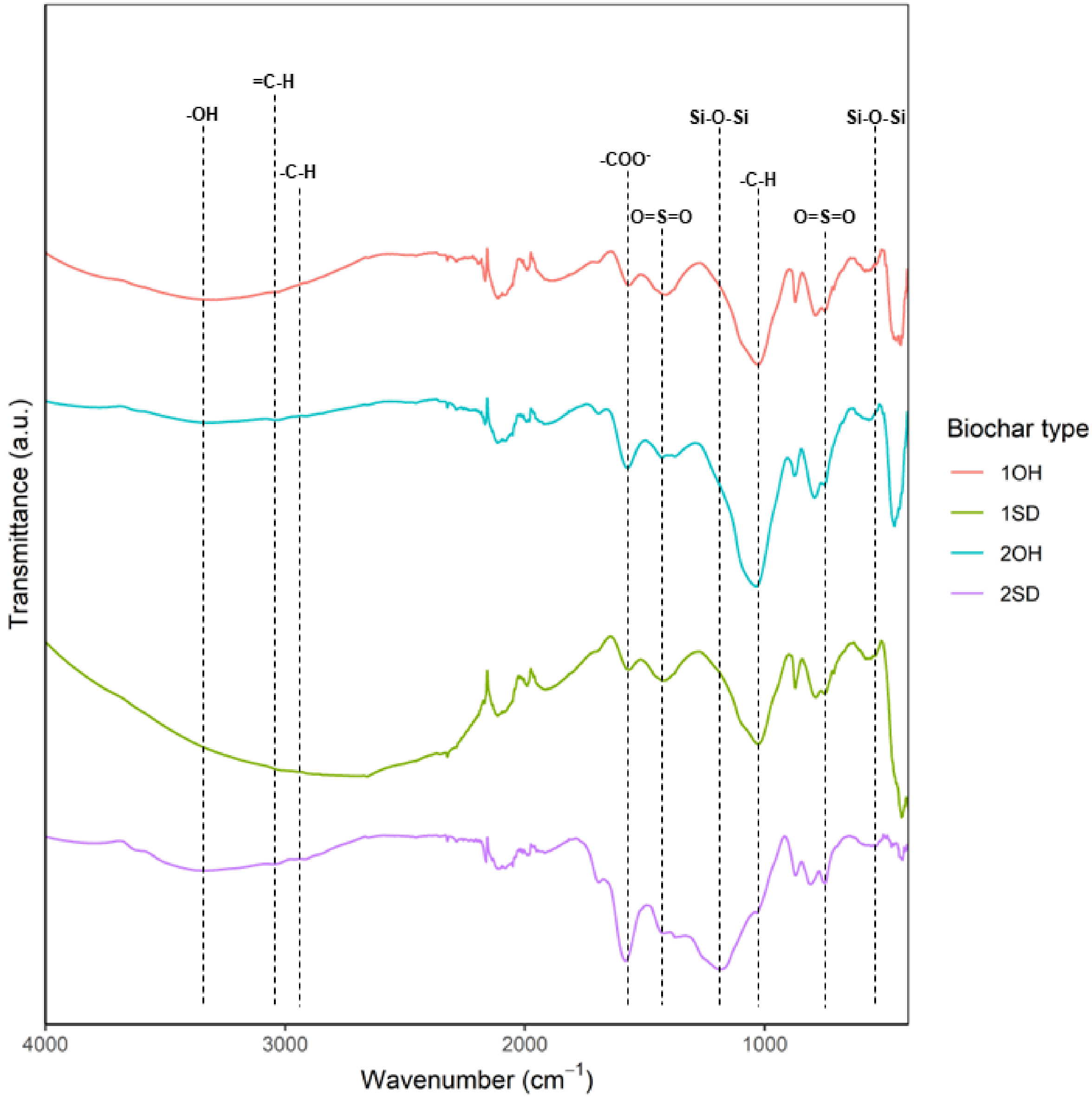

Figure 3.2 FTIR spectra of biochars. Abbreviations of biochar types are explained in the footnote of Table 3.1.

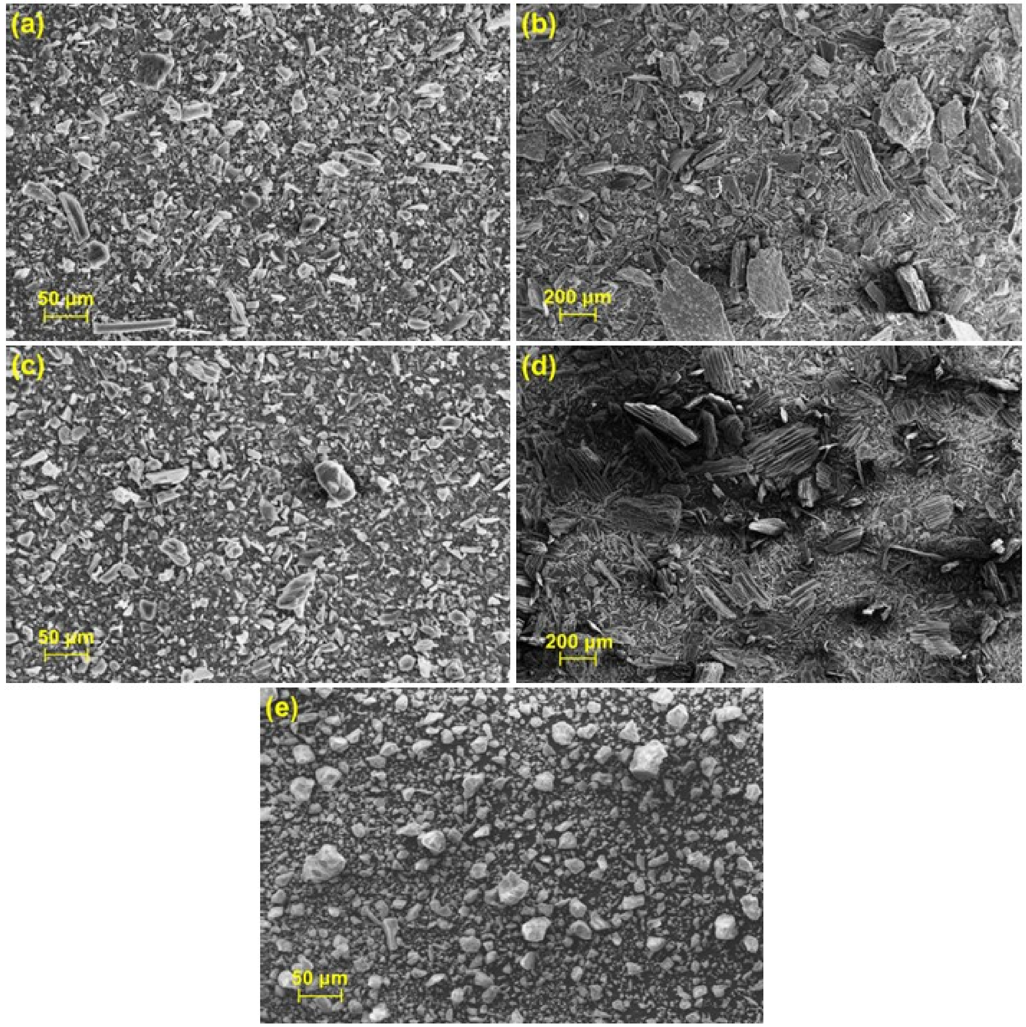

Figure 3.3 SEM images of (a) 1OH, (b) 2OH, (c) 1SD, (d) 2SD biochar and (e) cement particles. Figures (b) and (d) have larger scales than other figures. Abbreviations of biochar types are explained in the footnote of Table 3.1.

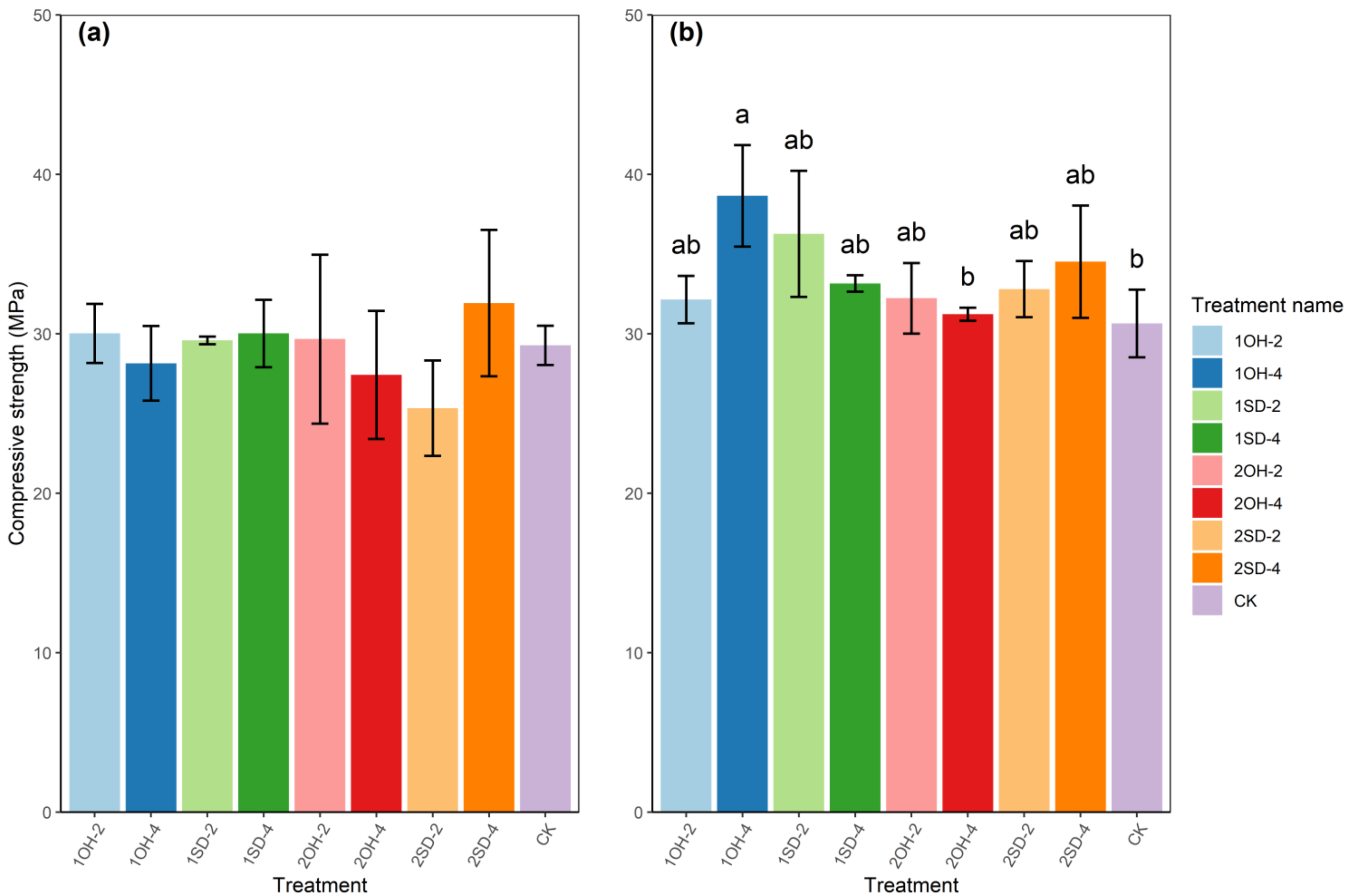

Figure 3.4 The (a) 7-day and (b) 28-day compressive strength of concrete. Black bars represent standard deviations. Different letters in figure (b) represent significant significances among the treatments. The meanings of treatment codes are explained in the footnote of Table 3.1.

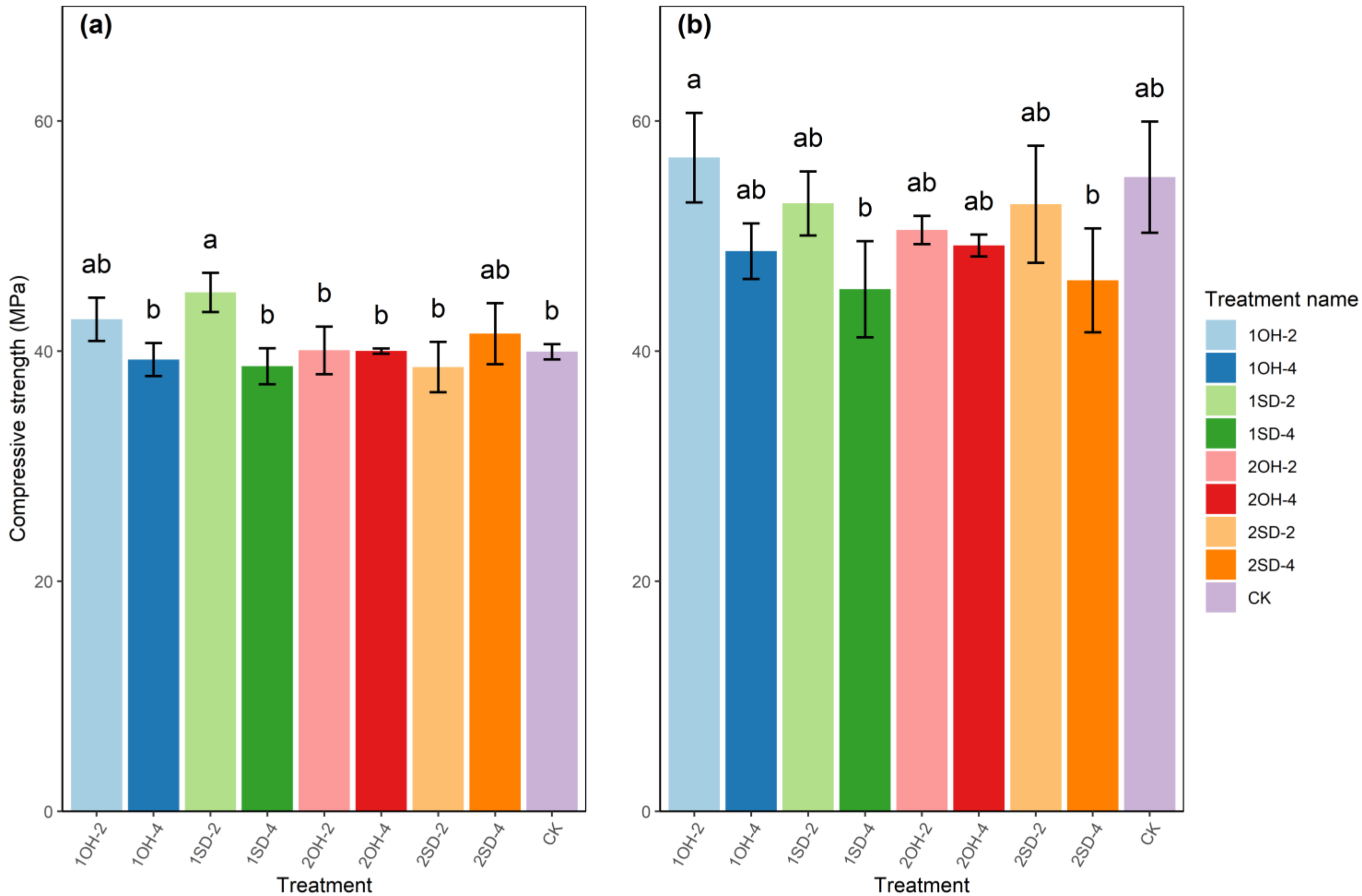

Figure 3.5 The (a) 7-day and (b) 28-day compressive strength of cement paste. Black bars represent standard deviations. Different letters of each figure represent significant significances. The meanings of treatment codes are explained in the footnote of Table 3.1.

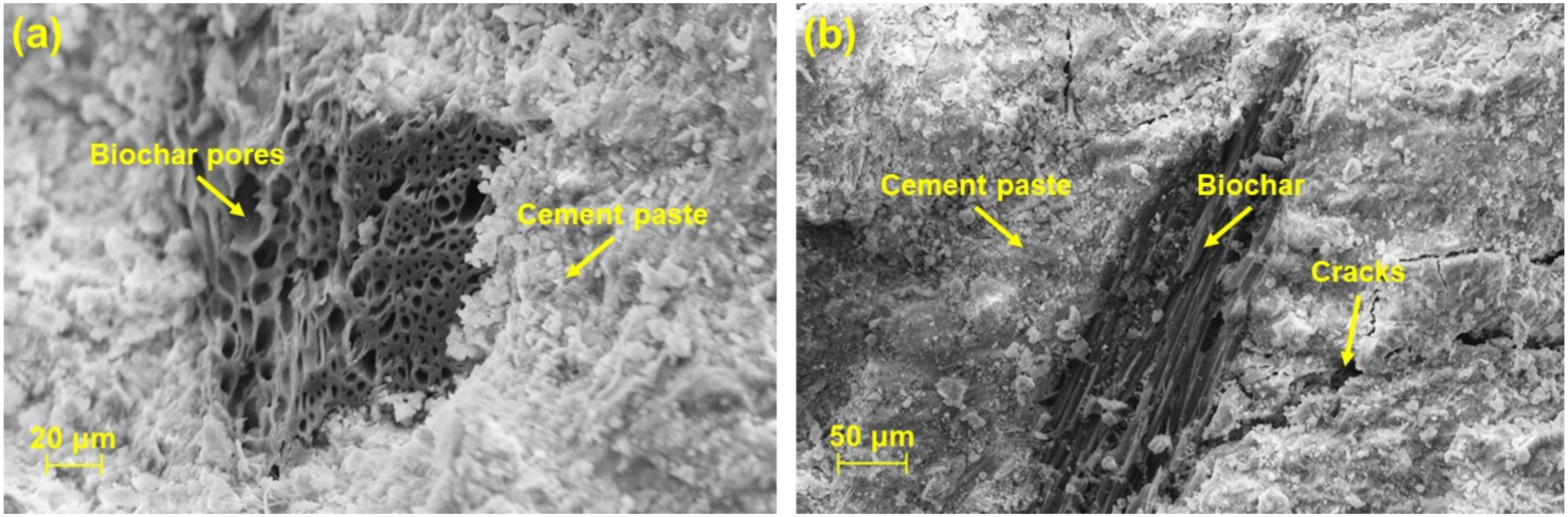

Figure 3.6 The SEM images of pieces of cement paste after 28-day compressive strength measurements for treatments (a) 1OH-2, and (b) 2SD-4. Cracks are shown in (b) around the biochar. The meanings of treatment codes are explained in the footnote of Table 3.1.

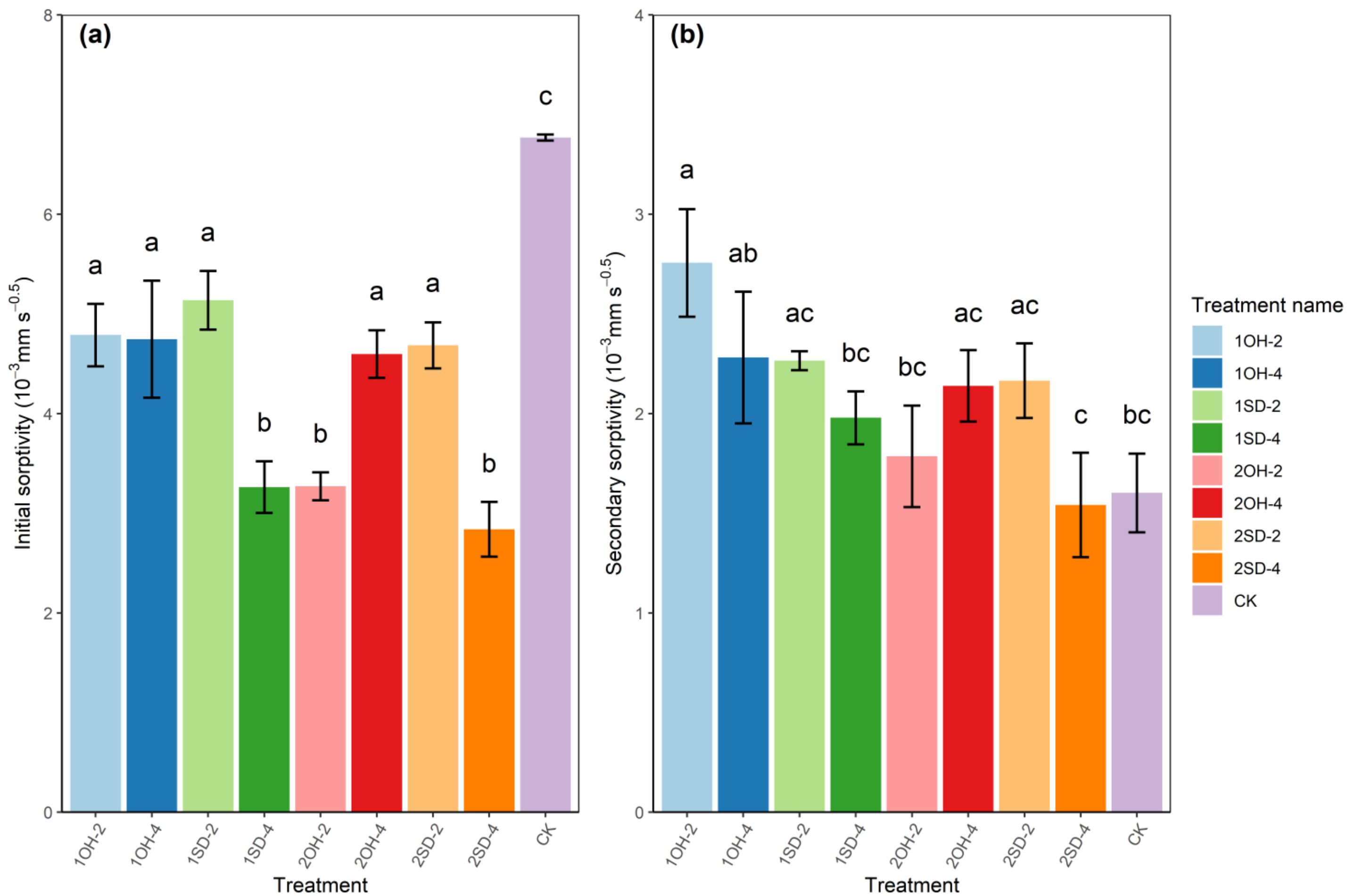

Figure 3.7 The (a) initial sorptivity and (b) secondary sorptivity of concrete. Black bars represent standard deviations. Different letters of each figure represent significant significances among the treatments. The meanings of treatment codes are explained in the footnote of Table 3.1.

Chapter 4: Synthesis and Perspectives

4.1 Synthesis of findings

This thesis research aimed to assess and elucidate the effects of biochar addition on the performances of Portland cement composites through meta-analysis and laboratory experiments. The key findings can be summarized as follows:

(1) Overall, adding biochars can significantly alter the quality of Portland cement composites, based on the meta-analysis and laboratory experiments.

(2) The meta-analysis revealed that plant-based biochars, rather than industrial waste and manure biochars, were more suitable for addition to Portland cement composites. In addition, biochar produced at high temperatures (> 450 °C) with a slow pyrolysis rate (around 10 °C min^{-1}) resulted in the highest positive effects on the performance of Portland cement composites. The smaller particle size of biochars, at least similar to cement particle size (D_{90} is around 40 μm), had better acceleration in the cement hydration process. Although water-presoaking of biochars provides more available water for cement hydration; however, more research is required to validate this.

(3) The meta-analysis also showed that low biochar dosages (< 2.5% of binder weight) improved compressive strength. Biochars can also cooperate with aggregates to affect compressive strength. However, the performance of biochar addition on concrete and mortar was highly affected by the type of Portland cement composites due to the complexity of compositions and properties of aggregates.

(4) Laboratory experiments concluded that the effects of biochar feedstock type were not significant on the performance of Portland cement composites. Biochar addition can alter the slump, fresh air content, 28-day compressive strength, and water sorptivity of concrete. In addition, biochar application significantly altered the quality of cement paste, including compressive strength, final setting time, and hardened density. It was apparent that the performance parameters between cement paste and concrete and their effect sizes were partially correlated, indicating that the binder quality can affect concrete performance.

(5) Although the microstructure of concrete had been affected by biochar addition, the concrete macroscopical structure was not significantly affected due to the low biochar dosage used in this study. However, with the presence of aggregate, the significance of the effects of binder quality alteration was obscured. On the other hand, studying interactions between biochar and other admixtures and exploring the kinetic mechanism during cement hydration is necessary to expand the understanding of the effect of biochar addition. The low molar O/C ratio and high specific surface area of biochars were highly correlated to the improvement effects of biochar addition, which were substantially affected by biochar feedstock type, pyrolysis condition and pre-treatment.

4.2 Limitations and perspectives

The lack of some essential details in the literature included in the meta-analysis is one of the major limitations of the meta-analysis chapter. This limitation may cause uncertainties in the meta-analysis. For instance, misestimation of the variances of parameters is possible due to the missing variance data. In addition, missing information on pyrolysis conditions, including pyrolysis temperature, residence time and heating rate, element content, specific surface area, pore structure, ash and volatile matter contents, and chemical properties of biochars, limited the explanations on the mechanism of the effects and did not enable me to conduct additional correlation analyses, including establishing a structural equation model. Furthermore, due to the lack of data, this study did not include other performance parameters of Portland cement composites, such as flexural strength and durability. In addition, the meta-analysis chapter did not include papers focusing on a combination of biochar and other SCMs, although some studies illustrated improvements in the mechanical performance of Portland cement composites (Akhtar and Sarmah, 2018b; Chen et al., 2022a; Gupta and Kua, 2020), which may limit the application of results in the meta-analysis study. The meta-analysis also did not include the pre-treatment and modification of feedstocks. Finally, this study only focused on one aspect of the mechanical performance of Portland cement composites. More research and review should be conducted in the future on how biochar affects the other aspects of the mechanical performance of Portland cement composites.

Although the laboratory experiments explored the effects of biochar addition on concrete's mechanical performance and water sorptivity, they only explored the general scenario of concrete performance. Expanding this study to more concrete performance parameters is necessary. However, except for general mechanical performances and durability, which included flexural strength, elastic modulus, freeze-thaw resistance and chloride immobilization (Qu et al., 2024; Zaid et al., 2024), exploring kinetic mechanisms of how biochar works in the cementitious matrix are more critical. Several studies have been published to expand the views on the effects of biochar addition in ITZ between aggregate and cementitious matrix (Chen et al., 2024; Zhu et al., 2023) and the foam stabilizing mechanism driven by biochar in foam concrete (Song et al., 2023); those studies provided detailed information on how biochar works. Such explorations are more valuable than adding biochar into Portland cement composites and finding the performance differences, which could pave the way for designing novel SCMs. Besides, since the metal phase in biochar is different in cement, which provides biochar with various physical and chemical properties (Li et al., 2018), the working process during cement hydration might differ from single oxides, which needs further research. In addition, exploring the cooperation between biochar and other admixtures to offset the adverse effects of biochar addition and solve the floating problem of biochar on concrete is critical in future research to expand the potential of biochar utilization. Since biochar potentially contains heavy metals and toxic substances, the health risks of biochar addition to Portland cement composites should also be considered in the future (Duan et al., 2019). Although there are some life-cycle assessments to evaluate how biochar addition could mitigate carbon emissions (Chen et al., 2022c, 2022b), it is necessary to conduct a complete assessment of how this strategy affects any concerned environmental problems.

Based on the review of the literature and my research, I suggest that several aspects need to be addressed in future research:

(1) Details on biochar production conditions and properties, including pyrolysis conditions, proximate analysis, element contents, and physicochemical properties, need to be provided for further analysis and modelling of relationships and mechanisms.

(2) The potential application of modified biochar needs to be explored to increase biochar dosage while maintaining the properties of Portland cement composites.

(3) The analysis of batching factors, including cement types, concrete batching type and combining biochar with other SCMs and reinforcement, needs to be conducted to determine how biochar works in specific Portland cement composites.

(4) The measurements for performance parameters and their relationships of Portland cement composites need to be extended to quantify more effects of biochar addition.

(5) The leaching risk of biochar constituents, such as polycyclic aromatic hydrocarbons and heavy metals, and environment assessment, including carbon sequestration and life-cycle assessment, need to be explored to evaluate the health risks of biochar addition to Portland cement composites and its potential environment benefits.

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Appendix

Appendix. A: Supplementary information for the meta-analysis

Table A1 List of papers used for meta-analysis with the parameters of 7- and 28-day compressive strength.

No.	Papers included in the meta-analysis	7-day compressive strength	28-day compressive strength
1	Ahmad, M.R., Chen, B., Duan, H., 2020. Improvement effect of pyrolyzed agro-food biochar on the properties of magnesium phosphate cement. Sci. Total Environ. 718, 137422.		✓*
2	Akhtar, A., Sarmah, A.K., 2018. Novel biochar-concrete composites: manufacturing, characterization and evaluation of the mechanical properties. Sci. Total Environ. 616–617, 408–416.	✓	✓
3	Aziz, M.A., Zubair, M., Saleem, M., Alharthi, Y., Ashraf, N., Alotaibi, K.S., Aga, O., Al Eld, A.A.A., 2023. Mechanical, non-destructive, and thermal characterization of biochar-based mortar composite. Biomass Conversion and Biorefinery.	✓	✓
4	Belaadi, A., Boumaaza, M., Alshahrani, H., Bourchak, M., 2023. Optimization of Palm Rachis Biochar Waste Content and Temperature Effects on Predicting Bio-Mortar : ANN and RSM Modelling. Journal of Natural Fibers, 20(1), 2151547.	✓	✓
5	Chen, X., Li, J., Xue, Q., Huang, X., Liu, L., Poon, C.S., 2020. Sludge biochar as a green additive in cement-based composites:	✓	✓

	mechanical properties and hydration kinetics. Constr. Build. Mater. 262, 120723.		
6	Chen, T., Zhao, L., Gao, X., Li, L., Qin, L., 2022. Modification of carbonation-cured cement mortar using biochar and its environmental evaluation. Cem. Concr. Compos. 134, 104764.	✓	✓
7	Chen, Z., Wu, N., Song, Y., Xiang, J., 2022. Modification of iron-tailings concrete with biochar and basalt fiber for sustainability. Sustainability 14, 10041.	✓	✓
8	Chen, L., Wang, L., Zhang, Y., Ruan, S., Mechtcherine, V., Tsang, D.C.W., 2022. Roles of biochar in cement-based stabilization/solidification of municipal solid waste incineration fly ash. Chem. Eng. J. 430, 132972.	✓	✓
9	De Carvalho Gomes, S., Zhou, J.L., Zeng, X., Long, G., 2022. Water treatment sludge conversion to biochar as cementitious material in cement composite. J. Environ. Manage. 306, 114463.	✓	✓
10	Gupta, S., 2021. Carbon sequestration in cementitious matrix containing pyrogenic carbon from waste biomass: a comparison of external and internal carbonation approach. J. Build. Eng. 43, 102910.	✓	✓
11	Gupta, S., Kua, H.W., Tan Cynthia, S.Y., 2017. Use of biochar-coated polypropylene fibers for carbon sequestration and physical improvement of mortar. Cem. Concr. Compos. 83, 171–187.	✓	✓
12	Gupta, S., Kua, H.W., Koh, H.J., 2018. Application of biochar from food and wood waste as green admixture for cement mortar. Sci. Total Environ. 619–620, 419–435.	✓	✓
13	Gupta, S., Kua, H.W., Pang, S.D., 2018. Biochar-mortar composite: manufacturing, evaluation of physical properties and economic viability. Constr. Build. Mater. 167, 874–889.	✓	✓

14	Gupta, S., Kua, H.W., 2018. Effect of water entrainment by pre-soaked biochar particles on strength and permeability of cement mortar. Constr. Build. Mater. 159, 107–125.	✓	✓
15	Gupta, S., Kua, H.W., Low, C.Y., 2018. Use of biochar as carbon sequestering additive in cement mortar. Cem. Concr. Compos. 87, 110–129.	✓	✓
16	Gupta, S., Kua, H.W., 2019. Carbonaceous micro-filler for cement: effect of particle size and dosage of biochar on fresh and hardened properties of cement mortar. Sci. Total Environ. 662, 952–962.		✓
17	Gupta S., Kua, H.W., Pang S.D., 2019. Biochar-concrete composite: manufacturing, characterization and performance evaluation at elevated temperature. Academic Journal of Civil Engineering, 37(2), 507-513.	✓	✓
18	Gupta, S., Krishnan, P., Kashani, A., Kua, H.W., 2020. Application of biochar from coconut and wood waste to reduce shrinkage and improve physical properties of silica fume-cement mortar. Constr. Build. Mater. 262, 120688.	✓	✓
19	Gupta, S., Palansooriya, K.N., Dissanayake, P.D., Ok, Y.S., Kua, H.W., 2020. Carbonaceous inserts from lignocellulosic and non-lignocellulosic sources in cement mortar: preparation conditions and its effect on hydration kinetics and physical properties. Constr. Build. Mater. 264, 120214.	✓	✓
20	Gupta, S., Kua, H.W., 2020. Combination of biochar and silica fume as partial cement replacement in mortar: performance evaluation under normal and elevated temperature. Waste Biomass Valorization 11, 2807–2824.	✓	✓

21	Gupta, S., Kua, H.W., Pang, S.D., 2020. Effect of biochar on mechanical and permeability properties of concrete exposed to elevated temperature. Constr. Build. Mater. 234, 117338.	✓	✓
22	Gupta, S., Kashani, A., Mahmood, A.H., Han, T., 2021. Carbon sequestration in cementitious composites using biochar and fly ash – effect on mechanical and durability properties. Constr. Build. Mater. 291, 123363.	✓	✓
23	Gupta, S., Muthukrishnan, S., Kua, H.W., 2021. Comparing influence of inert biochar and silica rich biochar on cement mortar – hydration kinetics and durability under chloride and sulfate environment. Constr. Build. Mater. 268, 121142.	✓	
24	Gupta, S., Kashani, A., 2021. Utilization of biochar from unwashed peanut shell in cementitious building materials – effect on early age properties and environmental benefits. Fuel Process. Technol. 218, 106841.	✓	
25	Gupta, S., Mahmood, A.H., 2022. A multi-method investigation into rheological properties, hydration, and early-age strength of cement composites with admixtures recovered from inorganic and bio-based waste streams. Constr. Build. Mater. 347, 128529.	✓	✓
26	Gupta, S., Mahmood, A.H., 2022. A multi-method investigation into rheological properties, hydration, and early-age strength of cement composites with admixtures recovered from inorganic and bio-based waste streams. Constr. Build. Mater. 347, 128529.	✓	
27	Haque, M.I., Khan, R.I., Ashraf, W., Pendse, H., 2021. Production of sustainable, low-permeable and self-sensing cementitious composites using biochar. Sustain. Mater. Technol. 28, e00279.	✓	✓
28	Haris Javed, M., Ali Sikandar, M., Ahmad, W., Tariq Bashir, M., Alrowais, R., Bilal Wadud, M., 2022. Effect of various biochars on		✓

	physical, mechanical, and microstructural characteristics of cement pastes and mortars. J. Build. Eng. 57, 104850.		
39	Jafari, A., Sadeghian, P., 2023. Influence of biochar and recycled gypsum on the strength and microstructure of conventional and sustainable cementitious composites. Construction and Building Materials, 408, 133715.	✓	✓
30	Khalid, A., Khushnood, R.A., Mahmood, A., 2019. Impact of pyrolytic carbonaceous nano inerts addition on fracture and electromagnetic interference shielding characteristics of cementitious composites. Theor. Appl. Fract. Mech. 103, 102320.		✓
31	Khan, K., Aziz, M.A., Zubair, M., Amin, M.N., 2022. Biochar produced from Saudi agriculture waste as a cement additive for improved mechanical and durability properties—SWOT analysis and techno-economic assessment. Materials 15, 5345.	✓	✓
32	Kim, Y.U., Park, J.H., Yun, B.Y., Yang, S., Wi, S., Kim, S., 2021. Mechanical and thermal properties of artificial stone finishing materials mixed with PCM impregnated lightweight aggregate and carbon material. Constr. Build. Mater. 272, 121882.		✓
33	Liu, W., Li, K., Xu, S., 2022. Utilizing bamboo biochar in cement mortar as a bio-modifier to improve the compressive strength and crack-resistance fracture ability. Constr. Build. Mater. 327, 126917.	✓	✓
34	Li, Z., Xue, W., Zhou, W., 2023. Mechanical Properties of Concrete with Different Carya Cathayensis Peel Biochar Additions. Sustainability, 15(6), 4874.		✓
35	Maljaee, H., Paiva, H., Madadi, R., Tarelho, L.A.C., Morais, M., Ferreira, V.M., 2021. Effect of cement partial substitution by waste-based biochar in mortars properties. Constr. Build. Mater. 301, 124074.	✓	✓

36	Mo, L., Fang, J., Huang, B., Wang, A., Deng, M., 2019. Combined effects of biochar and MgO expansive additive on the autogenous shrinkage, internal relative humidity and compressive strength of cement pastes. Constr. Build. Mater. 229, 116877.	✓	✓
37	Navaratnam, S., Wijaya, H., Rajeev, P., Mendis, P., Nguyen, K., 2021. Residual stress-strain relationship for the biochar-based mortar after exposure to elevated temperature. Case Stud. Constr. Mater. 14, e00540.		✓
38	Park, J.H., Kim, Y.U., Jeon, J., Yun, B.Y., Kang, Y., Kim, S., 2021. Analysis of biochar-mortar composite as a humidity control material to improve the building energy and hygrothermal performance. Sci. Total Environ. 775, 145552.	✓	✓
39	Qin, Y., Pang, X., Tan, K., Bao, T., 2021. Evaluation of pervious concrete performance with pulverized biochar as cement replacement. Cem. Concr. Compos. 119, 104022.	✓	✓
40	Qing, L., Zhang, H., Zhang, Z., 2023. Effect of biochar on compressive strength and fracture performance of concrete. Journal of Building Engineering, 78, 107587.	✓	✓
41	Restuccia, L., Ferro, G.A., 2016. Promising low cost carbon-based materials to improve strength and toughness in cement composites. Constr. Build. Mater. 126, 1034–1043.	✓	✓
42	Restuccia, L., Ferro, G.A., 2018. Influence of filler size on the mechanical properties of cement-based composites. Fatigue Fract. Eng. Mater. Struct. 41, 797–805.	✓	✓
43	Roychand, R., Patel, S., Halder, P., Kundu, S., Hampton, J., Bergmann, D., Surapaneni, A., Shah, K., Pramanik, B.K., 2021. Recycling biosolids as cement composites in raw, pyrolyzed and	✓	✓

	ashed forms: a waste utilisation approach to support circular economy. J. Build. Eng. 38, 102199.		
44	Sirico, A., Belletti, B., Bernardi, P., Malcevschi, A., Pagliari, F., Fornoni, P., Moretti, E., 2022. Effects of biochar addition on long-term behavior of concrete. Theoretical and Applied Fracture Mechanics, 122, 103626.	✓	✓
45	Suarez-Riera, D., Lavagna, L., Bartoli, M., Giorcelli, M., Pavese, M., Tagliaferro, A., 2022. The influence of biochar shape on cement-based materials. Mag. Concr. Res. 1–6.		✓
46	Tan, K., Pang, X., Qin, Y., Wang, J., 2020. Properties of cement mortar containing pulverized biochar pyrolyzed at different temperatures. Constr. Build. Mater. 263, 120616.	✓	✓
47	Tan, K., Qin, Y., Wang, J., 2022. Evaluation of the properties and carbon sequestration potential of biochar-modified pervious concrete. Constr. Build. Mater. 314, 125648.	✓	✓
48	Wang, L., Chen, L., Tsang, D.C.W., Kua, H.W., Yang, J., Ok, Y.S., Ding, S., Hou, D., Poon, C.S., 2019. The roles of biochar as green admixture for sediment-based construction products. Cem. Concr. Compos. 104, 103348.	✓	✓
49	Yang, X., Wang, X.Y., 2021. Hydration-strength-durability-workability of biochar-cement binary blends. J. Build. Eng. 42, 103064.	✓	✓
50	Zhang, Y., Xu, H., Fang, S., Li, D., Xue, W., Chen, B., Zhao, L., 2022. Biochar as additive for improved building performances and heavy metals solidification of sediment-based lightweight concrete. Environ. Sci. Pollut. Res. 30, 4137–4150.		✓
51	Zhang, Y., Maierdan, Y., Guo, T., Chen, B., Fang, S., Zhao, L., 2022. Biochar as carbon sequestration material combines with		✓

sewage sludge incineration ash to prepare lightweight concrete.

Constr. Build. Mater. 343, 128116.

*: Indicating that a particular publication includes data for the parameter listed in the column heading.

Table A2 Particle size distribution (mean ± standard deviation) of ground and original biochars used for testing 7-day compressive strength. D_{10}, D_{50} and D_{90}: the maximum diameter containing 10%, 50% and 90% of the mass of the sample. WPM: without physical modification.

Modification	**Cement**			**Biochar**			**Sample size (n)**
	D_{10} (µm)	**D_{50} (µm)**	**D_{90} (µm)**	**D_{10} (µm)**	**D_{50} (µm)**	**D_{90} (µm)**	
Grinding	3.6 ± 1.6	17.1 ± 5.1	39.3 ± 8.4	3.4 ± 1.4	13.4 ± 9.1	46.4 ± 36.1	63
WPM	2.4 ± 0.9	14.9 ± 3.1	45.1 ± 5.0	4.2 ± 3.1	63.6 ± 65.4	280 ± 293	54

Table A3 Particle size distribution (mean ± standard deviation) of ground and original biochars used for testing 28-day compressive strength. D_{10}, D_{50} and D_{90}: the maximum diameter containing 10, 50 and 90% of the mass of the sample. WPM: without physical modification.

Modification	**Cement**			**Biochar**			**Sample size (n)**
	D_{10} (μm)	**D_{50} (μm)**	**D_{90} (μm)**	**D_{10} (μm)**	**D_{50} (μm)**	**D_{90} (μm)**	
Grinding	3.4 ± 1.5	16.3 ± 4.4	41.4 ± 11.2	3.2 ± 1.1	11.8 ± 3.9	43.6 ± 22.5	65
WPM	3.1 ± 1.3	15.0 ± 2.4	43.5 ± 4.5	3.3 ± 2.9	45.2 ± 61.0	201 ± 271	78

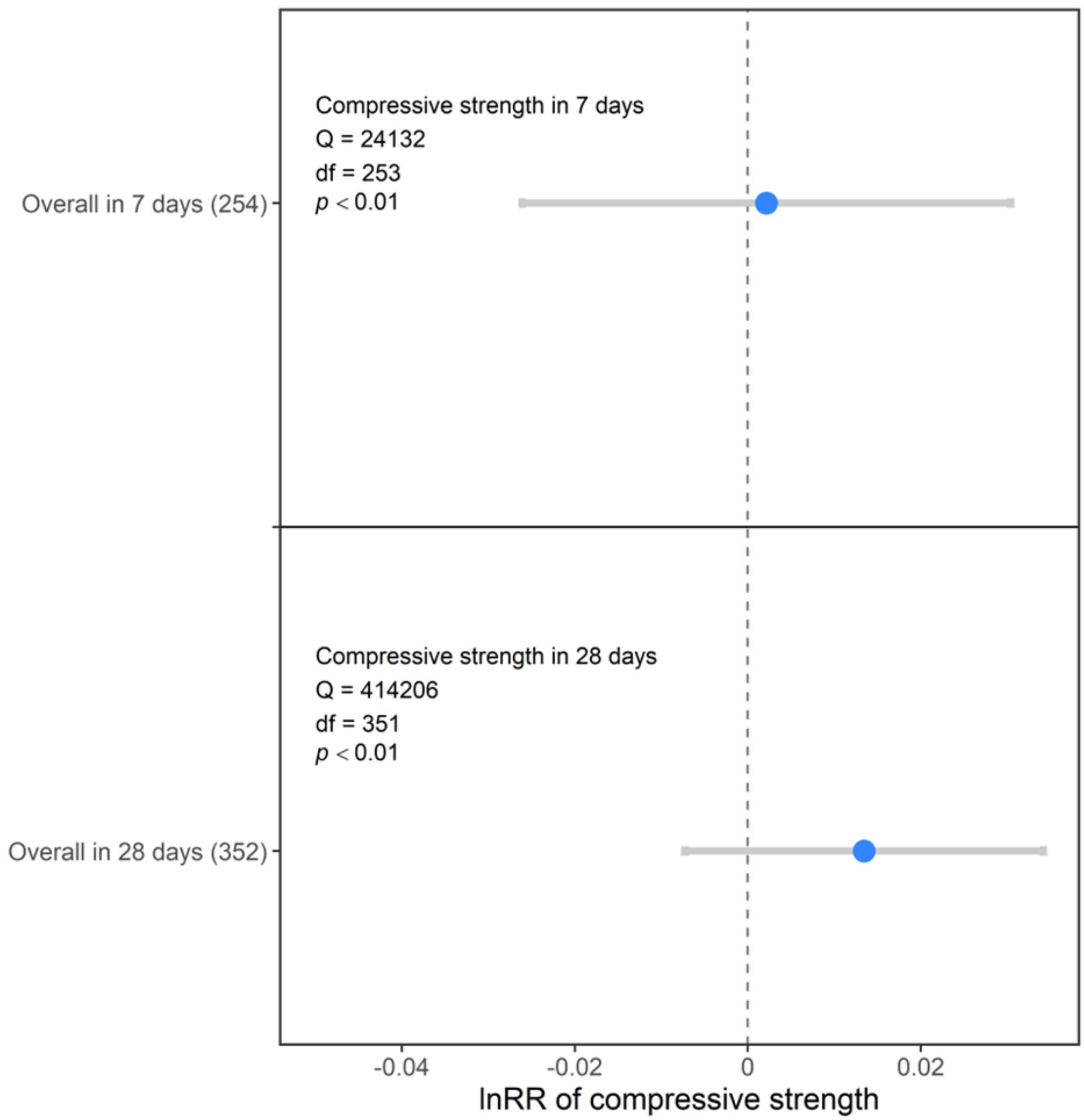

Figure A1 The overall effect sizes of biochar addition on 7- and 28-day compressive strength of Portland cement composites. Each point represents effect sizes, and the size of the point represents the relative number of records compared to the total records. Grey bars represent 95CI. The vertical dash line represents the value of 0. The numbers of records are indicated in the brackets.

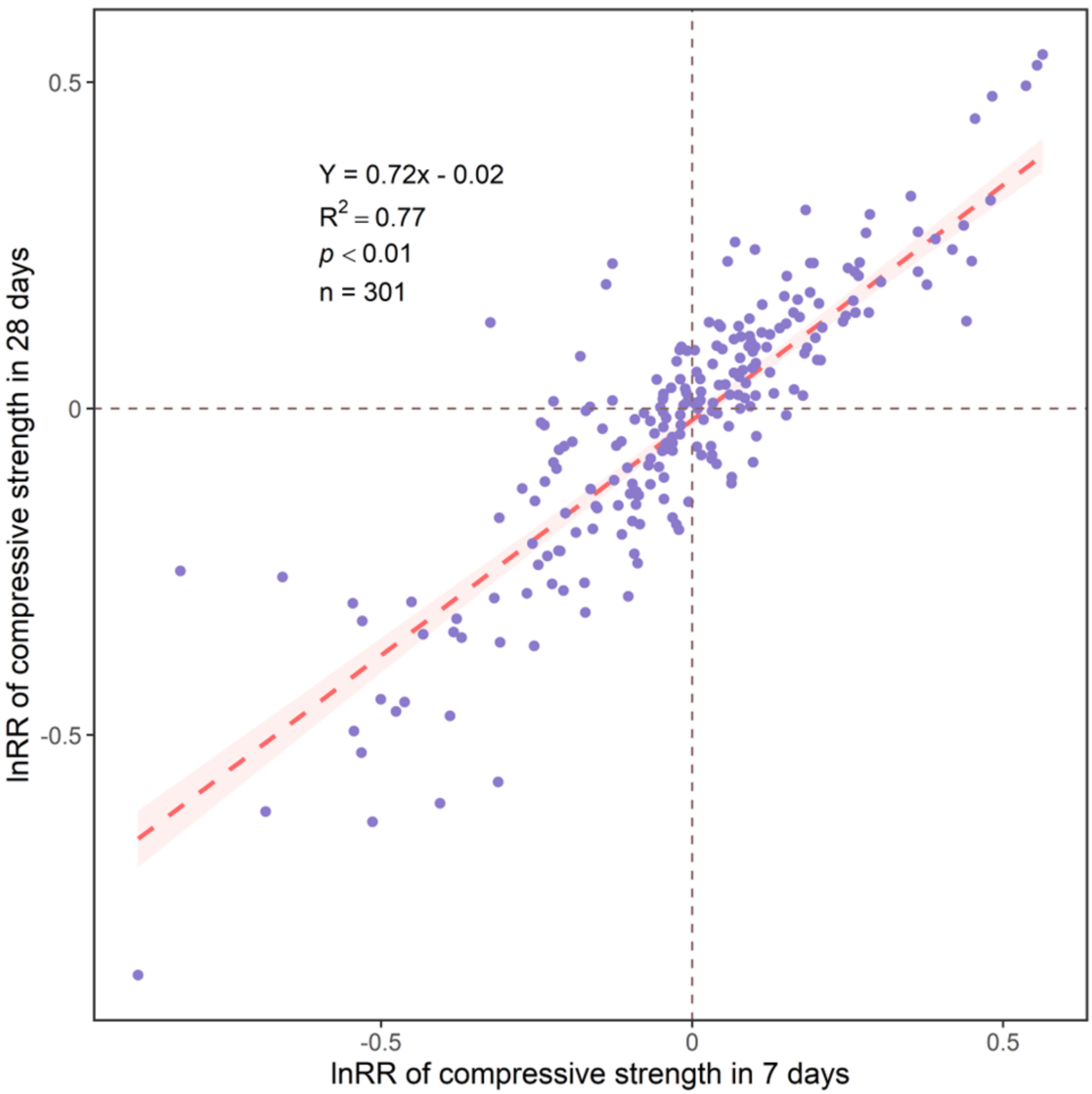

Figure A2 Linear relationship between effect sizes of biochar addition on 7- and 28-day compressive strength of Portland cement composites. Points in the figure represent paired records. The simple linear regression line with 95% confidential intervals is shown, with the number of records (n) presented. The horizontal dash lines represent the value of 0.

Appendix. B: List of symbols and abbreviations

°C – degree Celsius

μm – micrometre

CK – batching without adding biochar, for concrete and cement paste

1OH – oat hull biochar with blender grinding

1OH-2 – batching in adding oat hull biochar with blender grinding with the dosage of 2wt% of binder, for concrete and cement paste

1OH-4 – batching in adding oat hull biochar with blender grinding with the dosage of 4wt% of binder, for concrete and cement paste

1SD – sawdust biochar with blender grinding

1SD-2 – batching in adding sawdust biochar with blender grinding with the dosage of 2wt% of binder, for concrete and cement paste

1SD-4 – batching in adding sawdust biochar with blender grinding with the dosage of 4wt% of binder, for concrete and cement paste

2OH – oat hull biochar with automatic mill grinding

2OH-2 – batching in adding oat hull biochar with automatic mill grinding with the dosage of 2wt% of binder, for concrete and cement paste

2OH-4 – batching in adding oat hull biochar with automatic mill grinding with the dosage of 4wt% of binder, for concrete and cement paste

2SD – sawdust biochar with automatic mill grinding

2SD-2 – batching in adding sawdust biochar with automatic mill grinding with the dosage of 2wt% of binder, for concrete and cement paste

2SD-4 – batching in adding sawdust biochar with automatic mill grinding with the dosage of 4wt% of binder, for concrete and cement paste

Al – aluminum

C – carbon

Ca – calcium

C-S-H – calcium silicate hydrate

D_{10} – 10% of the total volume of material in the sample is below the named micron size

D_{50} – 50% of the total volume of material in the sample is below the named micron size

D_{90} – 90% of the total volume of material in the sample is below the named micron size

Fe – iron

FTIR – fourier-transform infrared spectroscopy

h – hour

H – hydrogen

ICP-OES – inductively coupled plasma-optical emission spectrometry

ITZ – interfacial transition zone

K – potassium

kg - kilogram

kN – kilonewton

lnRR – logarithmic effect size

Mg – magnesium

min – minute

mm – millimeter

MPa – megaPascal

n – replication

Na – sodium

O – oxygen

OH – oat hull

s - second

SCMs – supplementary cementitious materials

SD – sawdust

Si - Silicon

XRD – X-ray diffraction analysis