Journal of Environmental Management 222 (2018) 76–85

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Effects of biochar on availability and plant uptake of heavy metals – A meta-analysis

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ARTICLE INFO

Keywords:
Availability
Biochar
Heavy metal contamination
Meta-analysis
Plant uptake
Soil remediation

ABSTRACT

Biochar can be an effective amendment for immobilizing heavy metals in contaminated soils but has variable effects depending on its chemical and physical properties and those of the treated soil. To investigate the range of biochar's effects on heavy metal accumulation in plants in responses to the variation of soil, biochar and plant, we carried out a meta-analysis of the literature that was published before March 2016. A total of 1298 independent observations were collected from 74 published papers. Results showed that across all studies, biochar addition to soils resulted in average decreases of 38, 39, 25 and 17%, respectively, in the accumulation of Cd, Pb, Cu and Zn in plant tissues. The effect of biochar on heavy metal concentrations in plants varied depending on soil properties, biochar type, plant species, and metal contaminants. The largest decreases in plant heavy metal concentrations occurred in coarse-textured soils amended with biochar. Biochar had a relatively small effect on plant tissue Pb concentrations, but a large effect on plant Cu concentrations when applied to alkaline soils. Plant uptake of Pb, Cu and Zn was less in soils with higher organic carbon contents. Manure-derived biochar was the most effective for reducing Cd and Pb concentrations in plants as compared to biochars derived from other feedstock. Biochar having a high pH and used at high application rates resulted in greater decreases in plant heavy metal uptake. The meta-analysis provides useful guidelines on the range of effects that can be anticipated for different biochar materials in different plant-soil systems.

1. Introduction

Soil contamination with heavy metals is a major environmental concern that has emerged with the rapid development of industrial activities in the world over the last century. Heavy metals that are subsequently taken up by plants enter into the food chain and accumulate in animals and humans where they can cause toxicity (Dudka and Miller, 1999; Reeves and Chaney, 2008; Singh et al., 2010). Many factors affect the uptake process of metals by plants, such that remediation of heavy metal polluted soils presents a considerable challenge. Various methods for treatment of contaminated soils include phytoextraction (Kumar et al., 1995), chemical stabilization (Kumpiene et al., 2008), and soil washing (Abumaizar and Smith, 1999). In-situ immobilization of metals via chemical stabilization is a particularly convenient and cost-effective way to reduce heavy metal bioavailability and uptake by plants (Guo et al., 2006; Martin and Ruby, 2004). Among the amendments that are used to adsorb heavy metals and decrease their potential bioavailability, biochar has been shown to be particularly effective (Beesley et al., 2011; Houben et al., 2013).

Biochar is a carbon rich material produced by pyrolysis of straw, manure, wood, and other agricultural wastes under oxygen-limited conditions (Lehmann and Joseph, 2009). Under current agricultural and environmental practices, improper disposal or burning of agricultural organic wastes is a waste of resources and cause of environmental pollution (Segat et al., 2015; Zhang et al., 2016), while conversion of agricultural wastes into biochar is a multi-win strategy that is beneficial for soil carbon storage (Bolan et al., 2013), mitigation of greenhouse gases emissions (Zhang et al., 2010), improvement soil

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https://doi.org/10.1016/j.jenvman.2018.05.004
Received 9 December 2017; Received in revised form 2 May 2018; Accepted 2 May 2018

fertility (Doan et al., 2015), and immobilization of organic and heavy metal pollutants (Inyang et al., 2016; Lehmann and Joseph, 2015). Studies have shown that biochar is effective for reducing the bioavailability of heavy metals (Ahmad et al., 2014), thereby reducing plant uptake (Fellet et al., 2014) and food chain transfer (Khan et al., 2013). Biochar has highly condensed aromatic structures that make it resistant to microbial decomposition, which allows it to persist for decades to centuries, as evidenced by the anthrosol soils where charcoal was used as a soil amendment (Kuzyakov et al., 2014; Lehmann et al., 2006). Ageing of biochar also may enhance its ability to stabilize heavy metals in soil. Biochar undergoes oxidation over time, which promotes increases in carboxyl groups and the net negative charge that generates its cation exchange capacity (Bian et al., 2014). Thus there may be a long term effect of biochar on stabilizing heavy metals depending on its persistence in soil and increase in charge over time. Variations in the efficacy of biochar for immobilization of heavy metals can be attributed to differences in pH that affect the pH dependent charge (Yuan et al., 2011), as well as the pyrolysis temperature and feedstock that affect the abundance of functional groups that form metal complexes (Uchimiya et al., 2011). Immobilization of heavy metals is also affected by the mineral content of the associated ash in most biochar products (e.g. phosphate) (Cao et al., 2009) and by differences in the surface area and porosity of biochar (Harvey et al., 2011). These properties are dependent not only on the feedstock material, but can be manipulated by controlling the pyrolysis temperature, and other production conditions (e.g. heating rate and residence time) (Kloss et al., 2012; Zhao et al., 2013). The efficacy of particular biochar materials will further depend on soil properties (Ahmad et al., 2014), the specific heavy metals that are targeted (Beesley et al., 2011), and differences among plant species in their root growth patterns and in their abilities to take up and accumulate heavy metals (Rizwan et al., 2016).

To quantitatively and systematically examine the range of biochar's effects on soil heavy metal availability and plant uptake, we carried out a meta-analysis of data from previously published studies. Variables that were considered included soil physical and chemical properties, the type and application rate of biochar and crop type. The results provide useful insights into which biochars are most effective, and the extent to which heavy metal uptake may be affected by different biochar types and soils.

2. Materials and methods

2.1. Data sources and compilation

Relevant scientific articles were collected using the search terms “biochar” or “bio-char” to search for articles in the databases at the Web of Science, Elsevier, Springerlink, Wiley online, and Google Scholar. The search included all relevant articles up until March 1, 2016. Although the terms “char”, “black carbon” and “charcoal” have commonly been included in some former meta-analysis, here we mainly focused on “biochar” as this term intentionally denotes its use for agricultural and environmental applications (Lehmann et al., 2006; Lehmann and Joseph, 2009), as opposed to studies on naturally occurring black carbon. The definition of “biochar” was formally established in 2006 (Lehmann et al., 2006), therefore, the studies including “biochar” were published mainly after that year. The title and abstract of each article were examined and the articles relevant to heavy metal uptake by plant in soils treated with and without biochar were selected.

Data were compiled from the literature reporting the most common heavy metals of concern (Pb, Cd, Cu and Zn) in studies that specifically compared plant uptake of these metals in soils with and without a biochar amendment. Although of interest, there were too studies on the effect of biochar on arsenic uptake, which was not included in this meta-analysis because of the insufficient sample size. If the results were presented in figures, the data were numerically extracted using GetData software (version 2.26). In total, data were extracted from 74 scientific papers containing a total of 1298 individual observations comparing control (no biochar treatments) and biochar-amended treatments (see Supplementary Material 1). The basic properties of the soils and biochars were collected along with descriptions of the soil chemical and physical properties, and the crop type. Soil properties included soil organic carbon (SOC), total nitrogen, pH, cation exchange capacity (CEC), and texture. Biochar variables included feedstock, pyrolysis temperature, total organic carbon, total nitrogen, pH, and the amount applied. Experimental type included pot study and field study. Data collection also included the contents of heavy metals in the studied soils and biochars. Detailed description of the variables are listed in Supplementary Table 1.

The experiments that were evaluated in the present study were mainly conducted in China and Europe, accounting for 34 and 31% of the total studies, respectively (Fig. S1). Other reports were from South Korea (7%), Australia (7%), New Zealand (3%), and other areas (18%). Most of the studies (85%) employed pot trials, and 15% were conducted using field experiments.

2.2. Data normalization

Standard deviation (SD) was used as a measure of variance, and was calculated from the measured variance in each published study (Abalos et al., 2014). When standard errors (SE) were provided, they were transformed to standard deviations according to the following equation:

$$\mathrm{SD} = \mathrm{SE}\sqrt{\mathrm{n}} \tag{1}$$

where n is the number of replications. If the pH was measured with $CaCl_2$ solution, the values were transformed to acidity values predicted to be measured with deionized water using the following formula (Biederman and Harpole, 2013; Cayuela et al., 2014):

$$\mathrm{pH}_{[\mathrm{H2O}]} = 1.65 + 0.86{*}\mathrm{pH}_{[\mathrm{CaCl2}]} \tag{2}$$

Soil organic matter (SOM) values were converted to SOC content by multiplying them by the Bemmelen index value of 0.58 (Liu et al., 2015). Soil texture was classified into three categories of coarse (sandy loam, sandy clay loam, loamy sand and sand), medium (clay loam, loam, silty clay loam, silt, silt loam) and fine (clay, silt clay, loamy clay, sandy clay) according to the methods described in prior former studies (Cayuela et al., 2014; Liu et al., 2015).

2.3. Meta-analysis

Meta-analysis estimates the magnitude of change in a property (also named “effect size”) in response to an experimental treatment across a wide range of variables (Hedges et al., 1999; Kelley and Preacher, 2012). The *response ratio* (R), which is the ratio of measured quantity in experimental and control groups, is usually used to measure the effect size because it quantifies the proportionate change resulting from an experimental manipulation (Hedges et al., 1999). R was normally transformed to its natural logarithms (ln) to obtain a near normal distribution of data according to the following equation:

$$\mathrm{R} = \ln(\mathrm{X_t}/\mathrm{X_c}) = \ln(\mathrm{X_t}) - \ln(\mathrm{X_c}) \tag{3}$$

where X_t and X_c are means of the variable in biochar treatment and control groups, respectively. Its variance (υ) was estimated as:

$$\upsilon = \frac{S_t^2}{n_t x_t^2} + \frac{S_c^2}{n_c x_c^2} \tag{4}$$

where n_t and n_c are the sample sizes for the treatments and control groups, respectively; S_t and S_c are the standard deviations for the treatment and control groups. The Q statistic was used to measure the heterogeneity of effect sizes among studies (Zhou et al., 2016). The total heterogeneity (Q_t) of R among studies consists of within-group (Q_w) and between-group (Q_b) heterogeneity (Wang et al., 2016). A Q_b larger than a critical value indicates significant difference between groups

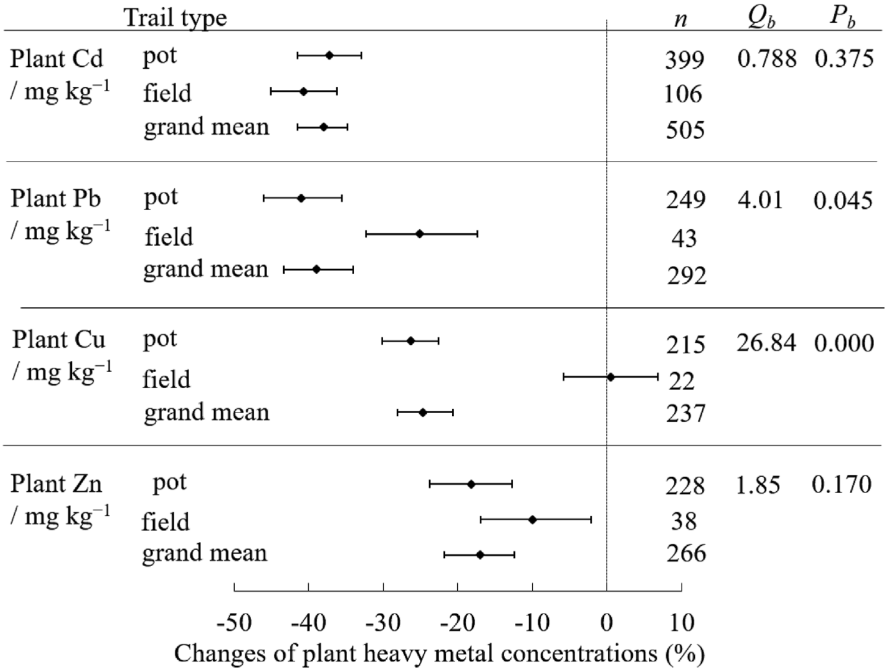

Fig. 1. Effect of biochar addition on heavy metal concentration in plants in field trials and pot studies. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

($P_b < 0.05$). Mean effect sizes and 95% confidence intervals (CI) were calculated using MetaWin (version 2.1). The response ratio and CI of treatments presented were transformed to percent change from lnR (Nguyen et al., 2017). Biochar treatment was considered significant if the 95% CI of response ratio did not overlap with zero in each figure. Responses among groups were considered different if their 95% CIs did not overlap (Wang et al., 2016). Figures were generated using Microsoft Excel (version 2016).

3. Results and discussion

3.1. Overall effect of biochar on plant heavy metal concentrations

In general, biochar consistently reduced the average concentrations of heavy metals in plant tissue as compared to plants grown in soils without biochar (Fig. 1, grand mean). Across all studies, the mean concentrations of Cd, Pb, Cu and Zn in plant tissues decreased by 38, 39, 25, and 17%, respectively, when the plants were grown in soils amended with biochar. The decreases in plant heavy metal concentrations corresponded with concomitant decreases in metal bioavailability as measured using various extraction methods and soil extractants (e.g. $CaCl_2$, NH_4NO_3, DTPA) (Rao et al., 2008). Fig. S2 shows that the average concentrations of available Cd, Pb, Cu, and Zn in soil were reduced by 52, 46, 29, and 36%, respectively, following biochar application. The decreases in soil heavy metal availability were primarily attributed to the immobilization of these heavy metals onto biochar particles, and also to indirect effects of biochar on soil properties and plant uptake processes. Mechanisms of heavy metal immobilization by biochar include adsorption (Inyang et al., 2016), ion exchange (Ding et al., 2014), complexation (Lu et al., 2012), and precipitation reactions (Cao et al., 2009). Along with direct interactions with biochar, changes in soil properties caused by biochar application can also indirectly increase the capacity of soil particles to absorb, complex, and precipitate heavy metals, which reduces their availability (Beesley et al., 2011; Rizwan et al., 2016). The results of the present meta-analysis indicated that biochar greatly increased pH, SOC, EC and CEC of soils (Fig. S3), which is beneficial for heavy metal retention. Besides, biochar may reduce the translocation of heavy metals from root to above-ground tissues. Root to shoot translocation is a key factor determining shoot and grain Cd accumulation in rice (Uraguchi et al., 2009; Yu and Zhou, 2009). Chen et al. (2016) found that the translocation of Cd from rice root to shoot decreased with wheat straw biochar application. Zhang et al. (2013) also found the decreased translocation of Cd in *Juncus subsecundus* when biochar was incorporated into the soil at 5% (w/w). One of the possible factors is that biochar can increase Si concentration in plant (Chen et al., 2016), which may co-deposit with heavy metals and further hinder heavy metal translocation within plants (Liang et al., 2005; Neumann and Zur Nieden, 2001).

Average concentrations of Cd, Pb, and Zn in plant tissues were reduced in both pot trials and field studies following biochar application (Fig. 1). Plant Cu concentrations were reduced only in pot experiments, without significant effects in field studies overall. The effects of biochar on plant Pb concentrations were greater in pot experiments than under field conditions. Possibly this is related to the generally higher biochar application doses in pot trials than in field studies. Differences may also be related to variations in root spread and depth, and in spatial concentrations of the metal in field soils, whereas there is more uniformity and homogeneity in pot studies.

3.2. Effect of biochar on plant heavy metal concentration in response to soil conditions

The effects of biochar on plant heavy metal uptake were generally dependent on soil pH, but were inconsistent and varied for different metals (Fig. 3). Biochar resulted in large reductions of plant Cd concentrations (32–40%) in acid, neutral, and alkaline soils, and there were no differences in the reductions among the three groups (Fig. 2), implying that soil pH exerted the least effect on the plant response to biochar for Cd. This may be because Cd is much more likely to accumulate in crops than other metals examined here (Hooda and Alloway, 1993), and biochar application effectively limits its bioavailability in soils with different pH. The effect of biochar on Pb uptake was highly dependent on soil pH, with an average decrease of 40, 44, and 20% in acid, neutral, and alkaline soils, respectively. Pb is more easily stabilized than other heavy metals and is especially sensitive to pH (Wu et al., 1999). As expected, the concentrations of available Pb in the soils studied here were much higher in acid soils than in alkaline soils; thus the application of alkaline biochar is effective for stabilizing Pb in acid soils. In contrast, plant uptake of Cu in biochar amended soils was inversely related to pH, and was greatest in alkaline soil (39%) as

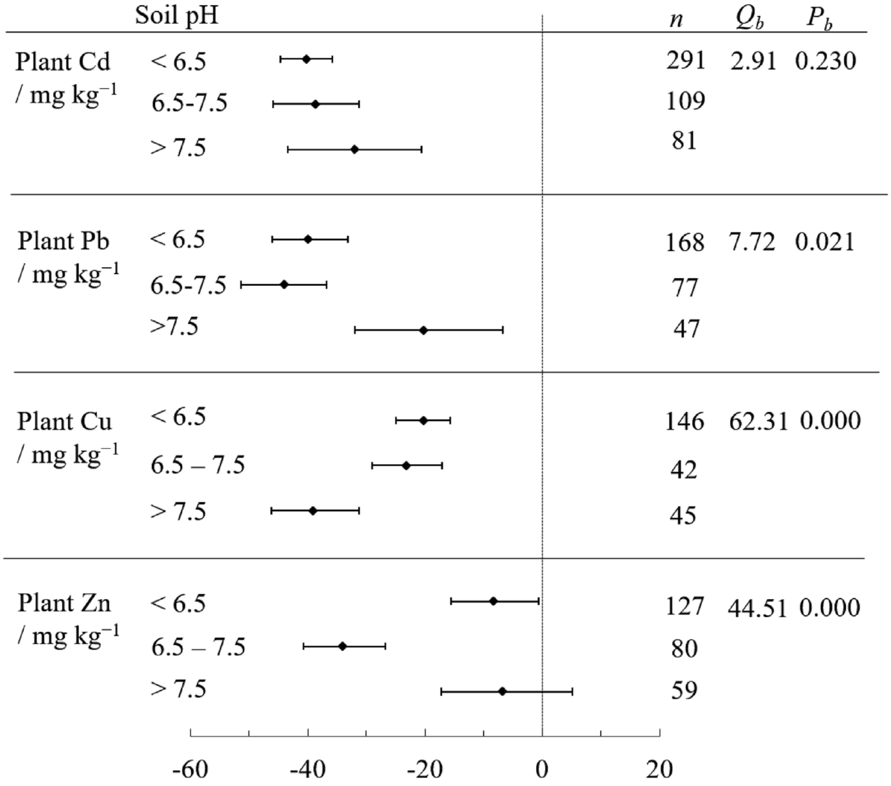

Fig. 2. Effect of biochar addition on plant heavy metal concentrations in response to soil pH. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

compared to neutral (23%) and acid soils (20%). This may be explained by the fact that Cu^{2+} is readily complexed by dissolved organic matter (DOM) as compared to other metals (Ginocchio et al., 2002; Temminghoff et al., 1997). Increases in soil pH promote dissolution of SOM, which further increases DOM content (Oste et al., 2002). McBride and Blasiak, 1979 found that the fraction of complexed Cu^{2+} in soil solution increased dramatically with pH until the soluble complexed Cu^{2+} accounted for 99.9% of the total soluble Cu^{2+} at pH 8, indicating that most of the solution Cu was in complexed form. Biochar application to acid soils increases soil pH, which may induce complexation of Cu^{2+} with DOM, increasing the mobility and availability of Cu. Biochar application to alkaline soil will have a smaller effect on pH, but the retention of Cu in soil is still enhanced by biochar. Possibly this may explain the greater decreases of plant Cu concentration in alkaline soils as compared to acidic soils. Showing yet another pattern, plant Zn concentration showed the greatest decrease in response to biochar in neutral pH soils (34%), as compared to acid soils (8%), but was not decreased in alkaline soils.

Biochar is usually alkaline, which is beneficial for increasing soil pH, especially for acid soils (Chintala et al., 2014; Yuan et al., 2011). Increases of soil pH may cause metal precipitation (Lindsay, 1979), decrease metal solubility (McBride et al., 1997) and promote metal adsorption onto soil (Ma et al., 2010) as soil net negative charge is increased (Naidu et al., 1997). Studies also indicate that biochar may change the redox of soil, which further affects the bioavailability of heavy metals (Choppala et al., 2012; Joseph et al., 2015). Choppala et al. (2012) found that biochar reduced the leaching of Cr in soil as a result of reduction of mobile Cr (VI) to less mobile Cr (III). Biochar also can change the speciation of heavy metals in soil. Both exchangeable Cd and Pb can be transformed to be more stable species that are associated with the oxidizable and residual fractions obtained using soil extraction (Zhu et al., 2015). The transformations of heavy metals to different hydrolysis species will vary depending on the pH and redox that control chemical equilibria between soluble hydrolysis species and the various mineral phases that are present in a particular soil (Tack, 2010). According to the Eh-pH diagram, the species of Cd in solutions were mainly free ion (Cd^{2+}), while the species were mainly free ion (Pb^{2+}) and hydroxide ($PbOH^{+}$) for Pb in a routine Eh-pH range (Brookins, 1986; Karbassi et al., 2018). Altogether, this various affects the various responses of plants to biochar amendments in heavy metals contaminated soils. As mentioned above, the heavy metals exist in different species in both soil solutions and soils due to their inherent nature, this may contribute to different responses to biochar application to a certain extent.

The efficacy of biochar for reducing Cd, Pb, Cu, and Zn concentrations in plant tissue varied depending on soil texture classified as fine (clay), medium (loam), and coarse (sandy) (Fig. 3). There were greater reductions in plant Cd concentrations in medium textured soil than in fine textured soil. Plant Pb concentrations underwent greater reductions in coarse textured soil than in medium or fine textured soil amended with biochar. Soil texture is an important factor controlling heavy metal availability. It is assumed that there were higher heavy metal availabilities in coarse and medium textured soils than in fine textured soils with a high content of clay minerals (Brümmer, 1986; Tiller et al., 1984). Thus biochar is more prone to reduce heavy metal availability in coarse or medium soils.

Average changes in heavy metal concentrations in plant tissues varied significantly in relation to SOM content (Fig. 4). Overall, there were greater decreases of plant heavy metal concentrations in soils containing higher levels of SOC than in soils with low SOC. With respect to Cd, the effects of biochar on reduction in plant tissue Cd concentrations were greater in soils having medium SOC levels ($15–30\,g\,kg^{-1}$) than in soils with low SOC levels ($< 15\,g\,kg^{-1}$). Plant Pb concentrations were reduced by 30 and 54%, respectively in soils with low SOC content and high SOC content ($> 30\,g\,kg^{-1}$). Plant Zn concentrations were not affected by a biochar amendment in soils with low SOC content, but were reduced in soils with medium and high SOC levels. The greater efficacy of biochar in soils with medium and high SOC may be explained by: 1) Soil pH is often inversely correlated with SOM content due to generation of acidity during organic matter decomposition (McCauley et al., 2009; Xiao et al., 2018), thus the addition of alkaline biochar would have a greater impact on higher organic

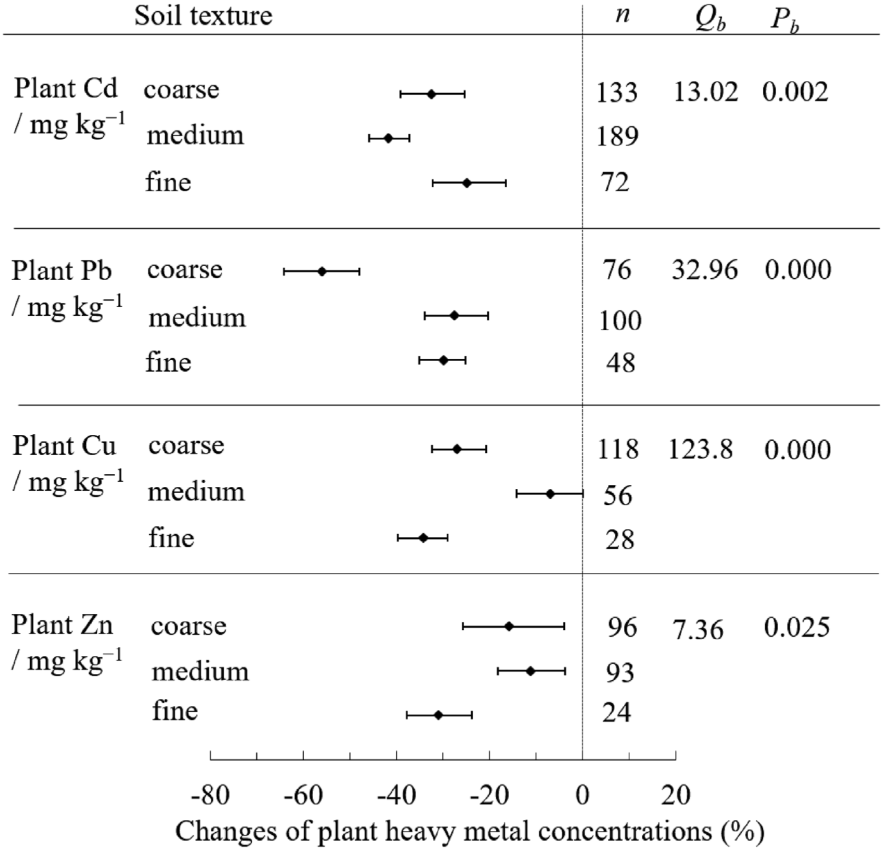

Fig. 3. Effect of biochar addition on plant heavy metal concentrations as affected by soil texture. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

carbon soils with a relatively low pH; 2) Complexation of heavy metals with DOM will promote the mobility and availability of heavy metals in soil-plant systems, especially in high organic matter soils (Kalbitz and Wennrich, 1998), while biochar can effectively retain these heavy metals (Li et al., 2018), limiting the uptake of metals by plant roots.

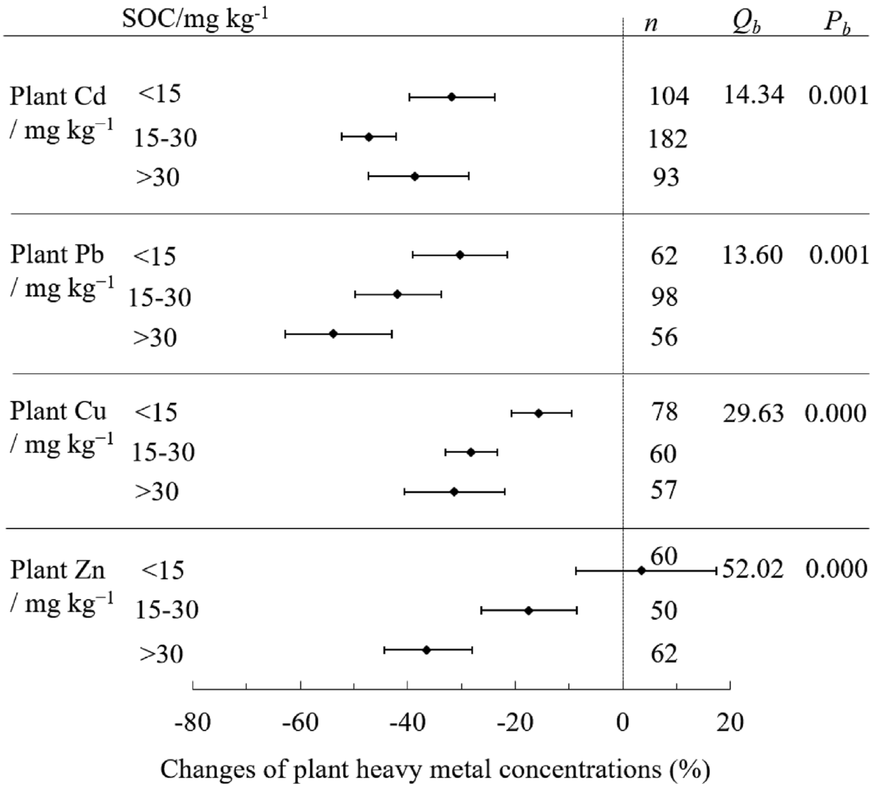

Fig. 4. Effect of biochar addition on plant heavy metal concentrations in soils having different SOC levels. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

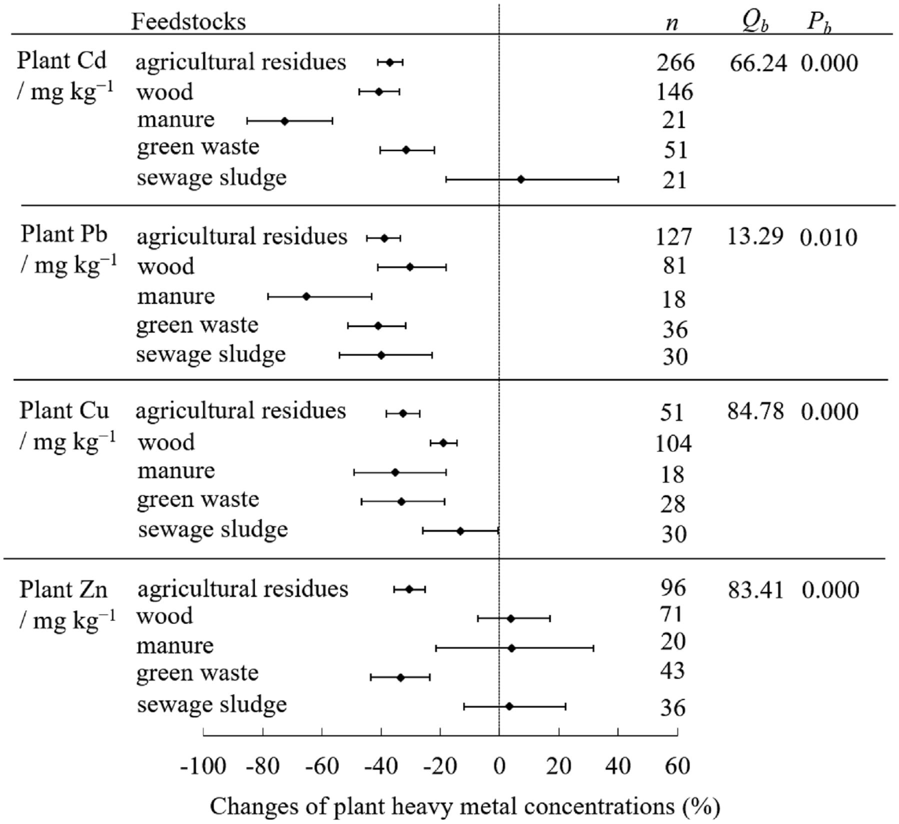

Fig. 5. Effect of different types of biochar addition on plant heavy metal concentrations. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

3.3. Effects of biochar variations on plant heavy metal uptake

Biochars from different feedstocks varied significantly in their capacity to reduce plant heavy metal concentrations (Fig. 5). Generally, the meta-analysis showed that biochars from different feedstocks effectively reduced Cd, Pb and Cu concentrations in plants (except for Cd when sewage sludge biochar was applied). The application of manure biochar resulted in the greatest reduction of plant Cd concentration (73%), which is superior to the other types of biochars evaluated. Agricultural residue biochar, wood biochar, and green waste biochar were also effective for decreasing plant Cd uptake (32–41%). In contrast, there was no significant change in plant Cd concentrations grown in sewage sludge biochar amended soils. Animal manure derived biochar reduced plant Pb concentration by 65%, wood biochar reduced Pb concentrations by 30%, whereas, the effects of other biochars were intermediate (39–41%). Cu concentrations in plants were reduced by the application of agricultural residue biochar (33%), wood biochar (19%), manure biochar (35%), green waste biochar (33%), and sewage sludge biochar (13%), respectively, as compared to un-amended soil. Plant Zn concentrations were only reduced by agricultural residue biochar (30%) and green waste biochar (33%).

Manure derived biochars appeared to be the most effective for reducing Cd and Pb concentrations in plants. Possibly this may be attributed to the high concentrations of phosphorus in animal manures, which may form insoluble precipitates with heavy metals (Cao et al., 2009); In addition, it is possible that nutrient enriched manure biochar enhanced plant growth (Liu et al., 2013), thereby diluting metal concentrations in the plant biomass (Alburquerque et al., 2013; Park et al., 2011). Precautions should be taken with biochar products derived from sewage sludge that may contain high concentrations of Cd and increase the risk for plant uptake (Ahmad et al., 2014; Lu et al., 2012). Changes in mean plant Zn concentrations varied for different biochars (Fig. 5). Crop Zn accumulation was affected by the type of biochar, soil conditions and plant species (Gartler et al., 2013; Khan et al., 2013; Zheng et al., 2015). In a previous study, we showed that Zn concentrations in rice grain were not reduced in biochar amended soils, although bioavailable Zn concentrations were greatly reduced (Chen et al., 2016). As shown in Fig. S5, biochar having a Zn concentration $\leq 500\ \mathrm{mg\,kg^{-1}}$ can reduce plant tissue Zn concentration, while plant Zn concentrations are increased in soils amended with biochar having a Zn content $> 500\ \mathrm{mg\,kg^{-1}}$. Fig. S6 shows that plant Zn concentration was significantly increased when biochar was added to soils with a higher Zn content ($\geq 300\ \mathrm{mg\,kg^{-1}}$), while there was no significant change of plant Zn concentration when biochar was added to low Zn soils ($< 300\ \mathrm{mg\,kg^{-1}}$). Therefore, the changes of plant Zn concentration were the results of interactions from multiple factors after biochar application.

The changes of plant heavy metal concentrations in response to biochar could also be affected by the pyrolysis temperature used in the manufacturing process (Fig. S4). The meta-analysis shows that biochars produced at 450–500 °C are preferred for effectively reducing plant uptake of Cd, Pb, Cu, and Zn, and also are more economical with respect to biochar yield and energy costs. Low temperature derived biochar is rich in oxygen containing functional groups that can effectively complex with heavy metals (Suliman et al., 2016; Uchimiya et al., 2011), while high temperature derived biochars usually have higher pH values due to the increased content of alkali and ash (Yuan et al., 2011). A pyrolysis temperature between 450 and 500 °C may drive the formation of oxygen containing functional groups and alkali. It should be noted that for raw materials with substantial phosphorus content, the pyrolysis temperature should be low to keep phosphorus available to form phosphate and allow precipitation with Pb (Cao et al., 2009; Ding et al., 2014). The availability of phosphorus in biochar will decrease with increasing pyrolysis temperature because of the formation of insoluble minerals such as crandallite ($CaAl_3(OH)_5(PO_4)_2$) and wavellite ($Al_3(OH)_3(PO_4)_2 \cdot 5H_2O$) (Xu et al., 2016). Kim et al. (2013) also recommended that production of biochar at a pyrolysis temperature of 500 °C is ideal for heavy metal stabilization and energy conservation

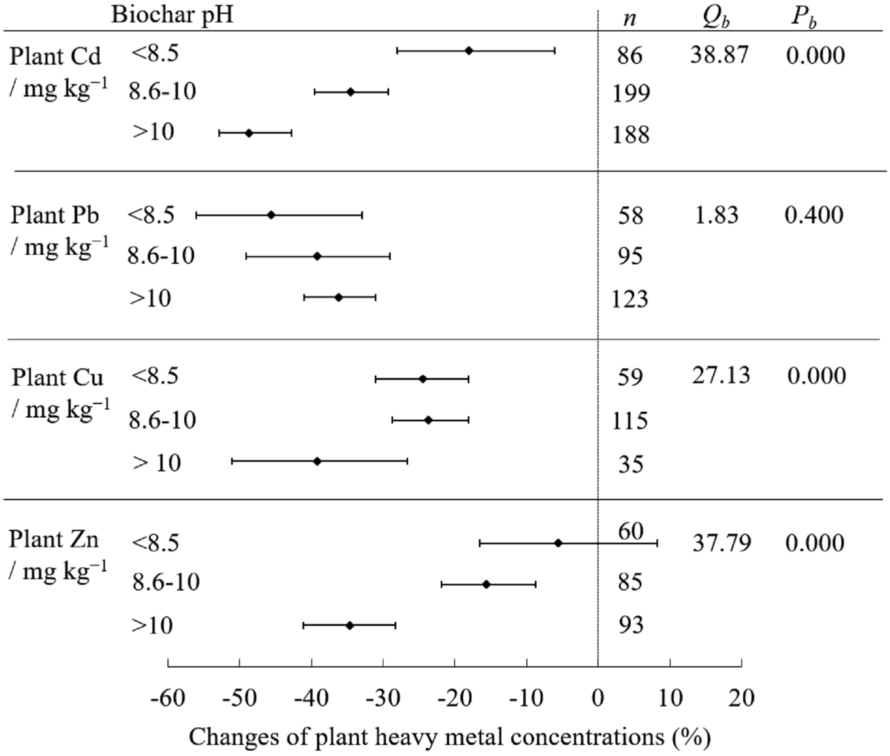

Fig. 6. Effect of biochar pH on plant heavy metal concentrations. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

compared to higher pyrolysis temperature, as pyrolysis is an energy-consuming process.

Differences in the pH of different biochar types had various effects on plant uptake of heavy metals (Fig. 6). Higher pH biochars resulted in greater reductions in plant Cd concentrations. Plant tissue Cd reductions were 18, 35, and 49%, respectively, for biochars grouped as having pH of < 8.5, 8.6–10, and > 10. The pH of the biochars likewise affected Pb and Cu concentrations in plants, while there were and were no differences among the three biochar pH groups for Cu and Pb. With respect to Cd, Cu, and Zn, the percentage reduction in plant uptake increased with increasing biochar pH. This suggests that the liming effect of biochar is a key consideration in remediation of heavy metal contaminated soils. As it is mentioned above, the possible mechanism lies that the alkaline biochar increased soil pH, which further promoted the transformation of heavy metal species to be more stable ones, and such transformation was affected by heavy metal properties.

As expected, the application rate of biochar significantly affected the percentage reductions in plant uptake of the heavy metals (Fig. 7). Biochar application at high levels (> 3%) resulted in the largest reductions in plant heavy metal concentrations. Plant Cd concentrations were reduced by 26, 38, and 51% at application rate of ≤1%, 1.1–3%, and > 3%, respectively. Likewise, plant Pb concentrations were reduced by 28, 37, and 50%, respectively at low, moderate, and high biochar application rates. The effect of application rate can be explained by the enhanced heavy metal retention and immobilization when more biochar was added and by biochar driven changes in soil chemistry.

3.4. *Effect of biochar on heavy metal concentrations in different plant types*

The reductions in plant heavy metal concentrations achieved with biochar application varied significantly for different types of plants (Fig. 8). On average, biochar reduced Cd concentrations in rice (40%), wheat (42%), maize (36%), vegetables (41%), grass (40%), and hyper accumulating plants (42%), respectively. The reduction in Cd concentrations in legumes was the least (21%) compared to other types of plants. Pb concentrations were reduced in crops in each of the eight categories, with the effect size ranging between 14 and 75%. The greatest decrease in Pb concentration occurred in maize, followed by vegetables, while the smallest decrease was in wheat. Cu concentrations were reduced by biochar in rice (32%), wheat (46%), maize (24%), vegetables (26%), and grass (15%). As for changes in plant Zn concentrations, biochar use led to declines only for vegetable crops, legumes, and hyper-accumulating plants, while it had no effects in reducing Zn concentrations in rice, wheat, maize and grasses. The latter plants are monocots in the family *Poaceae*, which produce phytosiderophores that are released into the rhizosphere to mobilize and transport iron and Zn (Reichman and Parker, 2005). Responses also varied among crops with respect to biochar effects on Cd uptake. The smallest effects of biochar on plant Cd concentrations were for legumes. This may be due to the rhizobia symbiosis with legume root, which promotes heavy metal uptake, and this partially offset the immobilization effect of biochar (Wei and Ma, 2010). Although this meta-analysis showed good effects on reducing heavy metal concentrations in plant tissues, it is also important to examine the absolute heavy metal concentrations of plants grown in biochar-treated soil as they may still exceed safe levels. Guidelines are needed for safe use of biochar to produce safe food crops.

While the main focus of most studies that were examined in this meta-analysis was to measure the extent to which biochar can reduce heavy metal uptake in plants, the biochar application rate must be sufficient to match and treat the amount of heavy metals that are contained in the soil (Figs. S5 and S6). In soils with relatively low heavy metal content, the application of biochar may result in lesser percentage decreases compared to those with higher heavy metal concentrations in soil. This may be because the availabilities of these heavy metals were already low in the lesser contaminated soils. If the biochar has a high content of heavy metals, it may also offset the immobilization effect normally associated with biochar. Generally, most of the biochars used in the investigated studies had low contents of heavy metals. However, standards such as those advocated by the International Biochar Initiative should be followed to restrict the application of heavy metal containing biochars and to reduce the risk when biochars are applied as soil amendment.

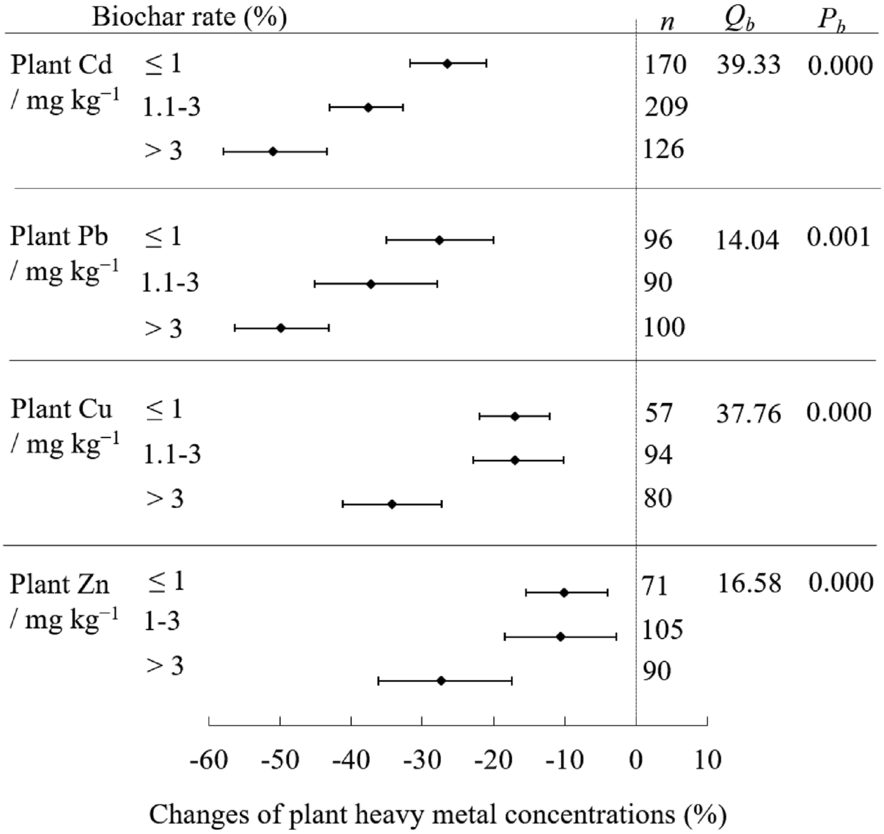

Fig. 7. Effect of application rate of biochar on plant heavy metal concentrations. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

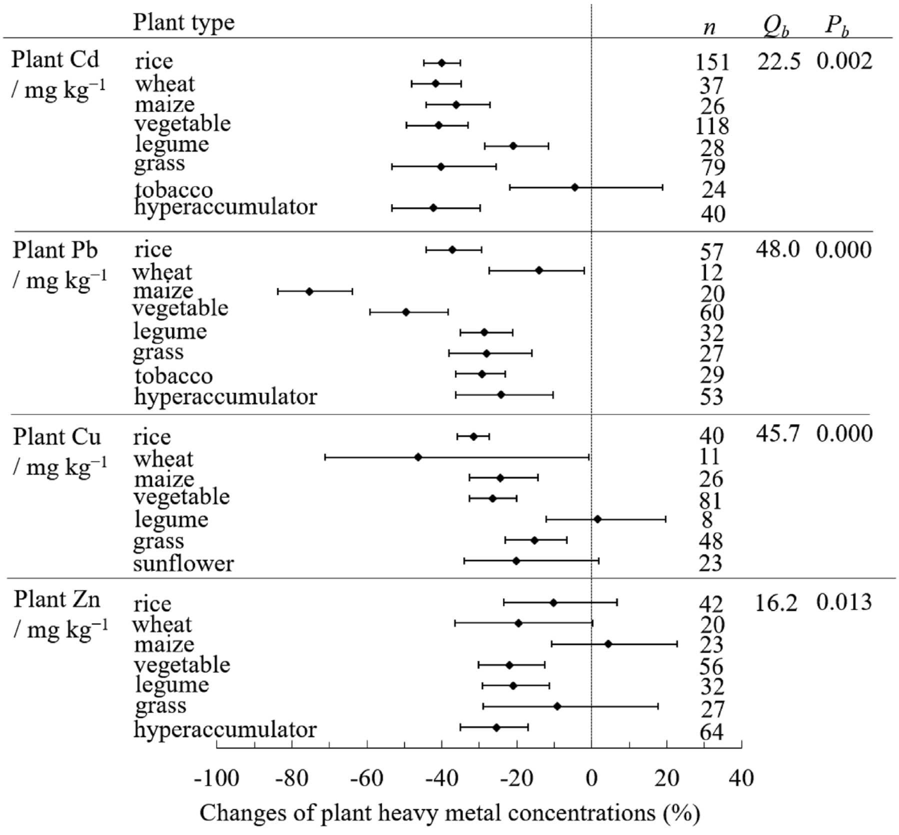

Fig. 8. Effect of biochar addition on heavy metal concentrations in different types of plants. Symbols represent mean effect sizes (percentage of change in plant heavy metal concentrations) with 95% bootstrap confidence intervals (CIs). *n* stands for sample sizes in each group. A significance test ($P < 0.05$) for between-group differences (P_b) of variables (Q_b) based on a permutation test (random effects design) was conducted.

4. Uncertainty analysis

The results of meta-analyses are constrained by the quality and quantity of the data collected from the published literature. The distribution of some variables in different groups may be unbalanced because of differences in research topic interest, variations in methods, and variations in research conditions. In those studies where the information such as experimental conditions, specific soil properties, and biochar properties were not given, this resulted in missing observations for some variables when sorted by groups. This will increase the uncertainties of meta-analysis. Various extraction methods were used as a characterization of available heavy metal contents in soils among the selected studies. The top 4 extractants were $CaCl_2$, DTPA, NH_4NO_3 and EDTA, which accounted for 21.6, 20.3, 10.8 and 5.4%, respectively, across the studies, with the available metal contents were not provided in some studies. As there was not a standard method in relation to normalization for these extraction methods, there was a possible uncertainty of the effect sizes of available metals in this meta-analysis. Zhang et al., 2013 found that $CaCl_2$ extractable Cd was significantly reduced, while EDTA extractable Cd was only reduced in low Cd contaminated soils treated by biochar. The meta-analysis shows that the magnitudes of biochar in field conditions were not as good as in pot conditions, while the latter one accounts for most (85%) of the studies. Cautions should be taken that the magnitudes of biochar in certain categories from this study may not accurately reflect practical amounts used in real world conditions.

5. Conclusions

This meta-analysis demonstrated biochar additions to soil resulted in overall reductions in average concentrations of Cd, Pb, Cu, and Zn in plant tissues by 38, 39, 25, and 17%, and simultaneous decreases in bioavailable metal concentrations in soils of 52, 46, 29, and 36%, respectively. The effect of biochar on heavy metal concentrations in plants varied depending on heavy metal speciation, soil chemical and physical properties, the type and application rate of biochar and its chemical properties, and on the crop type. Soil pH, texture, and organic matter contents were key variables determining the responses of plants to biochar. Understanding the responses to specific types of biochars for a particular crop and soil combination requires consideration of soil chemistry and the various factors that affect the bioavailability of metals. Meta-analysis provides a useful method for assessing the range of effects and predicting the efficacy of a particular biochar for stabilization of heavy metals in different soils and crop systems.

Acknowledgement

This work was financially supported by the project of "Source Identification and Contamination Characteristics of Heavy Metals in Agricultural Land and Products" (2016YFD0800300-6), the National Key Research and Development Program of China; the National Nonprofit Program by Ministry of Agriculture of China (201303095-11); and Jiangsu Collaborative Innovation Center for Solid Organic Waste Resource Utilization.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2018.05.004.

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