REVIEW

Key factors determining biochar sorption capacity for metal contaminants: a literature synthesis

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Abstract

The sorption capacity and affinity of biochar for metals are both important attributes that determine biochar's suitability as a soil amendment for contaminant mitigation, yet few analyses have considered both characteristics simultaneously. We present a systematic review of literature published between 2010 and 2018 to test the hypothesis that sorption capacity and affinity are affected by biochar properties, attributes of the metal contaminant, and experimental conditions, in that order. We used random forest (RF) and multi-objective optimization to analyze data of 559 individual Langmuir adsorption isotherms extracted from 133 studies covering the sorption capacity (C_{max}) and affinity (K_L) of biochar for 17 different metals, elaborated from six different feedstock classes, three different types of feedstock pretreatment, and five types of post-pyrolysis treatment. Highest sorption values were obtained for Pb(II), Cr(IV), and Cd(II). The feedstock used was the key determinant influencing biochar's capacity and affinity to sorb metal contaminants (first and fourth most important variable in RF models for C_{max} and K_L, respectively) with best results obtained for biochar elaborated from nutrient-dense feedstocks (animal biowaste, sludge, and manure). Biochars that had both high sorption capacity and affinity were the result of a longer duration of pyrolysis; they had lower C and higher N and O content, as well as lower C/N and higher O/C and H/C ratios, higher pore volumes and higher pH. Applying some form of pretreatment was better than none, whereas chemical modification was the best of the post-treatment methods analyzed. This review demonstrates clearly that multiple parameters during the preparation process influence the effectiveness of biochar to immobilize metal contaminants. Future research that focuses on mechanisms and the underlying factors for the correlations observed should allow the development of biochar formulations that are even more effective at immobilizing metal contaminants in soils and sediments.

Article Highlights

- **Sorption properties of biochar for heavy metals was assessed through a literature review using random forest and multi-objective optimization analyses**
- **Feedstock was the most important variable determining sorption capacity and affinity, with best results obtained for nutrient-dense feedstocks (animal biowaste, sludge, and manure)**
- **The best performing biochar had lower C and higher N and O content, as well as lower C/N and higher O/C and H/C ratios, higher pore volumes, and higher pH.**
- **Post-pyrolysis chemical treatment of biochar increased sorption properties more effectively than washing and magnetization.**

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1 Introduction

The contamination of soils with metals and other inorganic contaminants is a global concern (FAO and ITPS 2015; Friberg and Vahter 1983; Selin et al. 2018), resulting in crop contamination and posing serious threats to human health and our ability to reach sustainable development goals (Montanarella et al. 2016). In addition to reducing sources of new environmental contamination, it is also important to develop and improve strategies for the remediation of already contaminated soils. While soil remediation practices have traditionally relied on the use of organic matter (Alloway 2013; Bradl 2004; Wuana and Okieimen 2014), biochar, produced by burning biomass via pyrolysis, has been gaining interest due to its potentially superior ability to sorb metals (Ahmad et al. 2013; Borchard et al. 2012; Joseph et al. 2010).

Four main mechanisms for biochar's ability to sorb metals have been proposed: (1) electrostatic interactions between metallic ions and the charged biochar surface, (2) complexation or ionic exchange between ionizable protons on the surface of biochar and metallic ions, (3) sorptive interactions involving the delocalized π-electrons of biochar, and (4) the porous nature of biochar which may favor sorption of metals (Borchard et al. 2012; Dong et al. 2014; Vithanage et al. 2017). Regardless of the mechanisms involved, the ultimate suitability of biochar as a means to remediate metal-contaminated soils will depend on both the biochar's affinity (ability to attract) and its capacity to absorb metals. These attributes are described by the Langmuir adsorption constant (K_L) and maximum adsorption capacity (C_{max}), respectively (Volesky and Holan 1995).

The sorption capacity (C_{max}) and affinity (K_L) for metals may be influenced by multiple factors such as the range of feedstocks and the technologies used to manufacture biochar. Pre- and post-pyrolysis modifications of biochar may also enhance sorption properties. It is thought that such modifications may positively affect surface area, surface charge, functional groups on biochar surfaces, and pore volume, and/or improve pore size distribution in biochar (Rajapaksha et al. 2016; Sizmur et al. 2017). Approaches used so far to modify biochar surfaces include: (1) washing with water (Inyang et al. 2011) or acids (Park et al. 2013; Xu et al. 2014), (2) chemical and physical activation (Angin et al. 2013; Ippolito et al. 2012; Park et al. 2003), (3) chemical modification (Betts et al. 2013; Qian et al. 2013), and (4) magnetic polarization (Zhang et al. 2013a, b). However, despite the growing interest and understanding of the underlying mechanisms, available knowledge to predict and

influence biochar's potential for metal immobilization is still very limited.

Drawing on experimental data extracted from 133 peer-reviewed publications, we carried out an analysis to unravel how the affinity and capacity of biochar to sorb metals is influenced by (1) the processes used to prepare and modify the biochar (e.g., pyrolysis duration and maximum temperature, and pre- and post-treatment), (2) the characteristics of the biochar (e.g., feedstock type, elemental ratio, pH, and pore characteristics), (3) the characteristics of the metal contaminant (e.g., availability and ionic charge), and (4) the experimental conditions (e.g., pH, contact time, and buffer solution).

2 Material and methods

An exhaustive literature search was conducted focussing on peer-reviewed articles published between January 1st in 2010 and December 31st in 2018 using the Web of Science database (Thomson Reuters) using the term "biochar" in the "topic' field. Of the articles retained, only those that presented the results from batch experiments conducted to assess adsorption capacity and affinity of metals on biochar were selected ($n = 133$). These articles reported on a total of 559 individual Langmuir adsorption isotherms which is the commonly chosen model used to study biochar sorption of metals (see Appendix 1). The biochars were categorized into one of six types based on the feedstock used (see Table 1), and studies were grouped by metal contaminant ($n = 17$). We used the Langmuir equation parameters K_L and C_{max} as dependent variables in our analysis, because the equation has proven useful for describing natural systems where rates are low (e.g., limited sorption capacity) as is assumed for tested biochar-soil solution systems (Limousin et al. 2007; Vandenbruwane et al. 2007). K_L is a measure of affinity or how strongly the biochar attracts metals (L g^{-1}) and C_{max} is the maximum adsorption capacity of the biochar for the metals (g kg^{-1}). C_{max} and K_L were logarithmically transformed prior to analysis to reduce the influence of outliers (Reid 2003). Because our objective was to predict adsorption of metals on biochar, we collated the information shown in Table 1 from the source articles for use as independent variables.

Meta analytical statistical approaches have dominated attempts to quantitatively synthesize data. These statistics use mean values and error measurements to obtain robust measurements of effect size, but provide little information on the interaction of the independent variables being studied (Jeffery et al. 2011). In addition, for meta-analytical

Table 1 Metadata of variables used in the analyses

	Variable	Description
Independent/response variables	C_{max}, (g/kg)	The maximum adsorbate consumed in the given conditions (Volesky and Holan 1995)
	K_L, (L/g)	The coefficient related to the affinity between the adsorbent and adsorbate (Volesky and Holan 1995). This is a measure of the strength with which a biochar may sequester a contaminant, stickiness is less technical language
Biochar properties used as dependent variables in the analysis	Feedstock	We used six categories (animal biowaste, plant-based food industry waste, non-woody plant residues, woody plant residues, manure, and sludge) (a) Woody plant residues: Plant residues with high C:N ratios and high lignin content (b) Non-woody plant residues: Plant residues with low C:N ratio, and lignin content (c) Plant-based food industry waste: Waste from food processing plants, i.e., peels, seeds, etc. (d) Animal biowaste: bone, meat, blood, etc. (e) Manure: any type of manure (f) Sludge: any sludge, usually obtained from sedimentation ponds linked to waste water treatment but also including brewery sludge
	Max temp, (°C)	The maximum temperature during the pyrolysis process
	Duration, hours	The duration of the pyrolysis process
	Pre-treatment	Any treatment applied to the feedstock before pyrolysis: Chemical modification (The feedstock was composted, anaerobically digested, or modified by adding: $KMnO_4$, $AlCl_3$, KOH, NaOH, Fe_2O_3, flocculating agent, graphene, Na_3PO_4, H_3PO_4, or H_2O_2) H_2O washed (washed with water or deionized water) None (If there was no treatment mentioned)
	Post-treatment	Any treatment applied to the biochar after pyrolysis: Activation (the biochar was activated with KOH, H_2O_2, HNO_3, NH_3, steam, sonication, or other physical or chemical activators) Chemical modification (the biochar was modified by acidification, oxidation, Al, Br, Ca, P, Mg, Mn, Na, N, Zn, chitosan, gelatin, H_2O_2, KOH, NaOH, HNO_3, or $KMnO_4$) Magnetization (the biochar was magnetized with a treatment of Fe or Ni/Mn) Washing (the biochar was washed with water, deionized water, HCl, and CH_2Cl_2) None (if there was no treatment mentioned)
	Ash content (%)	The percentage ash of the biochar
	C (%)	The percentage carbon of the biochar
	H (%)	The percentage hydrogen of the biochar
	N (%)	The percentage nitrogen of the biochar
	O (%)	The percentage oxygen of the biochar
	O/C	The atomic ratio between oxygen and carbon found in the biochar
	H/C	The atomic ratio between hydrogen and carbon found in the biochar
	C/N	The mass ratio between carbon and nitrogen found in the biochar
	Pore diameter (nm)	The diameter of the pores found in the biochar
	Total pore volume (cm^3/g or m^3/t)	The volume of pores found in the biochar
	CEC (cmol/kg)	Cation-exchange capacity—capacity to sorb cations
	Surface area, (m^2/g N_2)	The surface area of the biochar; this is characterized by the Brunauer–Emmett–Teller (BET) method (Brunauer et al. 1938)
	pH (biochar)	The pH of the biochar

Table 1 (continued)

	Variable	Description
Attributes of the metals used as dependent variables in the analysis across all metal types	Metals	The species of metals used in the adsorption experiments (Ag, Al, As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb, U, and Zn)
	Anion/cation	Shows if the metal used has a positive (cations) or negative charge (anions)
	Goldschmidt classification	The geochemical behavior (of the metal studied) separates the chemical nature of the metal according to the classification of Goldschmidt. This classification is based on the formation of oxides and sulfides, and provides a view of the affinities/associations between elements. It also allows us to know which cations are more likely to enter spaces in the particles of mineral structures. This is determined based on the ionic radius and the ionic charge (Goldschmidt 1954; Scott and Pain 2019)
	Geochemical availability	This variable is a measure of the pH which causes a dissolution or maximum mobility of the metal being considered (Sarkar and Taylor 2010)
	Ionic radius Angström	The ionic radius is the distance between the nucleus and the electron in the outermost shell of an ion
	Ionic charge	The electrical charge of an ion, created by the gain (negative charge) or loss (positive charge) of one or more electrons from an atom or group of atoms
	Ionic potential	Relation between ionic radius and its charge. This measure allows us to know how strong or weak the ion will be electrostatically attracted to ions of opposite charge (Railsback 2015)
	Molar mass	A physical property defined as the mass of a given substance divided by the amount of substance
Experimental conditions used as dependent variables in the analyses	Normality	The normality of a solution is the gram equivalent weight of a solute per liter of solution
	Background solution	Solution where the adsorption experiment takes place. The solutions were categorized as deionized water (DI), mono-valent (e.g., $NaNO_3$), and di-valent (e.g. $CaCl_2$)
	pH (solution)	The pH of the solution is a categorical variable related to the solution used for the adsorption experiment. The solutions were $CaCl_2$, H_2O, or KCl
	Contact time (h)	The time in which the biochar was in contact with the metal for the adsorption process

statistics, there are no clear protocols for combining more than one dependent variable. In this synthesis, we have, therefore, endeavored to overcome these limitations using random forest analysis together with multi-objective optimization analysis, following the precedent set by Crane-Droesch et al. (2013) who synthesized biochar results via regression.

We developed random forest (RF) models using the *cforest* function in the *party* package for R (Strobl et al. 2007) to assess relationships between C_{max} and K_L as response variables and the explanatory variables shown in Table 1. Variable importance values were calculated with the *varimp* function and their variability was quantified by developing 20 RF models based on random selections of 80% of cases. Importance values were based on the mean decrease in model accuracy and were standardized across runs by dividing by the value of the most important variable. As a final step, the RF models were used to generate predicted values of both C_{max} and K_L values which were then plotted against the measured values reported in the source publications to give an indication of the accuracy (R^2) of the RF predictions. RF models were run for all metals combined and for specific metals when sufficient measurements (sample size) where obtainable from the literature for meaningful statistical analyses (i.e., for Cd, Pb, and Cu).

Given that both high affinity and capacity are important qualifiers of biochar sorption ability, we additionally carried out straightforward multi-objective optimization analyses by comparing attributes of biochar preparation properties between (1) study cases whose C_{max} and K_L scores were both above the respective median values (target group) and (2) all other cases (other group). Given that studies did not consistently report on the same biochar preparation properties, it was not possible to perform a multi-objective multivariate modeling exercise. Comparisons were, therefore, largely based on the use of *t* tests.
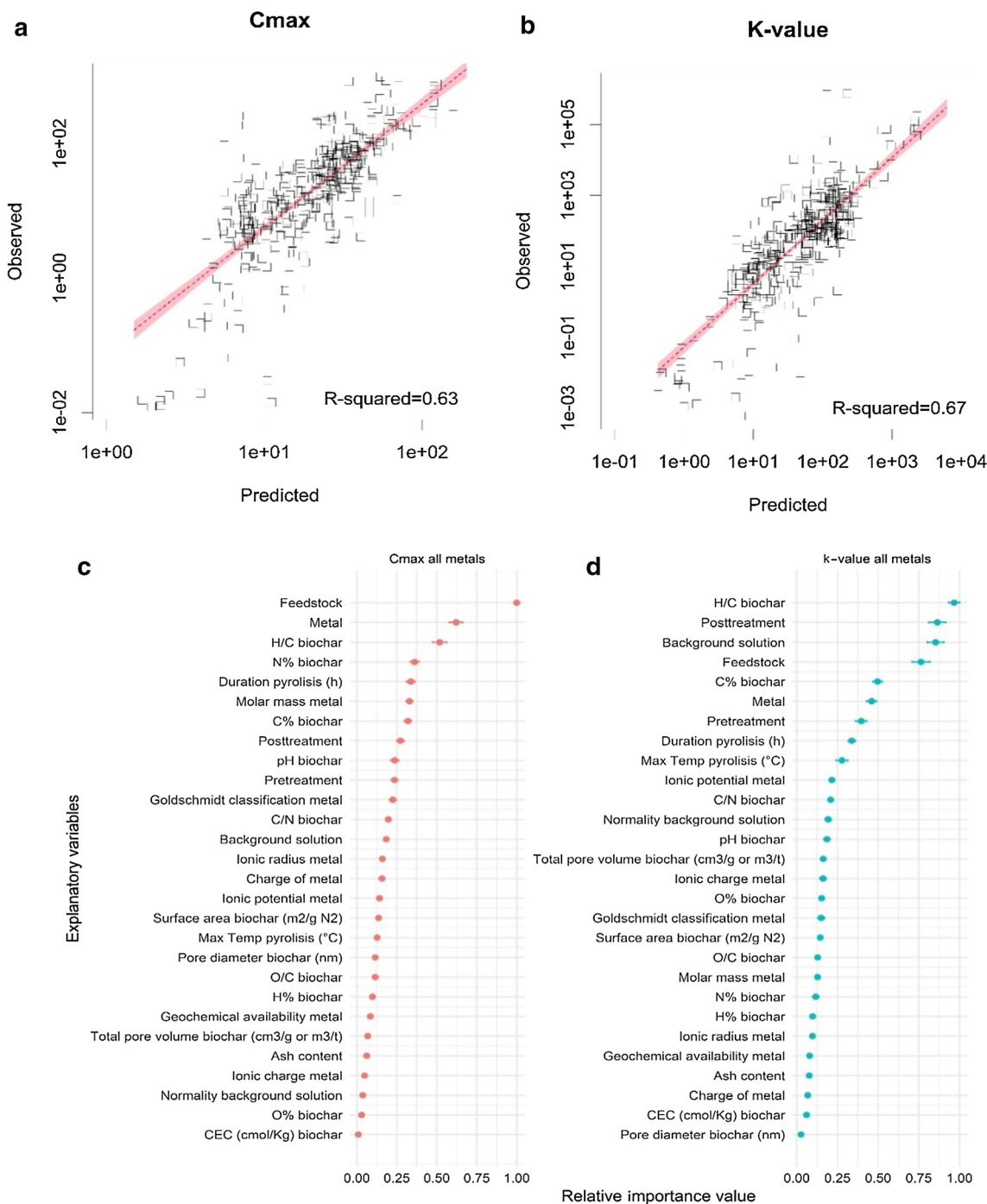
Fig. 1 **a** The correlation between predictions of C_{max} derived from the Random Forest model vs. values reported in the peer-reviewed literature ($R^2 = 0.63$). **b** The correlation between Random Forest K_L predictions and measured values ($R^2 = 0.67$). **c** The most important independent variables for C_{max} prediction. **d** The most important variables for K_L prediction. See Table 1 for detailed description of the independent variables used in the analyses, and Appendix S1 for raw data

3 Results

The RF model for C_{max} produced an $R^2 = 0.63$ (Fig. 1a) and the most important predictor variables were: (1) the feedstock used to make the biochar, (2) the metal contaminant under investigation, (3) the hydrogen-to-carbon atom mass ratio of biochar, and (4) the nitrogen content of the biochar (Fig. 1b). The RF model for K_L produced an R^2 of 0.67 (Fig. 1c), the most important predictor variables being: (1) the hydrogen-to-carbon ratio of the biochar, (2) the
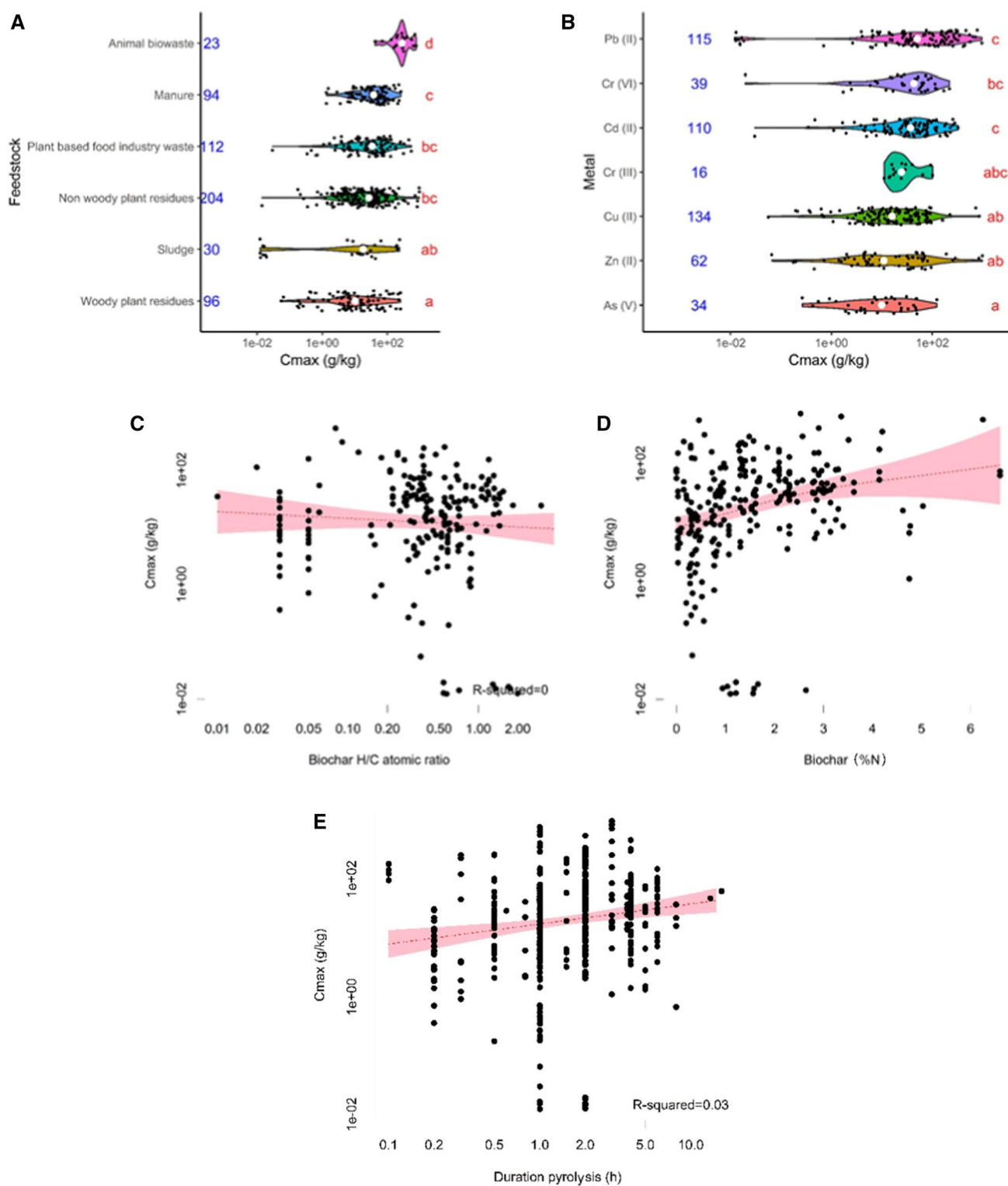
Fig. 2 Univariate plots of the five most important variables for C_{max} prediction across all metal species

background solution used in the sorption experiment, (3) post-treatment of the biochar, and (4) feedstock (Fig. 1d). The RF analyses done for individual metal contaminants (Cu, Pb, and Cd) produced results very similar to those obtained when all metal contaminants were analyzed simultaneously (see Appendix S2).
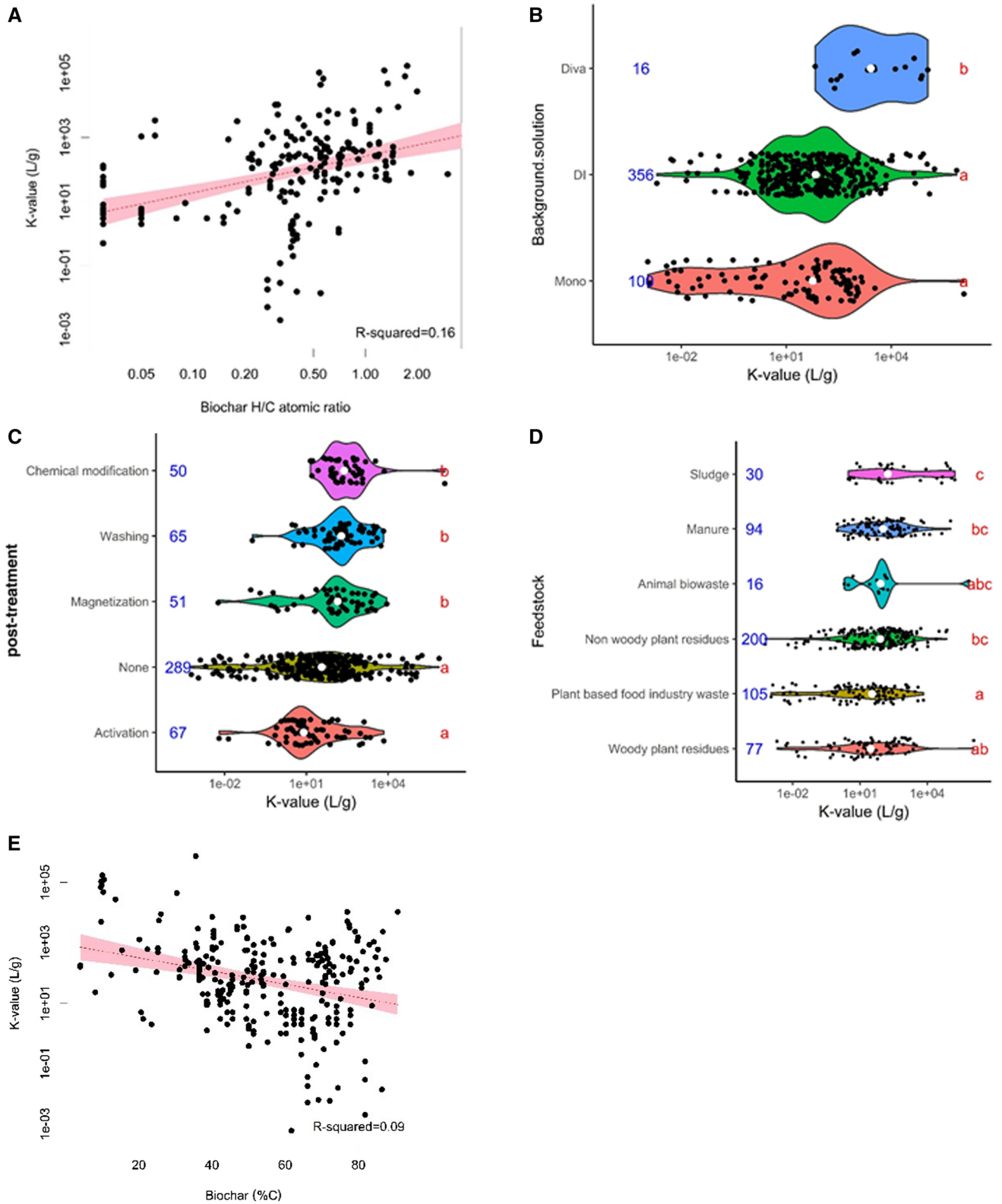
Fig. 3 Univariate plots of the five most important variables for K_L prediction across all metal species
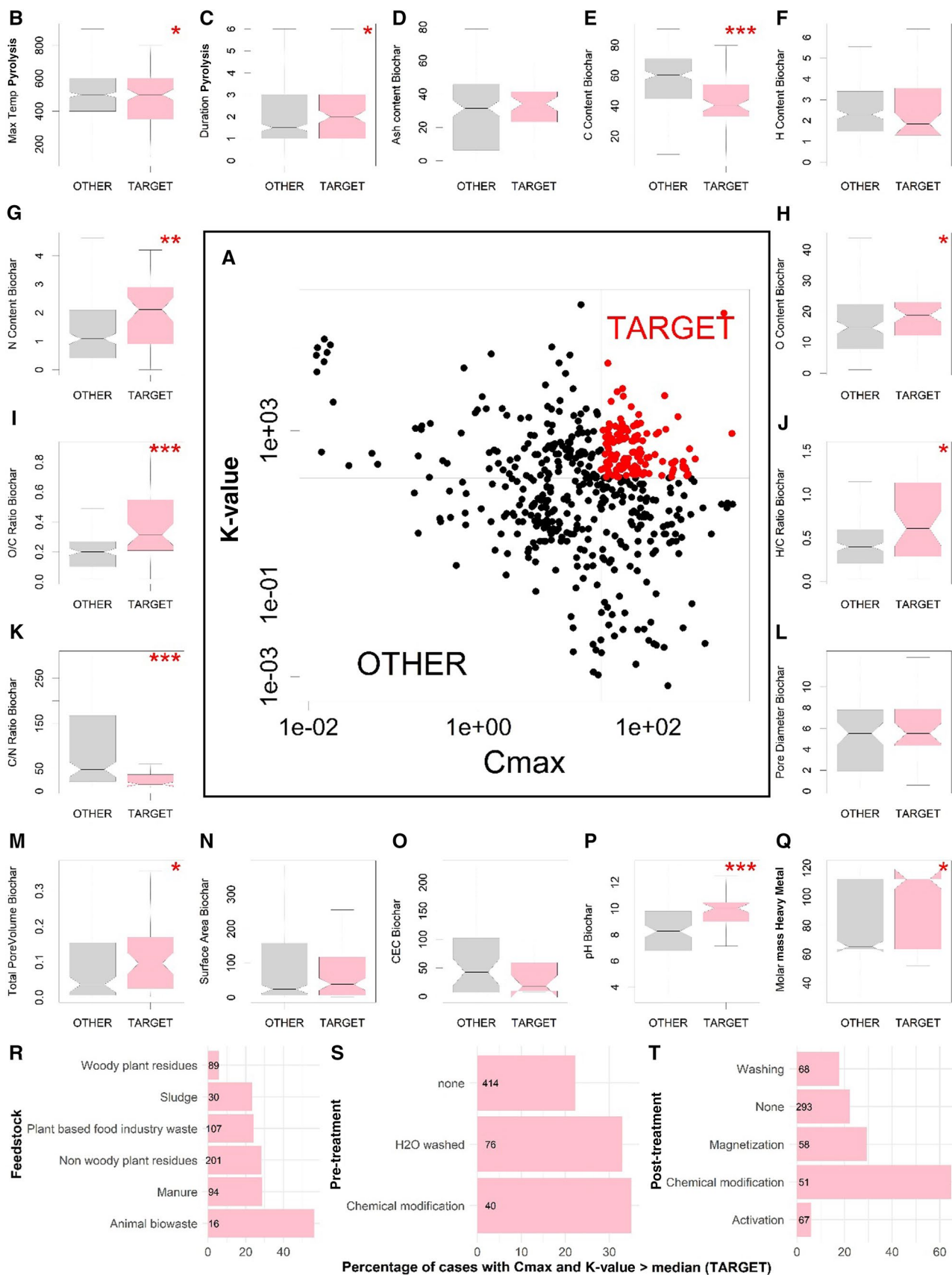
◀ **Fig. 4** Multi-objective optimization analyses comparing attributes of biochar preparation properties between (1) study cases whose C_{max} and K_L scores were both above the respective median values (TARGET group) and (2) all other cases (the OTHER group). In the central graph, the scatterplot, the red datapoints in the top right corner represent the data points that fall within the >50% quantile for both variables (marked as TARGET group). The box plots on the left, right, and top margins of the figure compare the values of cases in the TARGET group versus the OTHER group for continuous predictor variables. The horizontal histograms at the bottom margin of the figure demonstrate, for categorical predictor variables, the percentage (x-axis) and number (black numbers on the bars) of cases for each independent category where the relevant observations fell within the >50% quantile

One of the striking aspects of the data is the high variability reported by different studies (Figs. 2, 3). Nevertheless, it was possible to unveil some general trends: (1) biochar made from animal biowaste and manure seems to sequester metals more effectively than biochar made from woody plant residues, (2) biochar showed a better adsorption capacity for Pb(II) and Cd(II) than for As(V) and Zn(II), (3) C_{max} values decrease and K_L values increase as the hydrogen/carbon ratio of the biochar increases, (4) C_{max} values increase as the nitrogen content of biochar increases, (5) C_{max} and K_L values decrease as the carbon content of the biochar increases, (6) K_L values were higher in experiments that used a divalent background solution than in experiments that use a mono-valent background solution or deionized water, (7) K_L values are higher for biochars that were chemically modified, washed, or magnetized following pyrolysis, and (8) the duration of the pyrolysis process positively correlates with the sorption ability of biochar.

Results of comparisons of biochar preparation properties between study cases with K_L and C_{max} values above (target group) and below (other group) the medians aligned very closely with the results obtained from the RF analyses (Fig. 4). The biochars that simultaneously maximized both C_{max} and K_L were generally made at lower maximum temperatures and longer duration of pyrolysis and from nutrient-dense feedstocks such as animal biowaste. They tended to have low C, and high N content, as well as a low C/N and a high O/C ratio. The effectiveness of post-pyrolysis chemical treatment of biochar for metal sequestration was likewise borne out, with more than 60% of the biochars that received post-pyrolysis chemical treatment falling in the upper 50% quantiles area. Finally, analysis also suggests that alkaline biochars are best for simultaneously maximizing C_{max} and K_L (Fig. 4). Similar trends were found for individual metal contaminants (Cu, Pb, and Cd) (Table 2, appendix S2), although not all comparisons were statistically significant at $P = 0.05$ partly due to small-sample sizes.

4 Discussion

Our results demonstrate that the sorption capacity of biochar for metals is in the range of other commonly used soil amendments that are usually more expensive than biochar (e.g., activated carbon; Table 3) (Ng et al. 2003). The best performing biochars were made from feedstocks with high nutrient levels (e.g., animal biowaste or manure) and had high aromaticity (i.e., high oxygen-to-carbon ratio) favoring electrostatic sorption of metals (Ahmad et al. 2014, 2018; Harvey et al. 2012). However, due to the number of studies that were used, for example, animal biowaste was limited ($n = 23$), this result needs to be interpreted with caution. On the other hand, we also found a positive relationship between biochar sorption capacity and nitrogen content which also hints at the fact that biochars made from animal-based biomasses with high nutrient content (e.g., potassium and nitrogen) may sorb metals most effectively. The biochars with the lowest capacity to sorb metals were made from wood and these had a lower C_{max} than compost, which may be related to the fact that they had a higher C content, which was inversely correlated with biochar sorption ability. This adds further support to the idea that stoichiometric nutrient composition (e.g., N:K) and nutrient density of feedstock used to produce biochar is critical to the ability to sequester metals.

Several authors have suggested the modification or bespoking of biochar to enhance their sorption capacity and affinity (Huang 2019; Wang and Liu 2018). The results obtained here strongly support this growing consensus. Post-pyrolysis chemical modifications of biochar such as washing and magnetization increased the probability that a biochar will possess high values of both C_{max} and K_L. Washing biochar with water or acids post-pyrolysis is often done to neutralize pH and remove alkaline elements such as ash and soluble salts (Uchimiya et al. 2010; Yang et al. 2004; Zhang et al. 2013a, b). Our results indicate that the effectiveness of such washing is unlikely to be a pH effect, since alkaline biochars were more likely to be in the upper 50% quantile for simultaneous maximization of both C_{max} and K_L. More plausible explanations could be that post-pyrolysis chemical treatment may augment the effective surface area, or because of chemical reactions that induce the formation of oxygen-containing functional groups on biochar surfaces, but further study is needed to unravel the mechanistic basis of these correlations.

Even when we consider biochar manufactured from a single feedstock, the variance in the ability to sequester metals is significant, i.e., C_{max} values ranged from 0.01 to 980.39 mg g^{-1} for biochars made from non-wood plant residues. This demonstrates that we still need additional studies to be able to make clear recommendations about

Table 2 Results of comparisons of biochar preparation variables between cases with both K_L and C_{max} above the medians of all values (TARGET) and all other study cases (OTHER), as in Fig. 4 and appendix S2

	All metals	Cu	Pb	Cd
Maximum temperature pyrolysis	↓			↓
Duration pyrolysis	↑	↑↑↑		
Ash content biochar				
C content biochar	↓↓↓	↓↓	↓	(↓)
H content biochar				
N content biochar	↑	↑↑↑		(↑)
O content biochar	↑↑			
O/C ratio biochar	↑↑↑	↑	↑↑	(↑)
H/C ratio biochar	↑	↑↑		
C/N ratio biochar	↓↓↓	↓↓↓	↓	↓↓
Pore diameter biochar		↑		
Total pore volume biochar	↑	↑↑		
Surface area biochar				
CEC biochar			(↓)	
pH biochar	↑↑↑	(↑)	↑↑↑	(↑)
Molar mass metal	↑			
Feedstock	Animal biowaste, manure, non-plant residues	Sludge, manure	Animal biowaste, sludge, manure	Animal biowaste, manure
Pre-treatment	Chemical modification or H$_2$O washed	H$_2$O washed	H$_2$O washed	Chemical modification
Post-treatment	Chemical modification, magnetization	Chemical modification	Chemical modification, magnetization	Chemical modification

Results are presented for study cases covering all metals, and study cases limited to Cu, Pb, and Cd sorption experiments. Arrows indicate the nature of variable scores in the target group as compared to the other group: ↓ and ↑ mean that TARGET group scores are lower and higher than OTHERs group scores, respectively. The number of arrows is indicative of the significance levels: three arrows $P < 0.001$; two arrows $P < 0.01$; one arrow $P < 0.05$ (*t* tests). Arrows between brackets indicate clear differences which are nonetheless not significant at $P < 0.05$

biochar manufacture for specific purposes. Variables such as cation-exchange capacity are rarely reported and probably should be included more frequently in metal adsorption studies (Shackley and Sohi 2010; Shen et al. 2017). Finally, the results analyzed here from lab studies, together with theoretical studies, are providing important clues as to how we can best optimize biochar, so that it is fit for purpose, in this specific case, for the immobilization of metals (Dieguez-Alonso et al. 2019; Hagemann et al. 2017; Joseph et al. 2018). However, these conclusions need to be tested in the field where variations in water availability, temperatures, soil mineralogy, etc. may influence the effectiveness of biochar in addressing different challenges (Table 3).

The rapid growth in biochar research and funding witnessed over the last decade is a clear indication that both scientists and natural resource managers see potential in biochar for addressing environmental challenges. An especially attractive aspect of biochar is its potential to offset significant amounts of anthropogenic greenhouse-gas emission (Werner et al. 2018; Woolf et al. 2010), and to facilitate efforts to achieve a more circular economy, in which waste streams are reutilized to support production (Carus and Dammer 2018). However, for biochar to help in climate mitigation and contribute to the resolution of environmental problems more generally, biochar needs to be used at scale. To date, the economic value proposition of biochar has not been clear (Bach et al. 2017). While our results clearly demonstrate the need for future biochar research to address chemo-technical engineering challenges, demonstrating that biochar can immobilize metals, it is an important step forward in providing the motivation needed to fast forward research that would allow for scaling up. To conclude, the collective progress inherent in the data compiled here demonstrates that biochar has the potential to play an important role in remediation of contaminated soils and to help with the move towards a more circular economy.

Table 3 (a) C_{max} values of biochar and compost obtained by analyzing the peer reviewed literature, and (b) C_{max} values of three commonly used amendments soil remediation taken from empirical studies

Amendment	C_{max} (g kg^{-1})	Standard deviation	Metal	Source
(a) Estimates of absorption capacity of soil amendments obtained from literature analysis				
Biochar	65.1 (mean value)	120.01		This study (Appendix S1)
Biochar (animal biowaste)	317 (mean value)	189.62		This study
Biochar (wood)	33 (mean value)	55.77		This study
Compost	43.27 (mean value)	133.76		Anastopoulos and Kyzas (2015)
(b) Estimates of absorption capacity of soil amendments from individual empirical studies				
Activated carbon (heated with H$_2$S)	230		Hg(II)	Gomez-Serrano et al. (1998)
Activated carbon (modified with tetrabutyl ammonium)	9.9		Cu(II)	Monser and Adhoum (2002)
Granular-activated carbon	20		Zn(II)	Álvarez-Merino et al. (2005)
Chitosan	815		Hg(II)	Mckay and Blair (1989)
Chitosan	273		Cr(VI)	Udaybhaskar et al. (1990)
Sludge	560		Hg(II)	Srivastava et al. (1989)
Sludge	95		Zn(II)	Srivastava et al. (1994)
Sludge	1865		Pb(II)	Srivastava et al. (1994)

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