pH effects of the addition of three biochars to acidic Indonesian mineral soils

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Abstract

Soil acidity may severely reduce crop production. Biochar (BC) may increase soil pH and cation exchange capacity (CEC) but reported effects differ substantially. In a systematic approach, using a standardized protocol on a uniquely large number set of 31 acidic soils, we quantified the effect of increasing amounts (0-30%; w:w) of three types field-produced BC's (from cacao shell, oil palm shell and rice husk) on soil pH and CEC. Soils were sampled from croplands at Java, Sumatra and Kalimantan, Indonesia. All BCs caused a significant increase in mean soil pH with a stronger response and a greater maximum increase for the cacao shell BC addition, due to a greater acid neutralizing capacity (ANC) and larger amounts of extractable base cations. At 1% BC addition, corresponding to about 30 tons ha^{-1}, the estimated increase in soil pH from the initial mean pH of 4.7 was about 0.5 units for the cacao shell BC, whereas this was only 0.05 and 0.04 units for the oil palm shell and rice husk BC, respectively. Besides on BC type, the increase in soil pH upon the addition of each of the three BCs was mainly dependent on soil CEC (low CEC resulting in stronger pH increase), and to a lesser extent on initial soil pH (higher initial pH resulting in stronger pH increase). Addition of BC also increased the amount of exchangeable base cations (cacao shell >> oil palm and rice husk) and CEC. Through this systematic screening of the effect of BC on pH and CEC of acidic soils, we show that small addition of BC, in particular if made of cacao shell, to acidic agricultural soils increases soil pH and CEC. However, the response is highly dependent on type, quality and amount of the added BC as well as on intrinsic soil properties, mainly CEC.

Key words: Biochar; pH; soil; CEC; Indonesia.

1. Introduction

Biochar (BC) is the solid product formed after organic matter is charred via pyrolysis, i.e. without access to oxygen and at high temperature (250-900°C). Depending on its intrinsic properties and recalcitrance (Harvey *et al.*, 2012), BC may present a way of sequestering carbon in soils (Lehmann, 2007). In addition, BC can improve soil fertility (Atkinson *et al.*, 2010; Glaser *et al.*, 2002) and may serve as an attractive soil amendment for soils of low agricultural quality. Long-term use (100 to 1000 years) of BC as a soil amendment originates from the tropics: the *Terra preta* soils in the Amazonian forest have an improved fertility (Steiner *et al.*, 2007). It is known that the indigenous people added charcoal to these soils, and still today 100-1000 years later, these soils have enhanced physical and chemical properties due to the BC, compared to surrounding soils (Glaser *et al.*, 2001; Glaser *et al.*, 2002; Lehmann *et al.*, 2003; Neves *et al.*, 2003).

Biochar characteristics are strongly determined by source material and production procedure (Brewer *et al.*, 2011; Chun *et al.*, 2004; Jha *et al.*, 2010; Spokas *et al.*, 2012). The production temperature has been shown to have a profound effect on the C content, pH and CEC of the BC (Chen *et al.*, 2008). The feedstock of the BC has also been shown to be of importance for the liming capacity of the BC (Yuan and Xu, 2011; Yuan *et al.*, 2011). Upon mixing with BC, changes in soil pH are affected by CEC and levels of exchangeable acidity (acid saturation), which in turn depend on climatic conditions (leaching), mineralogy, clay content and amount and quality of soil organic matter (McBride, 1994; Ziadi and Sen Tran, 2007). Soils with high CEC and large acid saturation are well-buffered with respect to pH (Ziadi and Sen Tran, 2007). These factors combined with intrinsic BC characteristics may therefore be important in influencing how BC changes soil pH and fertility and thus BC's potential impacts on crop production (Spokas *et al.*, 2012).

Several studies have confirmed an increased crop production after BC amendment, although other studies found small or no effects (Jeffery *et al.*, 2011; Spokas *et al.*, 2012). The addition of BC to soil, in combination with fertilizer, has been reported to increase yield (Chan *et al.*, 2007; Asai *et al.*, 2009) or have no effect (Jeffery *et al.*, 2011). Yamato *et al.* (2006) experienced significantly increased yields of maize and peanut and attributed this to the increases in pH and CEC for Indonesian soils after the addition of BC of bark from *A. mangium*. In an acid Alfisol from NSW Australia, radish crop yields were significantly greater when BC was added in combination with fertilizer as compared to fertilizer only (Chan *et al.*, 2007). The positive effect of BC on crop production may be due to pH and CEC increases and changes in the physical properties of the soils, rather than to nutrients associated with the BC per sé (Chan *et al.*, 2007). Also, Asai *et al.* (2009) showed greater increases in rice yields in Laos, when fertilizer additions were combined with BC, as compared to the addition of fertilizer alone.

Increases in pH and CEC of acidic soils are commonly observed in response to BC amendments (Glaser *et al.*, 2002). However, most studies only included a limited number of soils (Alburquerque *et al.*, 2014; Atkinson *et al.*, 2010; Lehmann, 2007). In studies of pH-neutral soils from the USA Mid-west, Laird *et al.* (2010) showed only minor increases in both soil pH (<1 pH unit) and CEC (~3 $cmol_c$/kg) in response to 2% BC additions. Recently a meta-analysis (Jeffery *et al.*, 2011) revealed an overall positive effect of BC addition to soils on crop productivity with greatest effects in acidic and neutral pH soils with a coarse or medium texture. The main mechanisms were suggested to be liming effects and improved water holding capacity (Jeffery *et al.*, 2011). A significant increase in soil pH upon BC addition was also confirmed in a meta-analysis by Biederman & Harpole (2013), who used ~~underlying~~ a combination of several independent studies, which in contrast to our approach, involve a wide variation in protocol and applied techniques. The change in soil pH upon BC addition was related to the initial soil pH and the alkalinity of the BC. A systematic study of the effect of different types and doses of BC on such a large number of soils has to our knowledge not been conducted so far.

More than 50% of the agricultural soils in Indonesia are acidic (Uexküll and Mutert, 1995). Oxisols (USDA, Soil taxonomy) and Ultisols are common in the tropics and are characterized by low pH, low CEC and high contents of aluminium- ($Al(OH)_3$) and iron-hydroxides ($Fe(OH)_3$) (Van Wambeke, 1992). These features cause severe phosphorus (P) deficiency due to the strong sorption of PO_4^{3-} to oxide surfaces in the soils and the formation of insoluble iron (Fe) and aluminium (Al) phosphates (Cross and Schlesinger, 1995). At the same time, the low CEC of the ~~eroded~~ soils causes considerable leaching of nitrogen (N) making fertilization both inefficient and expensive (Chan *et al.*, 1993; Thomsen *et al.*, 1993). The low pH of these soils also results in elevated concentrations of Al. Dissolved Al in soils tends to increase exponentially to high values particularly at pH below 4.5 (Berggren and Mulder, 1995; Mulder *et al.*, 1989) and may reach levels toxic to plants (Kinraide, 2003).

We investigated the general effectiveness of three types of locally produced BC to acidic Indonesian soils in terms of changes in soil pH and CEC. In addition, we assessed if these changes can be directly related to initial soil characteristics and BC properties. According to our knowledge, this is the first study to systematically investigate effects of different BCs on selected soil chemical properties according to a standardized protocol, using a large number of soils (31). Most other studies focus on a limited number of soils only (Butnan *et al.*, 2015; Alburquerque *et al.*, 2014) or involve meta-analysis, where the underlying studies apply a range of techniques (Biederman & Harpole, 2013; Jeffery *et al.*, 2011). The large number of soils allowed us to draw conclusions on the effect of soil characteristics on the pH effect of the BCs.

In most earlier studies, laboratory-made BCs were used. Such biochars are mostly made in a muffle furnace or microwave oven. These approaches bear little relevance for a tropical rural situation where simple traditional kiln technologies are the norm. Thus here "real-world" BCs, actually made and used in field experiments using locally made pyrolysis units, were tested. The use of field-made biochars represents a realistic situation for small scale farmers because advanced BC production systems, microwave ovens and furnaces are unavailable in such situations (Spokas *et al.*, 2012).

Our main questions were: To what extent does BC increase in both soil pH (Q1) and CEC (Q2) with increasing concentrations of BC. How does the increase in soil pH depend on initial soil CEC and pH (Q3). How do changes in soil pH and/or CEC depend on BC type (Q4).

2. Material and methods

2.1. Site description and sampling

The samples were collected from 31 different sites (agricultural fields < 100 ha), from Java (site 1), Sumatra (site 2-22) and Kalimantan (site 23-31), Indonesia (Fig. 1 and Table 1). At each of the 31 sites, soil samples of 250-1000g from 10-15 points (depth 0-15 cm) were bulked, air dried at ± 35 ºC for 12 h and thoroughly mixed by hand. The sites were chosen to represent a wide range of well-drained acidic agricultural land in Indonesia, both regarding geographic distribution, agricultural use and soil properties. Soils at sites 2 to 12 were collected in close proximity to each other. However, both the content of C and the CEC varied by a factor of five, so the samples represented variable soil characteristics even though they were geographically close together.

In the laboratory, each of the 31 bulked soil samples (sieved at 2 mm) were divided into 21 subsamples (~10-50 g), to which was added either 0, 0.1, 0.3, 1, 3, 10 or 30 % (dw) of one of three BCs (sieved at 2 mm).-Assuming a bulk density of 1.5 g cm^{-3} (typical for A-horizons in tropical soils, cf. Batjes (1996)) and a soil depth of 20 cm (common depths for the plough layer) these amounts of BC correspond to 0, 3, 9, 30, 90, 300 and 900 tons BC ha^{-1}.

2.2. Biochar production

Three types of BC (cacao shell , oil palm shell, and rice husk) were produced in a locally constructed unit (Fig. S1) of 30-40 L, and a chamber temperature around 250-350 ^{0}C (average 300°C). Several pyrolysis times were tested and the ratio of BC:syngas and the BC yield were measured. The pyrolysis times were selected after charring the respective feedstock materials for 1, 2 and 3.5 h. The optimal pyrolysis time was selected on the basis of the amount of carbon recovered (i.e., the C content times the yield), as part of the motivation for using BC is the C sequestration effect. In addition, the characteristics of the BC (%ash, N, P) varied by < 5% between the three pyrolysis times (Table S1). The yield for each of the three BC produced (compared to dry weight of feed stock) were 22.0%, 53.5% and 30.4% for cacao shell, oil palm shell and rice husk, respectively (Table S1).

2.3. Analysis of soil and biochar soil mixtures

pH of the soil and the BC soil mixtures was determined electrometrically (*W/V Orion Model 410A*) in a soil suspension with distilled water as well as a 1 M KCl solution (weight soil: volume solution ratio of 1:5). The CEC and exchangeable cations were determined by percolation with 1M ammonium acetate (pH = 7.0) followed by extraction with 0.17 M sodium chloride after washing with alcohol. pH was measured on four soil samples without added BC and in each of the 0.1, 0.3, 1, 3, 10 or 30 % BC soil mixtures at each site. CEC was measured without BC at each site (for characterization) and in each of the 0, 0.1, 0.3, 1, 3, 10 or 30 % BC soil mixtures of each of the five sites 18, 19, 24, 30, 31 (i.e. on four soil samples without added BC at each of the five sites). Base cations (Ca, Mg, K, Mn and Na) replaced by ammonium ions (NH_4^+) were measured in the first eluent with a flame spectrophotometer (Perkin Elmer, AAS 3300). After washing with ethanol (96%) to remove excess ammonium acetate, adsorbed NH_4^+ was displaced by Na^+ - ions. The CEC was determined colorimetrically as the total amount of extracted NH_4^+ ions, using blue indofenol complexation (Ciesielski and Sterckeman, 1997; Rhine *et al.*, 1998), using a spectrophotometer (*Autoanalyzer 3 Bran Luebbe)* at 636nm. The procedure used for determining CEC might have underestimated the actual CEC due to dissolution of organic matter and subsequent hydrolysis as reported by Harada and Inoko (1980). The exchangeable acidity (H^+ and Al^{3+}) was measured in 1 M KCl solutions, where phenolphthalein was added and the solution titrated with 0.02 M NaOH to pH 7. For exchangeable Al^{3+}, 1 M NaF was added to the titrated sample, and the solution was titrated back to pH 7 with 0.02 M HCl (until colour disappears). The difference between these two measurements equals the approximate exchangeable H^+ concentration (Mc Lean in Black *et al.*(1965)). Total C and N were determined by the dry combustion (Nelson and Sommers, 1982) (Leco CHN-1000; Leco Corporation, Sollentuna, Sweden) and the Dumas method (Bremmer and Mulvaney, 1982), respectively. Due to the absence of carbonates in the native soils, suggested by their low pH, total C represents organic C before BC addition. All measured soil attributes are listed in the supporting information (Table S2).

2.4. BC analyses

pH of the three BCs was determined electrometrically (*Orion, model 720, Orion Research Inc., Cambridge, MA, USA*) in a suspension with distilled water and 1M KCl (10 ml BC and 25 ml water/KCl solution), respectively. The CEC and exchangeable cations of the BCs (air-dried and sieved at 2 mm) were determined by percolation with 1M ammonium acetate (pH = 7.0) followed by extraction with 1 M potassium chloride after washing with alcohol. In the first eluent base cation concentrations were determined using ICPOES (Optima 5300 DV, PerkinElmer Inc., Shelton, CT, USA). Extractable acidity was determined by back titration with 0.05 M sodium hydroxide to pH 7. The sum of exchangeable base cations and acidity was used to determine CEC (i.e. including base cations leached from ashes) according to Schollenberger & Simon (1945). After washing with propan-2-ol the samples were extracted with KCl and the CEC determined photometrically as the total amount of extracted NH_4^+ ions (Photometer, Gilford Instrument). In addition, CEC was determined after saturation with 1M KCl and subsequent extraction with 0.5M $NaNO_3$ according to the method described by Mukherjee *et al.* (2011). Total C and N were determined as described above.

The BCs (samples of approximately 10 and 100 mg) were analysed for moisture and ash content on a thermogravimetric analyzer (TGA). The samples were heated to 150 ºC and held at that temperature for 45 minutes. The percentage mass loss after this hold time is reported as the percentage moisture in the BC. The temperature was raised to 650 ºC and held for one hour then raised to 900 ºC and held for 45 minutes. The combined weight loss at these two temperatures is taken as the loss on ignition (LOI) and the percentage ash (100% less LOI) reported on a dry weight basis. In addition, approximately half a gram of sample was weighed into a polypropylene bottle, 50 mL of an aqueous solution of 0.05 N HCl and 0.1 N $NaNO_3$ was added and the mixture equilibrated on a rotator for 16 to 24 hours. The mixture was filtered through a nominal 0.7 um glass fiber filter and the filtrate back titrated with 0.05 N NaOH and 0.1 N $NaNO_3$ solution. The acid consumed is reported in $cmol_c$/kg and represents the acid neutralizing capacity (ANC).

The surface area (BET) of the chars was determined by adsorption of nitrogen (N_2) at -196°C, using an automated surface area analyzer at US Geological survey, Denver Colorado. The samples were out-gassed by heating at 110ºC under a flow of ultrahigh purity helium at 10 cm^3 min^{-1} for 16 to 24 hr prior to analysis. Isotherm data were recorded at partial pressures of N_2 between 0.05 and 0.95 atmospheres. The apparent surface areas of samples were obtained from the statistical monolayer capacities of N_2 from the BET plots (Atkins, 1990). For further details see Rutherford *et al.* (2005).

2.5. Statistical analyses

To describe the intrinsic nonlinear relationship between BC addition and the observed soil pH, a nonlinear regression model was used. For each of the three BCs oil palm shell, cacao shell, and rice husk, a three-parameter exponential function (Equation (1) was fitted to describe the response of soil pH (in H_2O and KCl, respectively) to BC addition (0, 0.1, 0.3, 1, 3, 10 and 30%).

$$y= a+b*(1- \exp^{(-x/c)}) \quad (1)$$

In Equation (1) x is the amount of added BC (%) and y denotes soil pH (the dependent variable). In the regression model parameter "a" represents the mean soil pH level for soils without BC addition (i.e., for 0% BC). Parameter "b" represents the maximum additional increase in soil pH (added to the level a) as BC addition is increased from 0% to 30%. Parameter "c" is the rate of change (i.e. a rate constant, which has the reciprocal unit of percentage BC added; the smaller the rate constant c the faster are the changes in pH per unit increase in BC in soil).

Initially, we assumed that the model parameters "b", and "c" differed between BC types, whereas only a single parameter "a" was used for all soils to denote the common mean level in soil pH without BC addition. Due to substantial variation in soil characteristics between the 31 sampling sites (Table 1), site-specific variation was modelled in the regression parameters "a", "b" and "c" by introducing random effects so that each model parameter was the sum of a contribution reflecting the pure BC effect and another contribution, reflecting the site-specific effect. This means that we extended the ordinary nonlinear regression model based on Equation (1), as it ignored variation between sites, to a nonlinear mixed-effects regression model with site-specific random effects (e.g. Crawley (2007) and Pinheiro and Bates (2000)). Likelihood ratio tests were used to simplify the fixed-effects structure of the models (Table S3), i.e., we investigated whether or not the model parameters "b" and "c" were in fact BC-specific (cf. Suuster *et al.* (2011)). The resulting estimated mean curves for the three BCs reflect the trends seen across all sites. Additionally, the same nonlinear mixed models were fitted to the subset consisting of sites 18, 19, 24, 30, 31 (Table 1) randomly selected for the analysis of CEC to determine the effect of BC addition on CEC and exchangeable base cations (Table S3).

Subsequently, linear regression was used for analysing the relationship between the estimated site-specific rates of change ("c" parameters) obtained from the nonlinear mixed model analysis (i.e., the estimated fixed effect and random effect added up) and initial pH, CEC and total C of the soils at each site. This analysis is independent of BC type as BC-specific differences in the rates of change amounts to vertical shifts in the rate constants. For the linear regression we used the cacao shell BC rates of change. We present parsimonious models obtained after model reduction using backwards stepwise elimination of non-significant terms.

The statistical software package "R", version 2.13.2 (R Development Core Team, 2011), was used for all statistical analyses. The nonlinear mixed-effects models were fitted using the function "nlme" in the R extension package nlme (Pinheiro *et al.*, 2011). Visualization of the fitted models was achieved using the package ggplot2 (Wickham, 2009).

3. Results

3.1. Properties of the soils and the biochars

The selected Indonesian soils from the 31 sites were acidic (mean $pH_{(H2O)}$ = 4.7 ± 0.47 (sd), n = 122, mean $pH_{(KCl)}$ = 3.9 ± 0.28 (sd), n = 122) with a moderate CEC (mean 7.5 ± 4.03 (sd)

$cmol_c/kg$, n = 43, Table S2). The mean percentage of organic C and N was 1.7 ± 1.67 (sd) % and 0.1 ± 0.1 (sd) %, respectively (n= 31, Table 1) with levels of C ranging from 0.43% to 8.9%. There were substantial differences in the properties of the three BCs (Table 2). pH and ANC were in the range of 6.7 (oil palm shell BC) to 10.5 (cacao shell BC) and 36 $cmol_c/kg$ (oil palm shell BC) to 217 $cmol_c/kg$ (cacao shell BC), respectively. NH_4Ac-extractable cations and CEC were greater for the cacao shell BC (197 and 30-37 $cmol_c/kg$, respectively) as compared to the oil palm shell BC (35 and 11-20 $cmol_c/kg$, respectively) and the rice husk BC (20 and 7-26 $cmol_c/kg$, respectively).

3.2. Changes in soil pH in response to BC addition

As pH_{H2O} and pH_{KCl} were significantly correlated (r= 0.96, $p<0.001$, n= 674) and both parameters responded to the addition of BC in a similar fashion, only the pH_{H2O} are shown in figures and used in the models presented. Recalling that model parameter "a" represents the mean pH for all sites without BC addition (thus the same for all 3 BC types), the pH response to BC addition is determined by model parameters "b" (maximum additional increase in pH) and "c" (the rate of change) only. The estimated parameters "b" for the $pH_{(H2O)}$ response upon the addition of cacao shell-, oil palm shell- and rice husk-BCs were significantly greater than 0 ($p<0.001$) for all three BCs (Table 3) therefore, resulting in a significant increase in pH with BC addition (Question 1; Fig. 2). In addition, the mean response in soil pH as a function of BC addition differed substantially between the three types of char (Question 4). More specifically, we found that parameter "b" was not significantly different between oil palm shell and rice husk (p=0.59), whereas parameter "c" was significantly different (p=0.048; Table S3). There were highly significant differences in both parameters "b" (greater) and "c" (smaller) between cacao shell on the one hand and oil palm shell and rice husk on the other ($p<0.0001$ in both cases) illustrating a stronger response and a greater maximum increase in soil pH with addition of cacao shell BC as compared to oil palm shell and rice husk BC. We found a significant linear relationship between the estimated parameters "c" (i.e. the rate of change in pH) for the cacao shell BC addition to the different soils and the initial CEC (c = 3.2 + 0.58*CEC, R^2 = 0.71, p<0.001, Fig. 3) indicating a more distinguished effect on soil pH upon BC addition for low CEC soil than for high CEC soil with high buffering capacity (Question 3).

3.3. Changes in soil CEC and levels of base cations with biochar addition

The cation exchange capacity (determined only for sites 18, 19, 24, 30, 31, Table 1) increased significantly (p<0.001) with BC addition (Question 2, Table 3, Fig. 4). At 30% BC addition, the CEC increased to 8.12 and 7.93 $cmol_c$/kg for the cacao shell/oil palm shell and rice husk BC soil mixtures, respectively, as compared to the initial CEC of 5.62 $cmol_c$/kg at the five sub-sites (Table 3). There was no significant (p= 0.42) difference in the "b" parameter (2.51) between the three BCs but parameter "c" was significantly greater (hence a smaller rate of change in CEC, Question 4) for the rice husk BC as compared to cacao shell and oil palm shell BCs (p<0.0001), which did not differ significantly from each other (p= 0.32). Furthermore, there was a highly significant increase in amounts of extractable Ca and Mg upon the addition of all three BCs (p<0.001; Table 3, Fig. 4). In contrast, only the addition of cacao shell BC significantly increased levels of K (p<0.001). For Ca, Mg and K the parameters "b" and "c" were significantly different for the cacao shell BC as compared to the oil palm shell and rice husk BC's (p<0.05; Table 3, Fig. 4). Significantly larger "b" parameters for the cacao shell BC indicate a larger maximum increase in the amount of base cations for this BC.

4. Discussion

The cacao shell, oil palm shell and rice husk BCs differed substantially in physical and chemical properties (Table 2). Large differences in the quality of BCs due to intrinsic feedstock properties and production procedures have been reported previously by e.g. Chen *et al.* (2008), Yuan and Xu (2011), Yuan *et al.* (2011) and Rutherford *et al.* (2012). Assessing the ameliorative effects of low temperature BC generated from nine different crop residues, Yuan and Xu (2011) found a great variation in soil pH, alkalinity and amounts of extractable base cations, with the most prominent difference occurring between legume vs non-legume feedstocks. The pH and alkalinity of the BCs was in general greater in legumes as compared to non-legume feedstocks due to a larger uptake of alkali ions in the former. In addition to feedstock properties affecting the quality of BC, the production temperature and method may also be of significant importance as e.g. reported by Budai *et al.* (2014) for corncob and miscanthus BCs. In our study the production procedure is similar for the three BCs, with the exception of a shorter pyrolysis time for the oil palm shell BC. This shorter time was selected for reasons described in the method section. However, the resulting differences in yield and ash content between 1h and 3.5h pyrolysis time for the oil palm shell was small (4.9% and 0.3% respectively, Table S1). In addition, the differences in properties are between the cacao shell BC on the one hand, and oil palm shell and rice husk on the other. If pyrolysis time was to explain the differences between the BC properties, the oil palm shell BC should have been the material with properties much different from the two other materials. Thus, the different properties of the three BCs are likely caused by the feedstock. Furthermore, the amount of NH_4Ac-extractable cations is in the order cacao shell>>oil palm shell>rice husk while the CEC (i.e. excluding the ash fraction) is in the order cacao shell>>rice husk~oil palm shell (Table 2). This clearly illustrates the importance of methodology when determining CEC, viz. sum of base cations and acidity as compared to analysis of extractable NH_4-N or K. Potassium in particular was found at the highest concentration in the cacao shell BC, which also had the greatest K content in the feedstock (Table S1). This suggests s a better K-fertilizing effect of the cacao shell BC when applied to soil, as compared to the oil palm shell BC and rice husk BC. However, the addition of rice husk BC may also increase the levels of K in soils, as reported by Haefele *et al.* (2011). Adding 4.13 kg m^{-2} of carbonized rice husk BC combined with a medium fertilizer rate significantly increased levels of K from 441 mg kg^{-1} to 620 mg kg^{-1} in anthraquic Gleysols (depth 0-0.15 m), at the IRRI lowland research farm in the Philippines (Haefele *et al.*, 2011).

Soil pH was more sensitive and had a greater maximum increase with the addition of cacao shell BC as compared to oil palm shell and rice husk BC. Soil pH increased rapidly (parameter c = 8.59) with only small amounts of cacao shell BC added (Fig. 2A, Table 3). An increase in soil pH from the initial mean value of 4.73 to pH 5 required only addition of 0.6% cacao shell BC. In contrast, much more oil palm shell BC (10 times more) or rice husk BC (12 times more) were needed for the same increase in pH (from 4.73 to 5; Fig. 2A, B and C, Table 3). After 30% BC addition (corresponding to the unrealistic amount of ~900 tons BC ha^{-1} which was only tested for mechanistic purposes), the estimated soil pH was 8.95 for the cacao shell BC and 5.52 and 5.47 for the oil palm shell- and rice husk BCs, respectively. In most field experiments, application rates of 0.5-2% (or 15 to 60 ton/ha assuming an incorporation depth of 20 cm and a dry bulk density of 1.5 g/cm^3) are used (Jeffery *et al.*, 2011; Martinsen *et al*. 2014; Schimmelpfennig *et al*. 2014). Within this range of BC addition our findings clearly show the different potentials of the BCs as liming agents; viz. cacao shell BC has a large potential to act as a liming agent, whereas oil palm shell and rice husk BCs have not. As the alkalinity of the BC is a key factor controlling its liming effect (Yuan and Xu, 2011), the greatest response when adding cacao shell BC could be explained by its higher

ANC (217 $cmol_c$/kg) as compared to the oil palm shell and rice husk BCs (ANC = 36 and 45 $cmol_c$/kg , respectively; Table 2).

Different effects on soil pH of the addition of various types of BC were previously reported by Yamato *et al.* (2006) and Yuan and Xu (2011). The addition of 37 tons ha^{-1} (10 dm^3 m^{-2}, $BD_{biochar}$ 0.37 kg dm^3) bark charcoal (*Acacia mangium*) increased the pH between 0.9 and 1.4 units in soils from three sites in South Sumatra (Yamato *et al.*, 2006). The $pH_{(H2O)}$ of the charcoal was 7.4, hence more similar to the oil palm shell and rice husk BC than the cacao shell BC used in our study (Table 2). However, at 1% BC addition in our study (i.e. about 30 tons ha^{-1}) the estimated increase in soil pH was about 0.5 units for the cacao shell BC whereas this was only 0.05 and 0.04 units for the oil palm shell and rice husk BC, respectively. Furthermore, adding 1% of different BCs derived from legume and non-legume feedstocks to an acidic ultisol from China, Yuan and Xu (2011) found an increase in pH ranging from 0.18 (non-legume) to 1.05 (legume) units. This corresponds with our findings for the cacao shell BC and further indicates the limited liming potential of the BCs from oil palm shell and rice husk ~~BCs~~. Interestingly, both Yamato *et al.* (2006) and Yuan and Xu (2011) reported a significant reduction in exchangeable acidity (Al^{3+} and H^+ $cmol_c$/kg) upon the addition of BC. This positive effect of increased pH and thus reduced risk for Al toxicity was also observed with the addition of cacao shell BC in our study. Based on a subset from the 31 sites (five sites, numbered 18, 19, 24, 30, 31; Table 1) amounts of exchangeable Al^{3+} before BC addition (mean 2.3 ± 2.25 (sd) $cmol_c$/kg, n= 14) were reduced to zero at all sites after addition of 3% cacao shell BC (Table S2). By contrast, exchangeable Al^{3+} was not eliminated at all sites after the addition of 30% oil palm shell and rice husk BC (mean oil palm shell 0.31 ± 0.38 (sd) $cmol_c$/kg (n=4) and mean rice husk 0.87 ± 1.56 (sd) $cmol_c$/kg (n=5)).

There was a large variation in the response of soil pH to BC addition between the 31 sampling locations, which resulted in different parameter estimates (and thus response curves) for each of the sampling sites (Fig. 2A, B and C). This clearly illustrates the importance of intrinsic soil properties (Table 1) when determining effects of BC addition on changes in soil pH. There was a significant relationship between the estimated parameters "c" for the cacao shell BC additions and initial soil CEC and pH (R^2 = 0.58, p<0.001). The parameters decreased (i.e. a greater increase in pH) with an increase in initial soil pH and increased (i.e. smaller increase in pH) with initial CEC, suggesting a greater response in pH with BC addition at sites with low CEC and high pH and thus smaller amounts of exchangeable acidity. Contrary to expectations, the CEC of the soil was more important than the initial soil pH for the pH effect of BC. Of the two explanatory variables, the initial CEC (R^2 = 0.42) explained more of the variation seen in the parameter "c" than initial pH (R^2 = 0.16). The estimated "c" parameters for sites 9 and 12 were unduly large (19.29 and 15.08, respectively). If these two points are excluded from the regression model, initial CEC is the only parameter retained (R^2 = 0.71, p<0.001, Fig. 3). In accordance with the model including both CEC and pH, the latter predicts an increased (0.58) value for parameter "c" (thus a smaller pH response with BC addition) per unit increase in CEC (Fig. 3). This is in accordance with the results reported by Streubel *et al.* (2011) who found a greater increase in soil pH with the addition of herbaceous and woody BCs to a sandy soil (3.3 $cmol_c$/kg) as compared to silty loamy soils (CEC 15.4-16.6 $cmol_c$/kg) in Washington. As in our case, the different responses were attributed to an inherently lower buffering capacity of sands as compared to medium and fine texture soils (Streubel *et al.*, 2011).

Biochar addition to acidic soils has earlier been observed to increase CEC and amounts of exchangeable base cations (Yuan and Xu, 2011; Yuan *et al.*, 2011; Glaser *et al.*, 2002).

According to Glaser *et al.* (2002), the addition of BC, which naturally includes ash, adds free bases to the soil. This may increase the pH and the readily available nutrients for plant growth. In addition, the nutrient retention can be improved with BC (Hale *et al.*, 2013), an effect that does not derive from the ash but from the BC *per se* (Glaser *et al.*, 2002). Our results support these findings, despite a more pronounced BC specific effect on the quantity of base cations (and thus soil pH) as compared to CEC (Fig. 4). The addition of 2% BC increased the CEC about 1 and 0.4 $cmol_c$/kg for cacao shell/oil palm shell BC and rice husk BC soil mixtures, respectively (Table 3, Fig. S2). In accordance with its great amount of exchangeable base cations (Table 2), addition of 2% cacao shell BC caused the largest increase in soil exchangeable Ca, Mg and K (0.66, 0.47 and 1.58 $cmol_c$/kg, respectively). Increases in soil exchangeable Ca, Mg and K due to addition of 2% of the other BCs were significantly smaller (for oil palm shell 0.21, 0.16 and 0.04 $cmol_c$/kg, respectively and for rice husk 0.04, 0.07 and 0.09 $cmol_c$/kg, respectively; Fig. 4 and Fig. S2). For the sites included in this study, there was no significant relationship between organic C content and CEC, (p = 0.70, R^2 = 0.006, n= 29). However, when excluding the three sites (23, 25 and 26, Table 1) with organic C content > 3%, the relationship was significant (CEC = 4.16 + 3.78 %C, p = 0.04, R^2 = 0.16, n= 26). This relationship indicates that the CEC per percent organic C is similar to recently published values for acid forest soils from southern Poland (Gruba and Mulder, 2015). In addition, the average contribution of clay minerals to CEC (4.16 $cmol_c\ kg^{-1}$ soil) in our soils is of the same order of magnitude or slightly smaller than the contribution of soil organic matter (Fig. S3). The three BCs, having CEC values of 30-37, 11-20 and 7-26 $cmol_c$/kg (cacao shell, oil palm shell and rice husk BCs, respectively; Table 2), were added to soils that had a mean CEC of 5.62 $cmol_c$/kg (Table 3). If there would be no pH-dependent effects on CEC through the addition of 30 % BC, the CEC would potentially increase to ~13-15, 7-10 and 6-11.8 $cmol_c$/kg, respectively. As shown in Fig. 4, we found that at 30% BC addition the modeled mean increase of CEC was 8.12 and 7.93 $cmol_c$/kg for the cacao shell/oil palm shell and rice husk BC soil mixtures, respectively. The somewhat smaller change in CEC than expected based on the potential might be due to a reduction of the CEC of the BC (pH dependent binding sites at the BCs) when added to the acidic soils.

The present paper shows that BC has a pH-increasing effect on soil, and that the effect is dependent on both soil and BC characteristics. The strongest effects were observed for high-CEC BC in the least acid soils with relatively low-CEC. In these soils, the CEC was a more important characteristic than initial pH (Fig. 3). This work will aid in mapping the extent to which BC can have a beneficial effect on soil fertility in acidic agricultural lands.

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Supplementary data

The supplementary data contains one figure and three tables.

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Figure Legends

Fig. 1 Map of soil sampling locations in Indonesia.

Fig. 2 Estimated (curves) and observed (points) response in soil $pH_{(H2O)}$ following the addition of cacao shell BC (**A**. n = 186 for 0.1 to 30% BC), oil palm shell BC (**B**. n= 180 for 0.1 to 30% BC) and rice husk BC (**C**. n= 186 for 0.1 to 30% BC) in soils from 31 sites across Indonesia (Table 1). The figure shows fitted curves (Table 3) for the mean response in soil pH (bold curve; **A**: a = 4.73, b = 4.35 and c = 8.59; **B:** a = 4.73, b = 1.00 and c = 19.35; **C**: a = 4.73, b = 1.00 and c = 22.49) superimposed on predictions of the response in soil pH for each of the 31 sampling sites. Note: Y-axis scales differ between **A-C**. Oil palm shell was missing for site 30. The relationship between the observed response in pH and CEC of the BCs is given in Figure 3.

Fig. 3 Relationship between estimated parameters "c" (parameter "c" estimates deriving from nonlinear mixed model analysis; $pH_{(H2O)}$ vs. BC addition) and cation exchange capacity (CEC, $cmol_c$ /kg) of soils before BC addition from 30 sites across Indonesia (Table 1). The parsimonious model after removal of two large rate constants (site 9 and 12; grey dots) is shown). One sampling site (20) is omitted due to lack of CEC. Note: A decrease in the parameter "c" implies a greater change in soil pH in response to BC addition (cf. Fig. 2).

Fig. 4 Predicted response in soil CEC ($cmol_c$/kg) and of available Ca, Mg and K ($cmol_c$/kg) to added cacao shell BC, oil palm shell BC and rice husk BC in soils from 5 sites (site 18, 19, 24, 30, 31; Table 1) across Indonesia. The fitted curves derive from the model y = a + b *(1-

659 $\exp^{(-x/c)}$) (eq. 1) based on restricted maximum likelihood estimates (Table 3) for the mean response in CEC (n=101) and levels of cations (Ca; n=97[§], Mg; n=97[§§] and K; n=90) to BC addition. [§]One outlier excluded (site 18, cacao shell-BC level 30%). [§§] One outlier excluded (site 19, oil palm shell-BC level 10%). Note: Y-axis scales differ between the plots.

Table 1. Site ID, location, agricultural use (crop type), soil type (Soil Survey Staff, 1999), clay mineralogy, carbon (C) and nitrogen (N) content (in dry weight %), pH in H_2O and KCl (mean of 4 subsamples ± SE) and CEC ($cmol_c$ /kg ; ± SE for the sites 18,19,24,30,31) for 31 sampling sites across Indonesia. nd = not done.

Site ID	Site name	Coordinate	Agricultural use	Soil type	Clay mineralogy		C (wt%)	N (wt%)	pH $_{(H2O)}$	pH$_{(KCl)}$	CEC
					Dominant	Others					($cmol_c$/kg)
1	Babakan Dramaga Village. Bogor. W. Java	6.56363 S 106.72734 E	Maize and cassava	Typic Dystrudepts	Kaolinite	Halloysite	1.18	0.12	4.69 ± 0.07	3.79 ± 0.03	15.37
2	Portibi.Padang Lawas Utara. N. Sumatera	1.29492 N 99.68059 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.67	0.12	4.49 ± 0.06	3.77 ± 0.02	13.44
3	Portibi.Padang Lawas Utara. N. Sumatera	1.30941 N 99.67722 E	Palm oil	Hapludults	Kaolinite	Gibbsite	0.43	0.03	5.28 ± 0.06	3.91 ± 0.02	9.33
4	Portibi.Padang Lawas Utara. N. Sumatera	1.30362 N 99.68623 E	Palm oil	Hapludults	Kaolinite	Gibbsite	2.18	0.15	4.58 ± 0.06	3.85 ± 0.06	15.39
5	Portibi.Padang Lawas Utara. N. Sumatera	1.30194 N 99.67887 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.62	0.08	4.68 ± 0.05	3.99 ± 0.05	9.00
6	Portibi.Padang Lawas Utara. N. Sumatera	1.29922 N 99.67598 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.72	0.09	4.36 ± 0.02	3.90 ± 0.06	12.76
7	Portibi.Padang Lawas Utara. N. Sumatera	1.31621 N 99.67390 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.14	0.05	4.73 ± 0.06	3.95 ± 0.04	7.29
8	Portibi.Padang Lawas Utara. N. Sumatera	1.30428 N 99.67834 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.42	0.07	4.19 ± 0.07	3.81 ± 0.03	12.15
9	Portibi.Padang Lawas Utara. N. Sumatera	1.30124 N 99.67244 E	Palm oil	Hapludults	Kaolinite	Gibbsite	0.76	0.03	4.35 ± 0.01	3.60 ± 0.01	15.83
10	Portibi.Padang Lawas Utara. N. Sumatera	1.29720 N 99.69182 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.27	0.06	4.51 ± 0.01	3.87 ± 0.04	10.69
11	Portibi.Padang Lawas Utara. N. Sumatera	1.29735 N 99.67092 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.04	0.04	4.58 ± 0.04	3.81 ± 0.03	6.90
12	Portibi.Padang Lawas Utara. N. Sumatera	1.31126 N 99.67369 E	Palm oil	Hapludults	Kaolinite	Gibbsite	1.45	0.07	3.35 ± 0.04	3.10 ± 0.02	3.34
13	Riau	0.30383 N 100.91294 E	Maize, peanut and cassava	Hapludults	Kaolinite		0.97	<0.02	5.61 ± 0.04	4.75 ± 0.03	2.51
14	Riau	1.09086 N 102.11622 E	Maize	Sulfaquents	Kaolinite	Vermiculite	1.03	0.03	5.12 ± 0.09	3.93 ± 0.03	5.83
15	Riau	1.08622 N 102.13917 E	Cassava	Sulfaquents	Kaolinite	Vermiculite	1.23	0.06	4.16 ± 0.03	3.81 ± 0.02	7.08
16	Jambi	3.51574 S 104.88022 E	Rubber area	Tropaquepts	Kaolinite	Goethite	0.88	0.04	4.11 ± 0.03	4.03 ± 0.04	7.15
17	Jambi	3.36217 S 104.83245 E	Rubber area	Tropaquepts	Kaolinite	Goethite	1.62	0.09	4.80 ± 0.06	3.81 ± 0.03	9.05
18	Riau	0.89505 N 112.55643 E	Annual crop	Typic Kanhapludults	Kaolinite		0.81	0.04	4.43 ± 0.02	3.90 ± 0.01	6.95 ± 0.03
19	Jambi	0.00015 N 112.54450 E	Oil palm area	Typic Kandiudults	Kaolinite		1.34	0.09	4.87 ± 0.10	4.23 ± 0.04	2.22 ± 0.09
20	Kayu Agung. Palembang	0.00015 N 112.54450 E	Maize	Typic Kandiudults	Kaolinite		1.54	0.11	5.41 ± 0.01	4.31 ± 0.01	nd
21	Kayu Agung. Palembang	0.90170 N 112.54189 E	Maize	Typic Dystrudepts	Kaolinite		1.53	0.11	4.66 ± 0.07	3.78 ± 0.03	11.88
22	Tamanbogo. East Lampung	0.89570 N 112.55680 E	Maize, paddy and cassava	Typic Kanhapludults	Kaolinite	Goethite	0.90	0.05	4.44 ± 0.08	3.88 ± 0.03	4.97
23	W. Kalimantan	0.30769 N 100.90822 E	Scrubland	Hapludults	Kaolinite	Gibbsite	6.01	0.40	5.20 ± 0.07	4.28 ± 0.03	3.51
24	W. Kalimantan	0.89580 N 112.55636 E	Annual crop	Typic Kanhapludults	Kaolinite		0.81	<0.02	4.79 ± 0.11	3.82 ± 0.01	6.15 ± 0.25
25	W.t Kalimantan	0.31728 N 100.90089 E	Rubber area	Hapludults	Kaolinite	Gibbsite	8.91	0.37	4.32 ± 0.08	3.93 ± 0.08	7.52
26	E. kalimantan	5.01128 S 105.49458 E	Vegetable area	Kanhapludults	Kaolinite		3.06	0.28	5.18 ± 0.16	3.91 ± 0.03	10.04
27	E. kalimantan	0.41894 N 101.45967 E	Maize and vegetable area	Humitropepts	Kaolinite		1.94	0.21	5.19 ± 0.16	3.86 ± 0.04	10.73
28	E. kalimantan	1.01956 N 102.13917 E	Vegetable area	Sulfaquents	Kaolinite	Illite, vermiculite	1.84	0.14	4.57 ± 0.04	3.78 ± 0.03	12.28
29	E. Kalimantan	0.89520 N 112.55636 E	Maize, peanut and vegetables	Typic Kanhapludults	Kaolinite		0.98	0.07	4.39 ± 0.01	3.85 ± 0.01	3.41
30	E. Kalimantan	0.89535 N 112.55646 E	Vegetable area	Typic Kanhapludults	Kaolinite		0.82	<0.02	5.05 ± 0.11	4.43 ± 0.01	10.03 ± 0.02
31	E. Kalimantan	0.89501 N 112.55652 E	Cassava, pineapple and king grass	Typic Kanhapludults	Kaolinite		0.52	0.05	4.70 ± 0.17	4.19 ± 0.01	1.61 ± 0.01

Table 2. Selected attributes of biochars produced from cacao shell, oil palm shell and rice husk.

Biochar type	Pyrolysis time (h)	C (%)	N (%)	BET (m^2 g^{-1})	pH (H_2O)	pH (1M KCl)	CEC[1] (cmolc/kg)	CEC[2] (cmolc/kg)	CEC[3] (cmolc/kg)	Ca (cmolc/kg)	K (cmolc/kg)	Mg (cmolc/kg)	Na (cmolc/kg)	Ash[4] %	ANC (cmolc/kg)
Cacao shell	3.5	69.59	1.37	29	10.5	10.0	197 ± 1.2	37 ± 2.4	30 ± 3.6	37.1	126.8	32.8	0.3	18.9	217
Oil palm shell	1	61.49	1.73	<1	6.7	5.7	35 ± 5.2	20 ± 2.1	11 ± 3.6	9.0	6.5	7.4	0.2	10.5	36
Rice husk	3.5	41.24	1.00	51	7.3	6.1	20 ± 4.3	26 ± 4.8	7 ± 0.1	3.2	9.5	3.6	0.2	51.0	45

[1]CEC measured as the sum of base cations and exchangeable acidity in NH_4Ac-extracts (n=3, SE shown).
[2]CEC measured as the amount of extractable NH_4 after saturation with NH_4Ac and subsequent extraction with 1M KCl (n=3, SE shown).
[3]CEC measured after saturation with 1M KCl and subsequent extraction with 0.5M NaNO3 (n=2, SE shown) (Mukherjee et al. 2011).
[4]Large sample of approximately 100 mg.

Table 3. Estimated fixed-effects parameters for the model $y = a + b * (1 - \exp^{(-x/c)})$ (eq. 1) based on restricted maximum likelihood estimates from nonlinear mixed effects-models for the response in CEC, Ca, Mg and K ($cmol_c$/kg) to BC addition (%). The table shows biochar specific estimates (cacao shell, oil palm shell and rice husk) for the parameters a, b, and c ± SE. For each model capital letters for the parameters b and c indicate differences between the BC types. Different letters indicate difference in parameter estimate at a level of significance <0.05. Number of observations (n) for each model is shown.

Model	Biochar	Parameter estimates (± SE)		
		a	b	c
$pH_{(H2O)}$ ~ BC%, n= 674§	Cacao shell	4.73 ± 0.09	4.35 ^{A} ± 0.09	8.59 ^{A} ± 0.64
	Oil palm shell		1.00 ^{B} ± 0.10	19.35 ^{B} ± 1.85
	Rice husk			22.49 ^{C} ± 2.08
CEC ~ BC%, n= 101§§	Cacao shell	5.62 ± 1.47	2.51 ± 0.43	3.94 ^{A} ± 0.94
	Oil palm shell			
	Rice husk			11.77 ^{B} ± 3.13
Ca ~ BC%, n= 97	Cacao shell	0.50 ± 0.12	5.63 ^{A} ± 0.37	16.10 ^{A} ± 3.13
	Oil palm shell		1.29 ^{B} ± 0.15	11.45 ^{B} ± 2.90
	Rice husk			62.93 ^{C} ± 22.97
Mg ~ BC%, n= 97	Cacao shell	0.19 ± 0.03	5.79 ^{A} ± 0.51	23.60 ^{A} ± 3.67
	Oil palm shell		1.82 ^{B} ± 0.50	21.58 ^{B} ± 11.28
	Rice husk			50.71 ^{C} ± 20.38
K ~ BC%, n= 90	Cacao shell	0.05 ± 0.03	31.32 ^{A} ± 4.25	38.67 ^{A} ± 6.76
	Oil palm shell		1.94 ^{B} ± 3.56	85.94 ^{B} ± 187.34
	Rice husk		3.94 ^{C} ± 7.30	

§n=552 for 0.1-30% BC addition (Oil palm shell not analysed at site 30) + 122 0% BC addition (1 missing value at site 20 and 30); Table S2.

§§n=83 for 0.1-30% BC addition (Oil palm shell not analysed at site 30 and 1 missing value at site 24) + 18 0% BC addition (1 missing value at site 30 and 31); Table S2.


N
INDIAN OCEAN
AUSTRALIA
Kuala Terengganu
Ipoh
Kuantan
Strait of Malacca
Kuala Lumpur
Kelang
Singapore
MALAYSIA
SINGAPORE
Kuching
2-12
17
19
16
13
15
18
14
25-31
24
23
Borneo
Pontianak
Sumatra
Padang
Jambi
Karamata Strait
Palembang
20
21
Bengkulu
22
Java Sea
Tanjungkarang-Telukbetung
Tangerang
Jakarta
1
Bekasi
Cirebon
Tegal
Semarang
Surakarta
Surabaya
Bandung
Yogyakarta
Malang
Situbondo
0
250
500 km

Fig. 1.

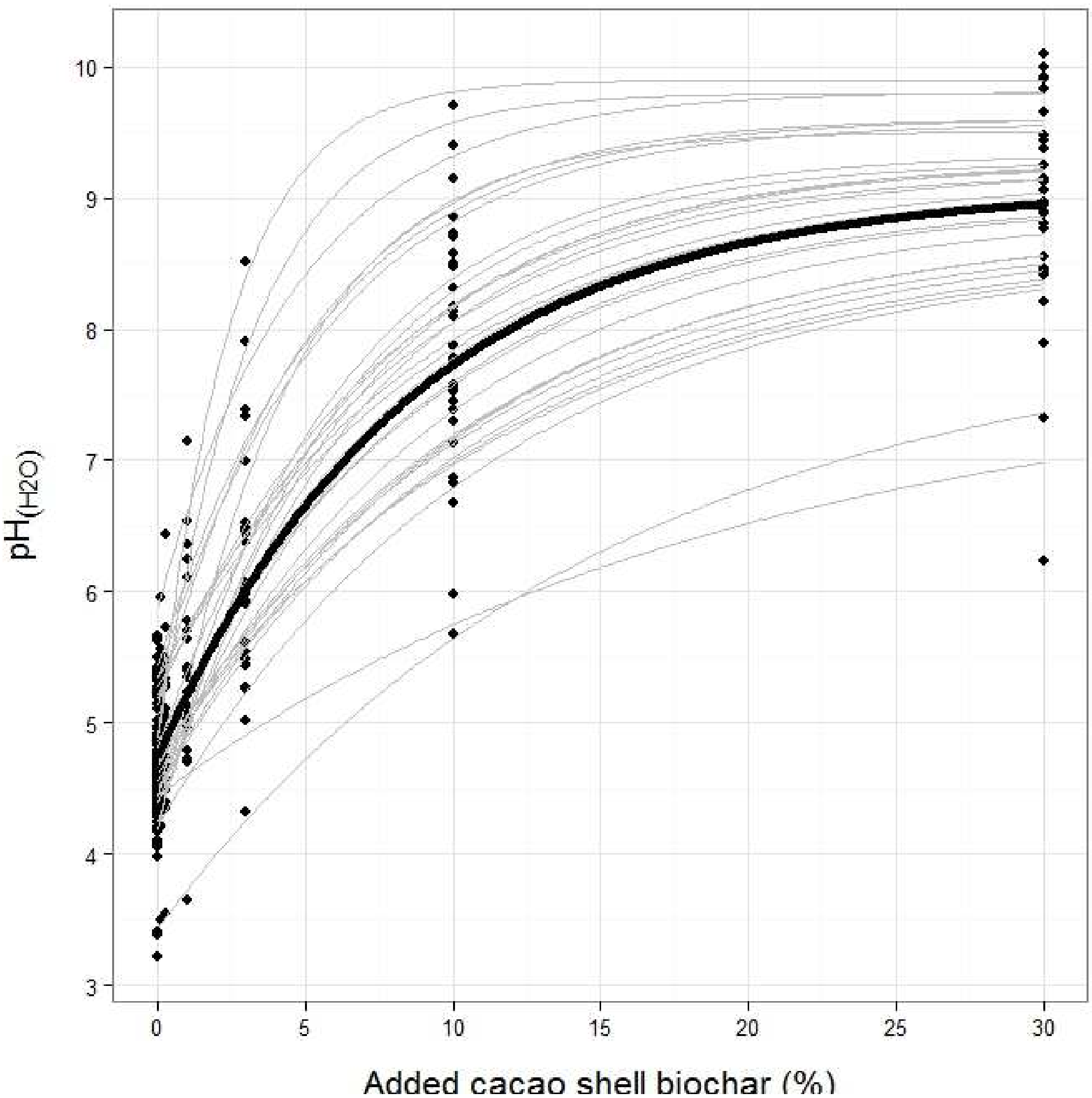


Fig. 2 A.

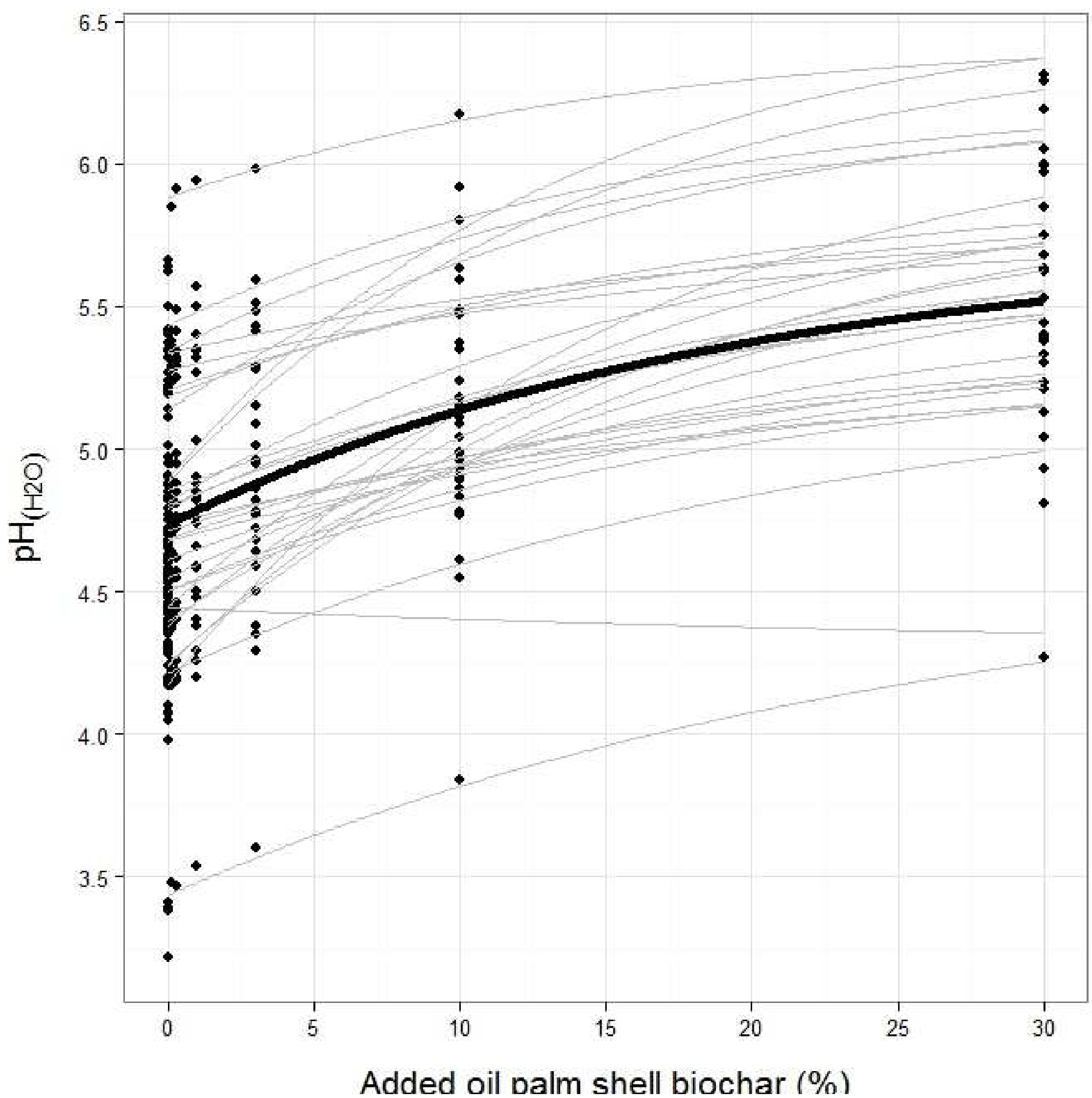

6.5
6.0
5.5
5.0
4.5
4.0
3.5
pH(H2O)
0
5
10
15
20
25
30
Added oil palm shell biochar (%)

Fig. 2 B.

3

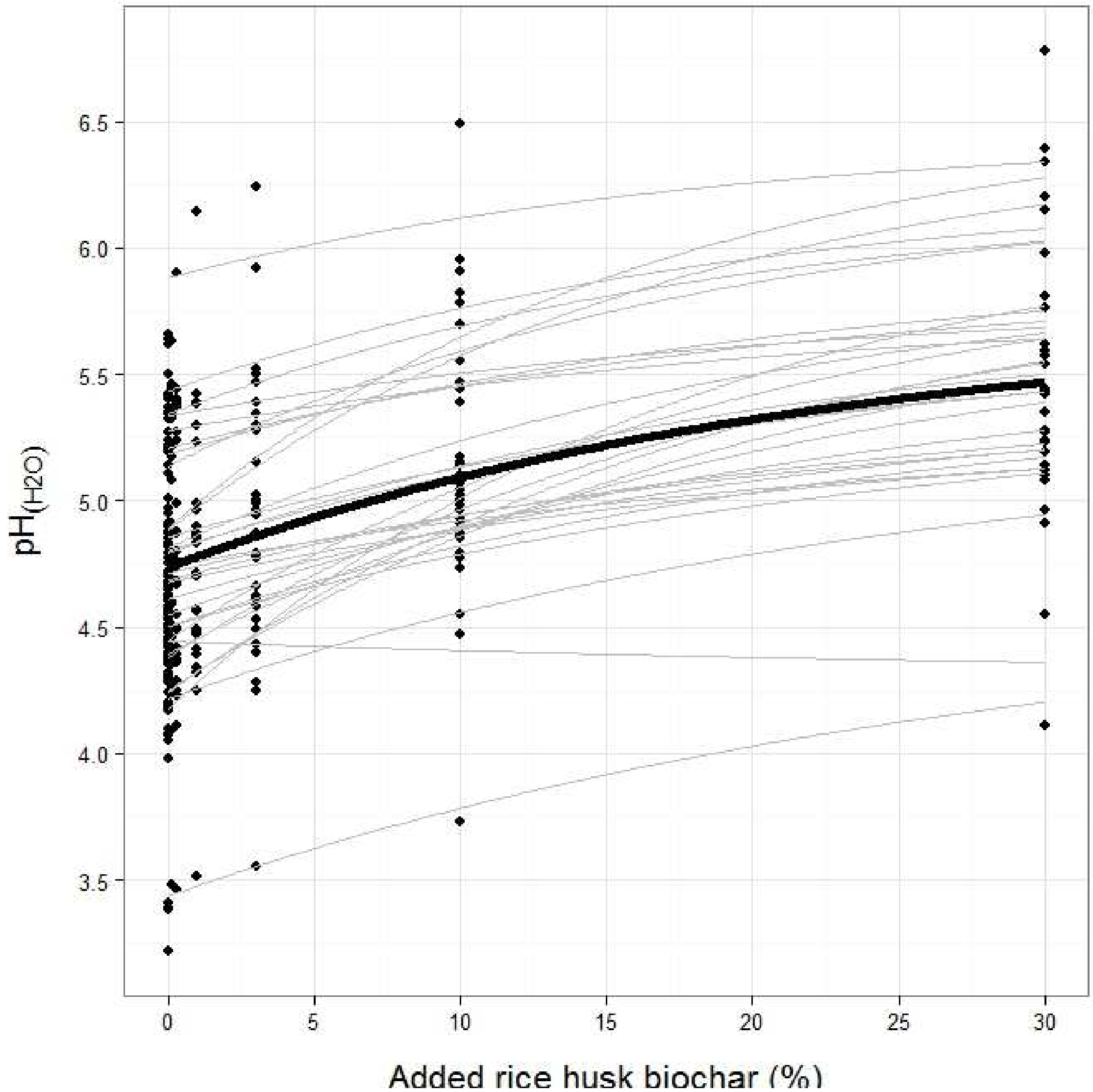
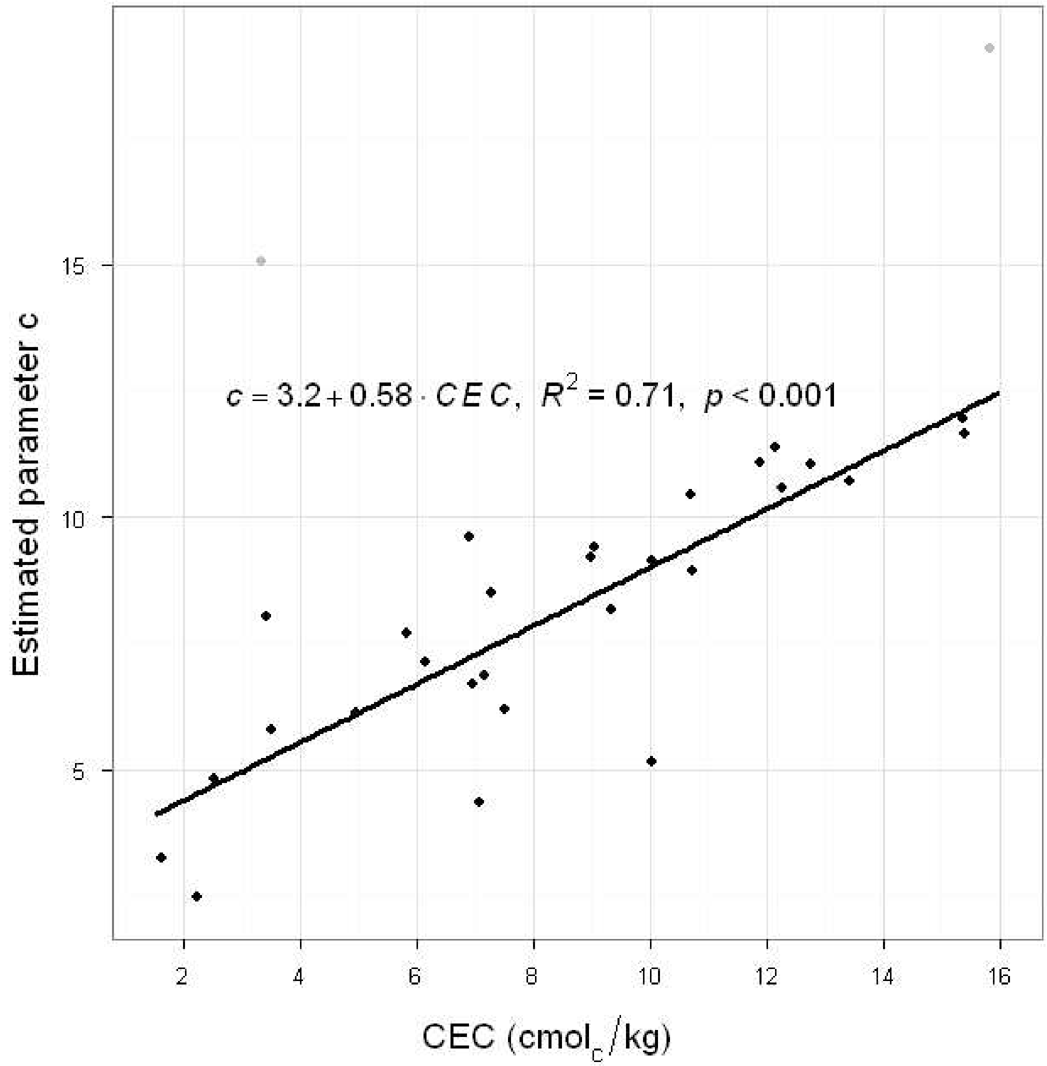

pH(H2O)
6.5
6.0
5.5
5.0
4.5
4.0
3.5
0
5
10
15
20
25
30
Added rice husk biochar (%)

Fig. 2 C.



$c = 3.2 + 0.58 \cdot CEC$, $R^2 = 0.71$, $p < 0.001$
Estimated parameter c
CEC (cmol$_c$/kg)
15
10
5
2
4
6
8
10
12
14
16

Fig. 3.

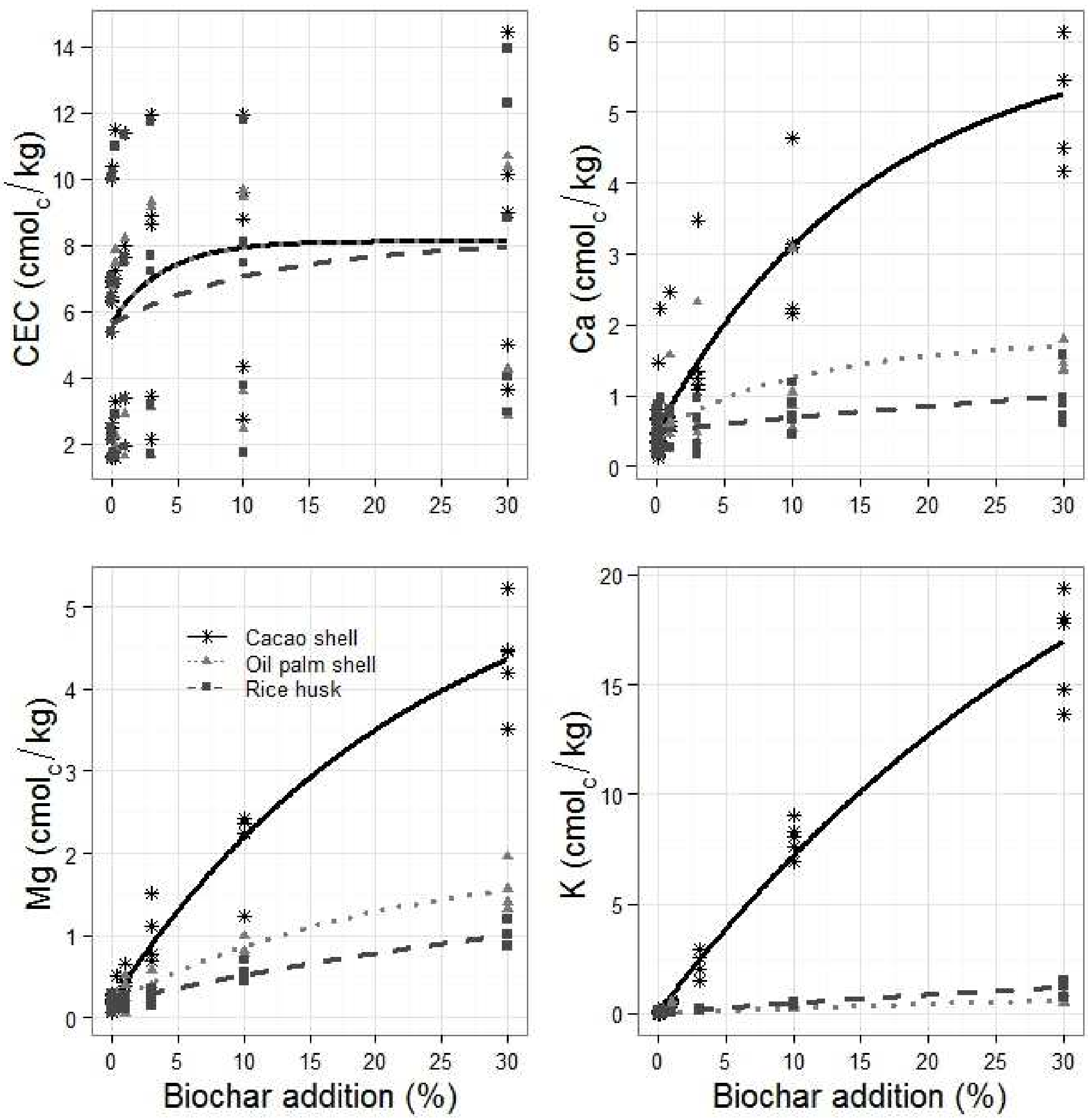

CEC (cmol$_c$/kg)
Ca (cmol$_c$/kg)
Mg (cmol$_c$/kg)
K (cmol$_c$/kg)
Cacao shell
Oil palm shell
Rice husk
Biochar addition (%)
Biochar addition (%)

Fig. 4.

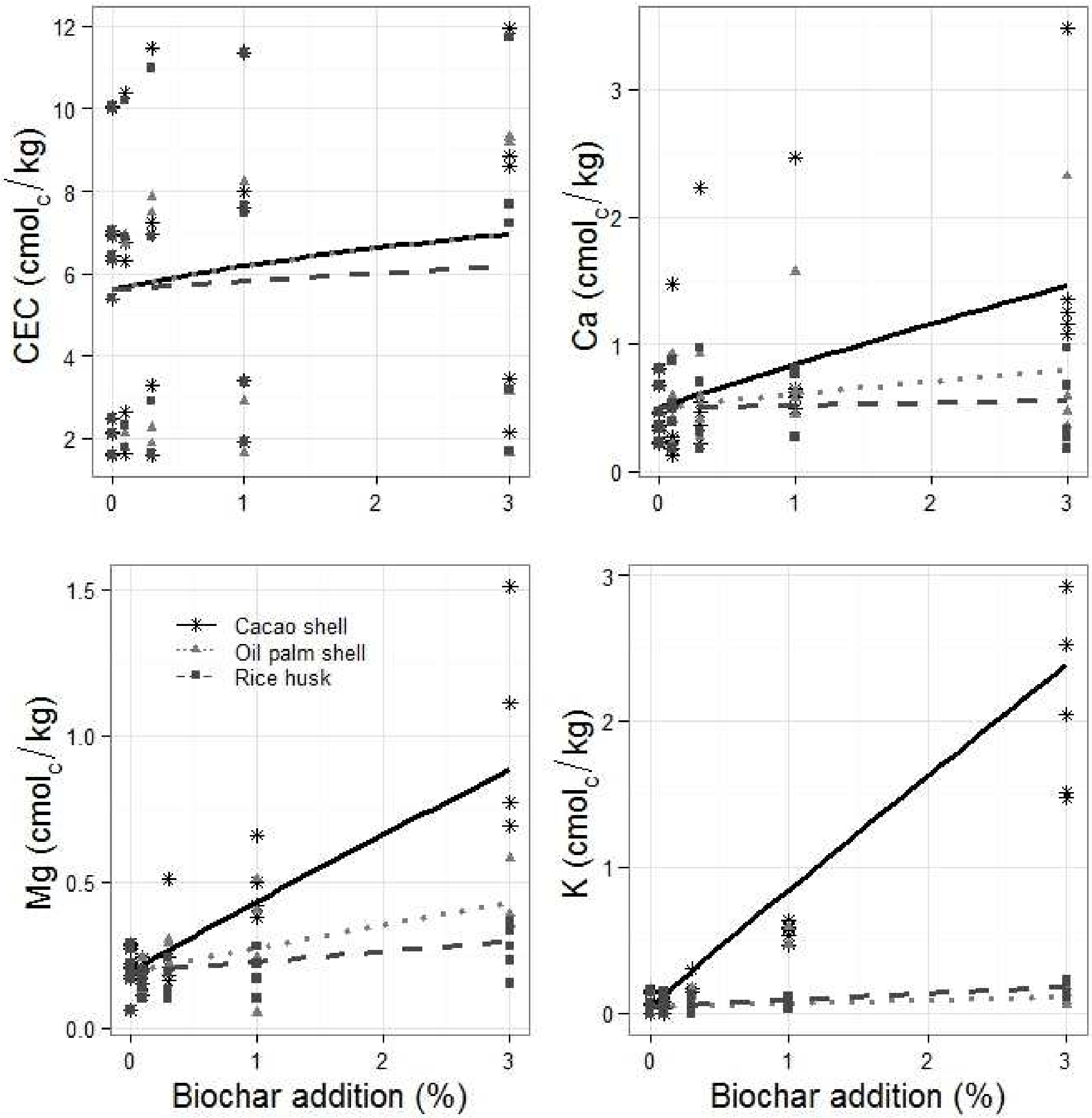
Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): "pH effects of the addition of three biochars to acidic Indonesian mineral soils"

Supporting information

Figure S1. Locally constructed unit for biochar production.

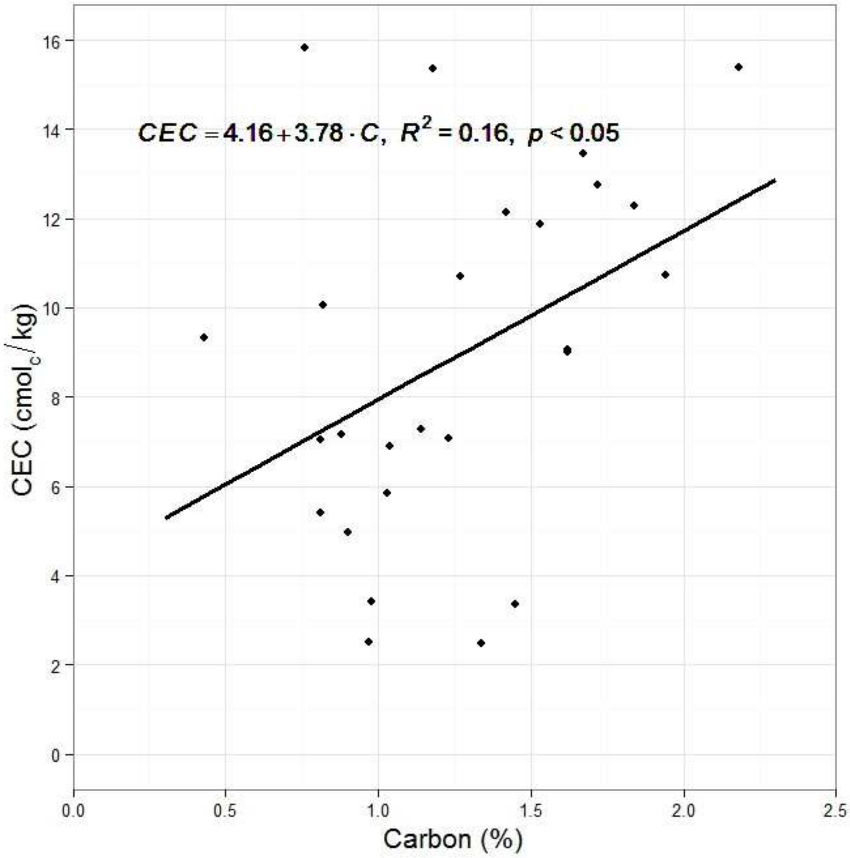
Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): "pH effects of the addition of three biochars to acidic Indonesian mineral soils"

Figure S2. Predicted response in soil CEC ($cmol_c$/kg) and of available Ca, Mg and K ($cmol_c$/kg) to added cacao shell BC, oil palm shell BC and rice husk BC in soils from 5 sites (site 18, 19, 24, 30, 31; Table 1) across Indonesia. The fitted curves derive from the model $y = a + b*(1-\exp^{(-x/c)})$ (eq. 1) based on restricted maximum likelihood estimates (Table 3) for the mean response in CEC and levels of cations (Ca, Mg and K) for 0-30% BC. Note: Predicted responses are presented for 0-3% only. For the full dataset, see Figure 4.



Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): "pH effects of the addition of three biochars to acidic Indonesian mineral soils"

Figure S3. Relationship between CEC ($cmol_c\ kg^{-1}$ soil) and the content of organic carbon (%) from 26 sites across Indonesia.



Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): "pH effects of the addition of three biochars to acidic Indonesian mineral soils"

Table S1. Selected attributes of biochars produced of cacao shell, oil palm shell and rice husk at pyrolysis times of 1, 2 and 3.5 hours. The pyrolysis times used for the different biochars are highlighted (bold).

Biochar type	Pyrolysis time (h)	BC yield (%)	Ash (%)	Water (%)	Total N (%)	Total P (%)	Total K (%)	Total Ca (%)	Total Mg (%)
Cacao shell (raw)	0	-	-	-	1.8	0.40	0.50	-	-
Cacao shell	1	18.7	27.9	18	1.1	0.40	1.10	1.29	0.85
Cacao shell	2	18	28.8	20	1.0	0.40	1.20	1.44	0.98
Cacao shell	**3.5**	**22**	**27.4**	**12**	**1.4**	**0.36**	**1.25**	**1.30**	**0.86**
Oil palm shell (raw)	0	-	-	-	1.1	0.10	<0.1	-	-
Oil palm shell	**1**	**53.5**	**26.1**	**5**	**1.7**	**0.25**	**<0.1**	**0.67**	**0.31**
Oil palm shell	2	45.6	40.9	3	1.3	0.40	<0.1	1.00	0.55
Oil palm shell	3.5	48.6	26.4	6	1.0	0.25	<0.1	0.66	0.31
Rice husk (raw)	0	-	-	-	0.8	0.15	<0.1	-	-
Rice husk	1	23.3	53.4	2	0.6	0.17	<0.1	0.31	0.13
Rice husk	2	23.3	47.0	3	0.6	0.10	<0.1	0.13	0.07
Rice husk	**3.5**	**30.4**	**48.9**	**3**	**1.0**	**0.21**	**<0.1**	**0.21**	**0.13**

Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): "pH effects of the addition of three biochars to acidic Indonesian mineral soils"

Table S2. Measured soil attributes in soils from 31 sites across Indonesia (Table 1) with and without addition of biochar (BC).

Site ID	BC type	BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
1		0	4.9	3.7					15.37			1.18	0.12	9.8
1	Rice husk	0	4.65	3.82										
1	Rice husk	0.1	4.69	3.83										
1	Rice husk	0.3	4.74	3.81										
1	Rice husk	1	4.71	3.82										
1	Rice husk	3	4.96	3.85										
1	Rice husk	10	4.91	3.89										
1	Rice husk	30	5.27	4.04										
1	Oil palm shell	0	4.55	3.83										
1	Oil palm shell	0.1	4.8	3.84										
1	Oil palm shell	0.3	4.72	3.83										
1	Oil palm shell	1	4.82	3.81										
1	Oil palm shell	3	4.77	3.86										
1	Oil palm shell	10	4.77	3.91										
1	Oil palm shell	30	5.04	4.07										
1	Cacao shell	0	4.67	3.82										
1	Cacao shell	0.1	4.86	3.85										
1	Cacao shell	0.3	4.96	3.87										
1	Cacao shell	1	4.96	3.96										
1	Cacao shell	3	5.61	4.39										
1	Cacao shell	10	7.39	6.48										
1	Cacao shell	30	7.89	7.74										
2		0	4.31	3.71					13.44			1.67	0.12	13.9
2	Rice husk	0	4.51	3.8										
2	Rice husk	0.1	4.78	3.82										
2	Rice husk	0.3	4.8	3.83										
2	Rice husk	1	4.85	3.82										
2	Rice husk	3	5.02	3.82										
2	Rice husk	10	4.96	3.87										
2	Rice husk	30	5.08	3.94										
2	Oil palm shell	0	4.61	3.8										
2	Oil palm shell	0.1	4.73	3.81										
2	Oil palm shell	0.3	4.76	3.81										
2	Oil palm shell	1	4.74	3.85										
2	Oil palm shell	3	4.82	3.86										
2	Oil palm shell	10	4.99	3.93										
2	Oil palm shell	30	5.3	4.17										
2	Cacao shell	0	4.54	3.78										
2	Cacao shell	0.1	4.81	3.83										
2	Cacao shell	0.3	4.86	3.86										
2	Cacao shell	1	5.03	3.92										
2	Cacao shell	3	5.53	4.12										
2	Cacao shell	10	7.55	6.39										
2	Cacao shell	30	8.21	7.79										
3		0	5.11	3.86					9.33			0.43	0.03	13.9
3	Rice husk	0	5.37	3.91										
3	Rice husk	0.1	5.32	3.9										
3	Rice husk	0.3	5.27	3.91										
3	Rice husk	1	5.3	3.95										
3	Rice husk	3	5.34	3.97										
3	Rice husk	10	5.44	3.97										
3	Rice husk	30	5.62	4.05										
3	Oil palm shell	0	5.27	3.91										
3	Oil palm shell	0.1	5.29	3.94										
3	Oil palm shell	0.3	5.31	3.93										

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Site ID		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^+	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
3	Oil palm shell	1	5.32	3.94										
3	Oil palm shell	3	5.29	3.93										
3	Oil palm shell	10	5.35	3.97										
3	Oil palm shell	30	5.23	4.13										
3	Cacao shell	0	5.37	3.95										
3	Cacao shell	0.1	5.27	3.95										
3	Cacao shell	0.3	5.27	3.98										
3	Cacao shell	1	5.35	4.08										
3	Cacao shell	3	6.05	4.37										
3	Cacao shell	10	8.58	7.61										
3	Cacao shell	30	9.25	8.55										
4		0	4.4	3.68					15.39			2.18	0.15	14.7
4	Rice husk	0	4.6	3.9										
4	Rice husk	0.1	4.59	3.93										
4	Rice husk	0.3	4.68	3.94										
4	Rice husk	1	4.7	3.95										
4	Rice husk	3	4.79	3.98										
4	Rice husk	10	4.93	3.96										
4	Rice husk	30	5.08	3.99										
4	Oil palm shell	0	4.63	3.91										
4	Oil palm shell	0.1	4.7	3.94										
4	Oil palm shell	0.3	4.73	3.93										
4	Oil palm shell	1	4.76	3.94										
4	Oil palm shell	3	4.82	3.96										
4	Oil palm shell	10	4.98	4.02										
4	Oil palm shell	30	5.23	4.19										
4	Cacao shell	0	4.68	3.92										
4	Cacao shell	0.1	4.81	3.93										
4	Cacao shell	0.3	4.88	3.97										
4	Cacao shell	1	5.13	4.04										
4	Cacao shell	3	5.53	4.2										
4	Cacao shell	10	6.83	5.75										
4	Cacao shell	30	8.44	7.55										
5		0	4.54	3.85					9			1.62	0.08	19.4
5	Rice husk	0	4.72	4.01										
5	Rice husk	0.1	4.77	4.08										
5	Rice husk	0.3	4.76	4.1										
5	Rice husk	1	4.86	4.1										
5	Rice husk	3	5	4.12										
5	Rice husk	10	5.15	4.14										
5	Rice husk	30	5.24	4.15										
5	Oil palm shell	0	4.7	4										
5	Oil palm shell	0.1	4.82	4.08										
5	Oil palm shell	0.3	4.85	4.08										
5	Oil palm shell	1	4.88	4.11										
5	Oil palm shell	3	5.01	4.14										
5	Oil palm shell	10	5.18	4.27										
5	Oil palm shell	30	5.62	4.76										
5	Cacao shell	0	4.77	4.08										
5	Cacao shell	0.1	4.92	4.09										
5	Cacao shell	0.3	5.04	4.14										
5	Cacao shell	1	5.41	4.26										
5	Cacao shell	3	5.93	4.68										
5	Cacao shell	10	7.58	6.69										
5	Cacao shell	30	8.81	8.06										
6		0	4.29	3.73					12.76			1.72	0.09	20.2

Martinsen, V., Alling, V., Nurida, N.L., Mulder, J., Hale, S.E., Ritz, C., Rutherford D.W., Heikens, A. , Breedveld, G.D. and Cornelissen, G., 2015 (Soil Science and Plant Nutrition): “pH effects of the addition of three biochars to acidic Indonesian mineral soils”

Site ID		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
6	Rice husk	0	4.39	3.96										
6	Rice husk	0.1	4.35	3.95										
6	Rice husk	0.3	4.36	3.96										
6	Rice husk	1	4.41	3.95										
6	Rice husk	3	4.49	3.97										
6	Rice husk	10	4.73	3.98										
6	Rice husk	30	5.28	4										
6	Oil palm shell	0	4.36	3.94										
6	Oil palm shell	0.1	4.42	3.95										
6	Oil palm shell	0.3	4.81	3.96										
6	Oil palm shell	1	4.83	3.95										
6	Oil palm shell	3	4.86	3.99										
6	Oil palm shell	10	4.89	4.02										
6	Oil palm shell	30	5.33	4.16										
6	Cacao shell	0	4.38	3.96										
6	Cacao shell	0.1	4.52	3.97										
6	Cacao shell	0.3	4.84	3.99										
6	Cacao shell	1	5.21	4.06										
6	Cacao shell	3	5.26	4.21										
6	Cacao shell	10	6.87	5.48										
6	Cacao shell	30	8.41	7.8										
7		0	4.55	3.84					7.29			1.14	0.05	21.6
7	Rice husk	0	4.79	3.98										
7	Rice husk	0.1	4.79	3.99										
7	Rice husk	0.3	4.8	3.99										
7	Rice husk	1	4.87	3.98										
7	Rice husk	3	4.96	3.99										
7	Rice husk	10	5.14	4.03										
7	Rice husk	30	5.43	4.02										
7	Oil palm shell	0	4.77	3.98										
7	Oil palm shell	0.1	4.76	3.99										
7	Oil palm shell	0.3	4.81	3.98										
7	Oil palm shell	1	4.85	4.01										
7	Oil palm shell	3	4.95	4.01										
7	Oil palm shell	10	5.11	4.09										
7	Oil palm shell	30	5.63	4.5										
7	Cacao shell	0	4.82	3.98										
7	Cacao shell	0.1	4.93	3.99										
7	Cacao shell	0.3	5.06	4.04										
7	Cacao shell	1	5.4	4.16										
7	Cacao shell	3	5.92	4.42										
7	Cacao shell	10	7.78	6.74										
7	Cacao shell	30	9.06	7.99										
8		0	3.98	3.72					12.15			1.42	0.07	21
8	Rice husk	0	4.29	3.84										
8	Rice husk	0.1	4.28	3.83										
8	Rice husk	0.3	4.29	3.82										
8	Rice husk	1	4.32	3.85										
8	Rice husk	3	4.25	3.84										
8	Rice husk	10	4.55	3.86										
8	Rice husk	30	4.91	4.05										
8	Oil palm shell	0	4.2	3.82										
8	Oil palm shell	0.1	4.17	3.82										
8	Oil palm shell	0.3	4.19	3.83										
8	Oil palm shell	1	4.2	3.83										
8	Oil palm shell	3	4.29	3.84										

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Site ID		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
8	Oil palm shell	10	4.55	3.89										
8	Oil palm shell	30	4.93	4.01										
8	Cacao shell	0	4.3	3.84										
8	Cacao shell	0.1	4.32	3.84										
8	Cacao shell	0.3	4.35	3.85										
8	Cacao shell	1	4.7	3.91										
8	Cacao shell	3	5.27	4.07										
8	Cacao shell	10	6.68	5.45										
8	Cacao shell	30	8.45	7.5										
9		0	4.32	3.58					15.83			0.76	0.03	25.9
9	Rice husk	0	4.36	3.61										
9	Rice husk	0.1	4.35	3.68										
9	Rice husk	0.3	4.37	3.68										
9	Rice husk	1	4.39	3.69										
9	Rice husk	3	4.4	3.67										
9	Rice husk	10	4.47	3.71										
9	Rice husk	30	4.55	3.74										
9	Oil palm shell	0	4.37	3.61										
9	Oil palm shell	0.1	4.37	3.67										
9	Oil palm shell	0.3	4.4	3.69										
9	Oil palm shell	1	4.4	3.68										
9	Oil palm shell	3	4.5	3.69										
9	Oil palm shell	10	4.61	3.72										
9	Oil palm shell	30	4.81	3.84										
9	Cacao shell	0	4.36	3.61										
9	Cacao shell	0.1	4.5	3.68										
9	Cacao shell	0.3	4.57	3.72										
9	Cacao shell	1	4.72	3.75										
9	Cacao shell	3	5.02	3.88										
9	Cacao shell	10	5.98	4.39										
9	Cacao shell	30	6.23	5.24										
10		0	4.49	3.76					10.69			1.27	0.06	20.4
10	Rice husk	0	4.5	3.91										
10	Rice husk	0.1	4.51	3.89										
10	Rice husk	0.3	4.49	3.92										
10	Rice husk	1	4.57	3.88										
10	Rice husk	3	4.62	3.9										
10	Rice husk	10	4.77	3.93										
10	Rice husk	30	5.1	3.92										
10	Oil palm shell	0	4.51	3.9										
10	Oil palm shell	0.1	4.55	3.88										
10	Oil palm shell	0.3	4.55	3.9										
10	Oil palm shell	1	4.59	3.9										
10	Oil palm shell	3	4.64	3.91										
10	Oil palm shell	10	4.78	3.96										
10	Oil palm shell	30	5.21	4.2										
10	Cacao shell	0	4.53	3.92										
10	Cacao shell	0.1	4.61	3.89										
10	Cacao shell	0.3	4.68	3.9										
10	Cacao shell	1	4.79	3.95										
10	Cacao shell	3	5.43	4.21										
10	Cacao shell	10	7.13	6.17										
10	Cacao shell	30	8.77	7.7										
11		0	4.7	3.73					6.9			1.04	0.04	23.5
11	Rice husk	0	4.54	3.84										
11	Rice husk	0.1	4.55	3.85										

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Site ID		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^+	C	N	CN
		(%)					cmol$_c$/kg soil					(%)	(%)	
11	Rice husk	0.3	4.55	3.84										
11	Rice husk	1	4.56	3.85										
11	Rice husk	3	4.61	3.83										
11	Rice husk	10	4.86	3.86										
11	Rice husk	30	5.12	3.9										
11	Oil palm shell	0	4.55	3.84										
11	Oil palm shell	0.1	4.54	3.84										
11	Oil palm shell	0.3	4.57	3.83										
11	Oil palm shell	1	4.58	3.84										
11	Oil palm shell	3	4.68	3.86										
11	Oil palm shell	10	5.04	3.93										
11	Oil palm shell	30	5.13	4.08										
11	Cacao shell	0	4.54	3.84										
11	Cacao shell	0.1	4.49	3.86										
11	Cacao shell	0.3	4.67	3.85										
11	Cacao shell	1	5.23	4.03										
11	Cacao shell	3	5.54	4.22										
11	Cacao shell	10	7.45	6.66										
11	Cacao shell	30	8.88	8.26										
12		0	3.22	3.04					3.34			1.45	0.07	19.8
12	Rice husk	0	3.38	3.12										
12	Rice husk	0.1	3.48	3.25										
12	Rice husk	0.3	3.46	3.26										
12	Rice husk	1	3.51	3.26										
12	Rice husk	3	3.55	3.26										
12	Rice husk	10	3.73	3.36										
12	Rice husk	30	4.11	3.52										
12	Oil palm shell	0	3.39	3.12										
12	Oil palm shell	0.1	3.48	3.24										
12	Oil palm shell	0.3	3.47	3.24										
12	Oil palm shell	1	3.54	3.24										
12	Oil palm shell	3	3.6	3.36										
12	Oil palm shell	10	3.84	3.45										
12	Oil palm shell	30	4.27	3.66										
12	Cacao shell	0	3.41	3.12										
12	Cacao shell	0.1	3.49	3.27										
12	Cacao shell	0.3	3.55	3.33										
12	Cacao shell	1	3.65	3.39										
12	Cacao shell	3	4.32	3.66										
12	Cacao shell	10	5.67	4.66										
12	Cacao shell	30	7.32	6.6										
13		0	5.66	4.67					2.51			0.97	0.02	40.6
13	Rice husk	0	5.5	4.78										
13	Rice husk	0.1	5.63	4.71										
13	Rice husk	0.3	5.9	4.73										
13	Rice husk	1	6.14	4.77										
13	Rice husk	3	6.24	4.82										
13	Rice husk	10	6.49	5.08										
13	Rice husk	30	6.78	5.46										
13	Oil palm shell	0	5.62	4.78										
13	Oil palm shell	0.1	5.85	4.73										
13	Oil palm shell	0.3	5.91	4.73										
13	Oil palm shell	1	5.94	4.78										
13	Oil palm shell	3	5.98	5										
13	Oil palm shell	10	6.17	5.34										
13	Oil palm shell	30	6.29	5.5										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
13	Cacao shell	0	5.64	4.78										
13	Cacao shell	0.1	5.96	4.85										
13	Cacao shell	0.3	6.43	5.11										
13	Cacao shell	1	7.15	5.86										
13	Cacao shell	3	7.9	6.89										
13	Cacao shell	10	8.73	8.03										
13	Cacao shell	30	9.44	8.72										
14		0	4.85	3.84					5.83			1.03	0.03	33.4
14	Rice husk	0	5.2	3.96										
14	Rice husk	0.1	5.21	3.98										
14	Rice husk	0.3	5.24	3.98										
14	Rice husk	1	5.23	3.99										
14	Rice husk	3	5.28	4.01										
14	Rice husk	10	5.47	4.05										
14	Rice husk	30	5.59	4.18										
14	Oil palm shell	0	5.21	3.96										
14	Oil palm shell	0.1	5.24	3.96										
14	Oil palm shell	0.3	5.29	3.98										
14	Oil palm shell	1	5.27	3.97										
14	Oil palm shell	3	5.28	4.01										
14	Oil palm shell	10	5.37	4.13										
14	Oil palm shell	30	5.75	4.7										
14	Cacao shell	0	5.22	3.97										
14	Cacao shell	0.1	5.25	3.97										
14	Cacao shell	0.3	5.46	4.03										
14	Cacao shell	1	5.78	4.19										
14	Cacao shell	3	6.46	4.85										
14	Cacao shell	10	8.17	7.2										
14	Cacao shell	30	9.38	8.33										
15		0	4.07	3.77					7.08			1.23	0.06	20.9
15	Rice husk	0	4.18	3.83										
15	Rice husk	0.1	4.24	3.84										
15	Rice husk	0.3	4.23	3.84										
15	Rice husk	1	4.34	3.85										
15	Rice husk	3	4.43	3.91										
15	Rice husk	10	5.04	4										
15	Rice husk	30	5.54	4.4										
15	Oil palm shell	0	4.17	3.81										
15	Oil palm shell	0.1	4.25	3.84										
15	Oil palm shell	0.3	4.22	3.83										
15	Oil palm shell	1	4.29	3.84										
15	Oil palm shell	3	4.35	3.92										
15	Oil palm shell	10	5.15	4.14										
15	Oil palm shell	30	5.85	4.88										
15	Cacao shell	0	4.2	3.84										
15	Cacao shell	0.1	4.22	3.87										
15	Cacao shell	0.3	4.49	3.95										
15	Cacao shell	1	5.35	4.22										
15	Cacao shell	3	6.99	5.61										
15	Cacao shell	10	8.85	8.1										
15	Cacao shell	30	9.66	8.33										
16		0	4.19	3.92					7.15			0.88	0.04	25
16	Rice husk	0	4.05	4.08										
16	Rice husk	0.1	4.09	4.07										
16	Rice husk	0.3	4.11	4.05										
16	Rice husk	1	4.49	4.05										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^+	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
16	Rice husk	3	4.78	4.05										
16	Rice husk	10	5	4.15										
16	Rice husk	30	5.44	4.46										
16	Oil palm shell	0	4.08	4.05										
16	Oil palm shell	0.1	4.17	4.03										
16	Oil palm shell	0.3	4.26	4.03										
16	Oil palm shell	1	4.38	4.04										
16	Oil palm shell	3	4.72	4.07										
16	Oil palm shell	10	4.93	4.22										
16	Oil palm shell	30	5.62	4.86										
16	Cacao shell	0	4.1	4.08										
16	Cacao shell	0.1	4.2	4.07										
16	Cacao shell	0.3	4.48	4.1										
16	Cacao shell	1	5.03	4.27										
16	Cacao shell	3	6.44	5.28										
16	Cacao shell	10	7.76	7.1										
16	Cacao shell	30	8.9	8.02										
17		0	4.61	3.72					9.05			1.62	0.09	17.6
17	Rice husk	0	4.87	3.82										
17	Rice husk	0.1	4.88	3.83										
17	Rice husk	0.3	4.87	3.82										
17	Rice husk	1	4.85	3.84										
17	Rice husk	3	4.94	3.85										
17	Rice husk	10	5.1	3.95										
17	Rice husk	30	5.42	4.08										
17	Oil palm shell	0	4.85	3.8										
17	Oil palm shell	0.1	4.88	3.82										
17	Oil palm shell	0.3	4.88	3.82										
17	Oil palm shell	1	4.9	3.83										
17	Oil palm shell	3	4.96	3.88										
17	Oil palm shell	10	5.09	3.97										
17	Oil palm shell	30	5.53	4.73										
17	Cacao shell	0	4.87	3.89										
17	Cacao shell	0.1	5.03	3.88										
17	Cacao shell	0.3	5.03	3.87										
17	Cacao shell	1	5.35	4.03										
17	Cacao shell	3	6.07	4.52										
17	Cacao shell	10	7.52	6.64										
17	Cacao shell	30	8.8	7.84										
18		0	4.48	3.86					7.04			0.81	0.04	20.8
18	Rice husk	0	4.42	3.92	0.81	0.28	0.07	0.07	6.93	4.12	0.24			
18	Rice husk	0.1	4.46	3.92	0.87	0.2	0.1	0.05	6.89	3.78	0.34			
18	Rice husk	0.3	4.42	3.91	0.97	0.24	0.1	0.05	6.89	3.81	0.16			
18	Rice husk	1	4.41	3.91	0.81	0.28	0.1	0.02	7.47	4.02	0.26			
18	Rice husk	3	4.53	3.93	0.97	0.36	0.23	0.07	7.22	3.18	0.32			
18	Rice husk	10	4.85	3.98	1.19	0.7	0.54	0.14	8.1	2.2	0.17			
18	Rice husk	30	5.19	4.18	1.58	1.19	1.48	0.16	8.82	0.67	0.21			
18	Oil palm shell	0	4.43	3.92	0.8	0.29	0.07	0.07	6.94	4.11	0.23			
18	Oil palm shell	0.1	4.38	3.91	0.92	0.2	0.1	0.07	6.93	3.55	0.24			
18	Oil palm shell	0.3	4.46	3.91	0.92	0.21	0.13	0.07	7.47	3.21	0.28			
18	Oil palm shell	1	4.5	3.92	1.57	0.24	0.07	0.07	7.51	3.53	0.24			
18	Oil palm shell	3	4.68	3.94	2.32	0.58	0.18	0.1	9.3	3.13	0.24			
18	Oil palm shell	10	4.86	4.03	3.08	0.99	0.37	0.09	9.65	1.83	0.21			
18	Oil palm shell	30	5.44	4.41	1.37	1.96	0.88	0.12	10.36	0.31	0.25			
18	Cacao shell	0	4.4	3.91	0.8	0.27	0.06	0.06	6.9	4.1	0.23			
18	Cacao shell	0.1	4.42	3.92	1.47	0.2	0.13	0.05	6.3	3.49	0.32			

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			$cmol_c$/kg soil							(%)	(%)	
18	Cacao shell	0.3	4.66	3.95	2.23	0.51	0.3	0.06	6.97	3.63	0.23			
18	Cacao shell	1	4.97	4.05	2.47	0.66	0.64	0.05	7.99	2.05	0.16			
18	Cacao shell	3	5.9	4.7	3.48	1.51	2.52	0.1	8.62	0	0.02			
18	Cacao shell	10	8.31	7.24	4.64	2.37	7.62	0.14	8.81	0	0.02			
18	Cacao shell	30	9.91	8.96	0.59	4.46	19.38	0.3	8.99	0	0			
19		0	4.58	4.1					2.48			1.34	0.09	14.3
19	Rice husk	0	4.97	4.27	0.47	0.22	0.15	0.05	2.14	1.18	0.2			
19	Rice husk	0.1	5.08	4.31	0.53	0.14	0.15	0.1	2.32	1.01	0.18			
19	Rice husk	0.3	4.99	4.33	0.7	0.13	0.12	0.05	2.9	0.57	0.17			
19	Rice husk	1	4.99	4.33	0.77	0.17	0.11	0.05	3.37	0.86	0.16			
19	Rice husk	3	5.15	4.33	0.68	0.33	0.21	0.07	3.19	0.52	0.23			
19	Rice husk	10	5.82	4.64	0.89	0.51	0.51	0.05	3.77	0	0.02			
19	Rice husk	30	6.39	5.21	0.61	1.01	1.24	0.1	4.01	0	0.07			
19	Oil palm shell	0	4.95	4.25	0.46	0.21	0.16	0.05	2.13	1.17	0.21			
19	Oil palm shell	0.1	4.95	4.26	0.59	0.13	0.14	0.05	2.11	1.03	0.16			
19	Oil palm shell	0.3	4.98	4.27	0.59	0.2	0.11	0.05	2.25	1.35	0.16			
19	Oil palm shell	1	5.03	4.38	0.45	0.05	0.07	0.05	2.91	0.5	0.17			
19	Oil palm shell	3	5.15	4.46	0.47	0.36	0.14	0.02	3.12	0.24	0.15			
19	Oil palm shell	10	5.8	4.8	0.53	3.54	0.22	0.05	3.59	0	0.09			
19	Oil palm shell	30	6.31	5.36	1.46	1.32	0.47	0.05	4.25	0	0.02			
19	Cacao shell	0	4.97	4.29	0.46	0.22	0.14	0.05	2.12	1.16	0.2			
19	Cacao shell	0.1	5	4.32	0.25	0.11	0.08	0.03	2.65	0.84	0.17			
19	Cacao shell	0.3	5.1	4.41	0.47	0.16	0.15	0.05	3.28	0.51	0.16			
19	Cacao shell	1	6.54	5.24	0.65	0.38	0.47	0.11	3.39	0	0.06			
19	Cacao shell	3	8.51	7.17	1.25	0.69	1.48	0.1	3.46	0	0			
19	Cacao shell	10	9.71	8.89	2.16	1.24	8.28	0.14	4.33	0	0			
19	Cacao shell	30	10.1	9.45	5.45	3.51	14.76	0.28	5.01	0	0			
20		0										1.54	0.11	13.7
20	Rice husk	0	5.41	4.32										
20	Rice husk	0.1	5.45	4.33										
20	Rice husk	0.3	5.44	4.36										
20	Rice husk	1	5.42	4.42										
20	Rice husk	3	5.5	4.4										
20	Rice husk	10	5.91	4.89										
20	Rice husk	30	6.34	5.25										
20	Oil palm shell	0	5.4	4.32										
20	Oil palm shell	0.1	5.41	4.28										
20	Oil palm shell	0.3	5.49	4.32										
20	Oil palm shell	1	5.57	4.48										
20	Oil palm shell	3	5.59	4.92										
20	Oil palm shell	10	5.63	5.18										
20	Oil palm shell	30	5.97	5.34										
20	Cacao shell	0	5.42	4.3										
20	Cacao shell	0.1	5.48	4.47										
20	Cacao shell	0.3	5.5	4.73										
20	Cacao shell	1	6.36	5.62										
20	Cacao shell	3	7.33	6.78										
20	Cacao shell	10	8.7	8.05										
20	Cacao shell	30	9.48	8.82										
21		0	4.45	3.7					11.88			1.53	0.11	14
21	Rice husk	0	4.75	3.81										
21	Rice husk	0.1	4.76	3.81										
21	Rice husk	0.3	4.74	3.79										
21	Rice husk	1	4.83	3.82										
21	Rice husk	3	4.87	3.86										
21	Rice husk	10	4.98	3.85										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^+	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
21	Rice husk	30	5.14	3.96										
21	Oil palm shell	0	4.71	3.81										
21	Oil palm shell	0.1	4.74	3.8										
21	Oil palm shell	0.3	4.76	3.8										
21	Oil palm shell	1	4.78	3.81										
21	Oil palm shell	3	4.78	3.87										
21	Oil palm shell	10	4.96	3.98										
21	Oil palm shell	30	5.39	4.35										
21	Cacao shell	0	4.72	3.8										
21	Cacao shell	0.1	4.73	3.81										
21	Cacao shell	0.3	4.82	3.84										
21	Cacao shell	1	5.13	3.93										
21	Cacao shell	3	5.49	4.1										
21	Cacao shell	10	7.14	6.2										
21	Cacao shell	30	8.46	7.66										
22		0	4.66	3.8					4.97			0.9	0.05	19.6
22	Rice husk	0	4.36	3.92										
22	Rice husk	0.1	4.36	3.88										
22	Rice husk	0.3	4.39	3.88										
22	Rice husk	1	4.47	3.9										
22	Rice husk	3	4.66	3.95										
22	Rice husk	10	5.07	4.09										
22	Rice husk	30	5.81	4.62										
22	Oil palm shell	0	4.35	3.91										
22	Oil palm shell	0.1	4.42	3.87										
22	Oil palm shell	0.3	4.44	3.86										
22	Oil palm shell	1	4.48	3.9										
22	Oil palm shell	3	4.72	4.01										
22	Oil palm shell	10	5.24	4.25										
22	Oil palm shell	30	5.99	5.41										
22	Cacao shell	0	4.37	3.9										
22	Cacao shell	0.1	4.51	3.91										
22	Cacao shell	0.3	4.66	3.98										
22	Cacao shell	1	5.35	4.29										
22	Cacao shell	3	6.48	5.74										
22	Cacao shell	10	8.13	7.68										
22	Cacao shell	30	9.15	8.53										
23		0	5.01	4.18					3.51			6.01	0.4	14.9
23	Rice husk	0	5.24	4.31										
23	Rice husk	0.1	5.46	4.29										
23	Rice husk	0.3	5.4	4.28										
23	Rice husk	1	5.39	4.3										
23	Rice husk	3	5.52	4.34										
23	Rice husk	10	5.78	4.39										
23	Rice husk	30	6.15	4.81										
23	Oil palm shell	0	5.21	4.31										
23	Oil palm shell	0.1	5.38	4.27										
23	Oil palm shell	0.3	5.32	4.29										
23	Oil palm shell	1	5.34	4.31										
23	Oil palm shell	3	5.43	4.33										
23	Oil palm shell	10	5.47	4.54										
23	Oil palm shell	30	6.05	5										
23	Cacao shell	0	5.35	4.31										
23	Cacao shell	0.1	5.56	4.3										
23	Cacao shell	0.3	5.72	4.36										
23	Cacao shell	1	6.24	4.65										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
23	Cacao shell	3	7.39	5.95										
23	Cacao shell	10	8.48	7.55										
23	Cacao shell	30	9.48	8.62										
24		0	4.46	3.85					5.4			0.81	0.02	42.3
24	Rice husk	0	4.9	3.81	0.35	0.18	0.07	0.07	6.43	5.35	0.53			
24	Rice husk	0.1	4.91	3.8	0.49	0.18	0.06	0.03	6.9	4.34	0.43			
24	Rice husk	0.3	4.88	3.81	0.31	0.1	0.03	0.05		4.32	0.45			
24	Rice husk	1	4.96	3.8	0.27	0.17	0.06	0.13	7.65	4.01	0.26			
24	Rice husk	3	4.99	3.81	0.18	0.23	0.11	0.08	7.68	3.42	0.3			
24	Rice husk	10	5.17	3.87	0.67	0.56	0.47	2.97	7.46	3.81	0.24			
24	Rice husk	30	5.23	4.09	0.9	0.86	0.75	0.18	13.93	3.62	0.47			
24	Oil palm shell	0	4.88	3.81	0.36	0.2	0.07	0.07	6.45	5.37	0.52			
24	Oil palm shell	0.1	4.89	3.81	0.22	0.18	0.08	0.08	6.73	2.34	0.19			
24	Oil palm shell	0.3	4.95	3.85	0.41	0.29	0.17	0.08	7.87	3.62	0.35			
24	Oil palm shell	1	5.03	3.95	0.57	0.4	0.47	0.11	8.22	2.03	0.38			
24	Oil palm shell	3	5.09	3.95	0.59	0.39	0.08	0.08	9.17	0.39	0.23			
24	Oil palm shell	10	5.15	3.95	1.05	0.77	0.23	0.08	9.46	0.8	0.15			
24	Oil palm shell	30	5.39	4.23	1.8	1.56	0.53	0.1	10.68	0.85	0.23			
24	Cacao shell	0	4.91	3.82	0.33	0.17	0.07	0.07	6.33	5.32	0.53			
24	Cacao shell	0.1	4.92	3.83	0.27	0.24	0.08	0.08	6.74	4.51	0.16			
24	Cacao shell	0.3	4.95	3.84	0.54	0.18	0.17	0.03	7.25	3.71	0.32			
24	Cacao shell	1	5.22	3.94	0.58	0.5	0.59	0.08	7.62	2.41	0.2			
24	Cacao shell	3	5.98	4.64	1.35	1.11	1.51	0.21	8.87	0	0.04			
24	Cacao shell	10	8.5	7.58	3.1	2.25	6.94	0.26	9.61	0	0			
24	Cacao shell	30	9.83	9.05	4.5	4.2	17.78	0.36	10.14	0	0			
25		0	4.56	4.15					7.52			8.91	0.37	24.1
25	Rice husk	0	4.24	3.88										
25	Rice husk	0.1	4.26	3.89										
25	Rice husk	0.3	4.25	3.89										
25	Rice husk	1	4.25	3.88										
25	Rice husk	3	4.28	4.03										
25	Rice husk	10	4.79	4.16										
25	Rice husk	30	5.57	4.35										
25	Oil palm shell	0	4.2	3.8										
25	Oil palm shell	0.1	4.22	3.88										
25	Oil palm shell	0.3	4.2	3.84										
25	Oil palm shell	1	4.26	3.9										
25	Oil palm shell	3	4.38	4.12										
25	Oil palm shell	10	4.98	4.27										
25	Oil palm shell	30	6	4.69										
25	Cacao shell	0	4.28	3.88										
25	Cacao shell	0.1	4.36	3.94										
25	Cacao shell	0.3	4.4	4.04										
25	Cacao shell	1	5.02	4.28										
25	Cacao shell	3	6.53	5.04										
25	Cacao shell	10	8.09	6.8										
25	Cacao shell	30	8.94	7.89										
26		0	4.7	3.81					10.04			3.06	0.28	11
26	Rice husk	0	5.34	3.95										
26	Rice husk	0.1	5.35	3.95										
26	Rice husk	0.3	5.37	3.96										
26	Rice husk	1	5.38	3.96										
26	Rice husk	3	5.39	3.96										
26	Rice husk	10	5.39	4.02										
26	Rice husk	30	5.57	4.06										
26	Oil palm shell	0	5.32	3.92										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^+	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
26	Oil palm shell	0.1	5.28	3.91										
26	Oil palm shell	0.3	5.34	3.92										
26	Oil palm shell	1	5.35	3.95										
26	Oil palm shell	3	5.41	3.97										
26	Oil palm shell	10	5.49	4.15										
26	Oil palm shell	30	5.63	4.82										
26	Cacao shell	0	5.35	3.95										
26	Cacao shell	0.1	5.4	3.96										
26	Cacao shell	0.3	5.43	3.98										
26	Cacao shell	1	5.64	4.17										
26	Cacao shell	3	6.37	4.87										
26	Cacao shell	10	7.87	7										
26	Cacao shell	30	8.97	8.13										
27		0	4.7	3.74					10.73			1.94	0.21	9.1
27	Rice husk	0	5.35	3.9										
27	Rice husk	0.1	5.33	3.89										
27	Rice husk	0.3	5.38	3.91										
27	Rice husk	1	5.42	3.92										
27	Rice husk	3	5.92	3.94										
27	Rice husk	10	5.95	3.98										
27	Rice husk	30	5.98	4.01										
27	Oil palm shell	0	5.35	3.9										
27	Oil palm shell	0.1	5.36	3.9										
27	Oil palm shell	0.3	5.41	3.91										
27	Oil palm shell	1	5.5	3.92										
27	Oil palm shell	3	5.51	3.97										
27	Oil palm shell	10	5.59	4.11										
27	Oil palm shell	30	5.68	4.65										
27	Cacao shell	0	5.35	3.9										
27	Cacao shell	0.1	5.36	3.91										
27	Cacao shell	0.3	5.4	3.91										
27	Cacao shell	1	5.42	4.09										
27	Cacao shell	3	6.01	4.43										
27	Cacao shell	10	7.88	7.18										
27	Cacao shell	30	9.12	8.37										
28		0	4.44	3.7					12.28			1.84	0.14	13.1
28	Rice husk	0	4.62	3.82										
28	Rice husk	0.1	4.69	3.81										
28	Rice husk	0.3	4.67	3.82										
28	Rice husk	1	4.71	3.81										
28	Rice husk	3	4.77	3.82										
28	Rice husk	10	4.87	3.86										
28	Rice husk	30	4.96	3.91										
28	Oil palm shell	0	4.6	3.8										
28	Oil palm shell	0.1	4.64	3.8										
28	Oil palm shell	0.3	4.62	3.81										
28	Oil palm shell	1	4.66	3.82										
28	Oil palm shell	3	4.72	3.85										
28	Oil palm shell	10	4.9	3.97										
28	Oil palm shell	30	5.4	4.36										
28	Cacao shell	0	4.61	3.8										
28	Cacao shell	0.1	4.72	3.81										
28	Cacao shell	0.3	4.74	3.85										
28	Cacao shell	1	4.97	3.89										
28	Cacao shell	3	5.45	4.11										
28	Cacao shell	10	7.3	6.23										

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		BC addition	$pH_{(H2O)}$	$pH_{(KCl)}$	Ca	Mg	K	Na	CEC	Al^{3+}	H^{+}	C	N	CN
		(%)			cmol$_c$/kg soil							(%)	(%)	
28	Cacao shell	30	8.55	7.59										
29		0	4.4	3.82					3.41			0.98	0.07	14.4
29	Rice husk	0	4.38	3.86										
29	Rice husk	0.1	4.38	3.89										
29	Rice husk	0.3	4.41	3.9										
29	Rice husk	1	4.48	3.9										
29	Rice husk	3	4.58	3.93										
29	Rice husk	10	5.02	4.03										
29	Rice husk	30	5.35	4.37										
29	Oil palm shell	0	4.37	3.85										
29	Oil palm shell	0.1	4.39	3.87										
29	Oil palm shell	0.3	4.4	3.89										
29	Oil palm shell	1	4.4	3.9										
29	Oil palm shell	3	4.59	3.97										
29	Oil palm shell	10	4.83	4.12										
29	Oil palm shell	30	5.38	4.66										
29	Cacao shell	0	4.39	3.87										
29	Cacao shell	0.1	4.42	3.9										
29	Cacao shell	0.3	4.48	4										
29	Cacao shell	1	5.07	4.22										
29	Cacao shell	3	6.02	5.12										
29	Cacao shell	10	7.57	7.52										
29	Cacao shell	30	9.14	8.58										
30		0	4.83	4.41					10.06			0.82	0.01	59.8
30	Rice husk	0	5.19	4.45	0.67	0.06	0	0.05	10.01	0.16	0.12			
30	Rice husk	0.1	5.17	4.46	0.39	0.15	0	0.03	10.2	0.11	0.16			
30	Rice husk	0.3	5.39	4.45	0.18	0.1	0.03	0.02	10.99	0.14	0.12			
30	Rice husk	1	5.42	4.45	0.27	0.1	0.03	0.02	11.34	0.1	0.14			
30	Rice husk	3	5.47	4.56	0.32	0.15	0.14	0.01	11.73	0.04	0.12			
30	Rice husk	10	5.55	4.79	0.72	0.44	0.45	0.03	11.8	0.02	0.06			
30	Rice husk	30	5.76	5.11	0.98	0.88	1.39	0.13	12.28	0	0.04			
30	Cacao shell	0	5.14	4.43	0.68	0.06	0	0.05	10.02	0.14	0.11			
30	Cacao shell	0.1	5.19	4.5	0.13	0.15	0	0.05	10.37	0.06	0.12			
30	Cacao shell	0.3	5.33	4.7	0.36	0.21	0.17	0.08	11.49	0	0.02			
30	Cacao shell	1	5.7	5.45	0.63	0.42	0.58	0.1	11.37	0	0			
30	Cacao shell	3	6.99	6.69	1.16	0.77	2.04	0.13	11.95	0	0.02			
30	Cacao shell	10	9.15	8.29	3.14	2.24	9.01	0.23	11.96	0	0			
30	Cacao shell	30	9.92	9.19	6.13	5.22	18.03	0.31	14.43	0	0			
31		0	4.57	4.15								0.52	0.05	10.3
31	Rice husk	0	4.5	4.2	0.22	0.18	0	0.08	1.62	0.04	1.29			
31	Rice husk	0.1	4.6	4.2	0.18	0.1	0.03	0.03	1.77	0.06	1.68			
31	Rice husk	0.3	4.5	4.2	0.18	0.19	0	0.25	1.64	0.19	1.46			
31	Rice husk	1	4.9	4.2	0.76	0.22	0.06	0.1	1.91	0.11	1.68			
31	Rice husk	3	5.3	4.3	0.27	0.28	0.14	0.05	1.7	0.21	1.73			
31	Rice husk	10	5.7	4.4	0.45	0.56	0.36	0.08	1.73	0.08	1.24			
31	Rice husk	30	6.2	4.8	0.72	1.02	1.26	0.13	2.96	0.05	0.96			
31	Oil palm shell	0	5.22	4.2	0.23	0.17	0	0.08	1.6	0.04	1.28			
31	Oil palm shell	0.1	5.23	4.3	0.41	0.24	0.06	0.03	1.76	0.17	1.55			
31	Oil palm shell	0.3	5.25	4.3	0.27	0.3	0.17	0.13	1.87	0.14	1.2			
31	Oil palm shell	1	5.4	4.7	0.63	0.51	0.59	0.08	1.64	0.04	1.42			
31	Oil palm shell	3	5.48	4.3	0.36	0.35	0.06	0.05	1.66	0.18	1.33			
31	Oil palm shell	10	5.92	4.5	0.85	0.81	0.25	0.08	2.47	0.16	1.1			
31	Oil palm shell	30	6.19	4.9	1.35	1.4	0.53	0.1	2.85	0.08	1.18			
31	Cacao shell	0	4.5	4.2	0.23	0.17	0	0.07	1.61	0.05	1.28			
31	Cacao shell	0.1	4.9	4.3	0.18	0.19	0.03	0.05	1.63	0.15	1.32			
31	Cacao shell	0.3	5.3	4.4	0.22	0.24	0.17	0.05	1.6	0.14	1.29			
31	Cacao shell	1	6.1	4.7	0.49	0.5	0.53	0.08	1.92	0.02	1.51			
31	Cacao shell	3	7.9	6.5	1.08	1.11	2.92	0.15	2.14	0	1.82			
31	Cacao shell	10	9.4	8.3	2.24	2.42	8.09	0.23	2.76	0	1.11			
31	Cacao shell	30	10	9.3	4.17	4.47	13.65	0.37	3.64	0	1.18			

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Table S3. Estimated fixed-effects parameters for the model y = a + b *(1-exp$^{(-BC\%/c)}$) (eq. 1) based on restricted maximum likelihood estimates from nonlinear mixed effects-models for the response in $pH_{(H2O)}$, CEC, Ca, Mg and K ($cmol_c$/kg soil) to BC addition (%). The table shows biochar specific contrasts (cacao shell (CS), oil palm shell (OP) and rice husk (RH)) for the parameters a, b and c. p-values are based on likelihood ratio tests between selected models. $p<0.05$ indicates significant difference between two models. The selected model is indicated in bold and italic.

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Model	Model nr.	Assumed structure	Parameter estimates (± SE) a	Parameter estimates (± SE) b	Parameter estimates (± SE) c	Test	p-value
$pH_{(H2O)}$ ~ BC%, n= 674	1	a: CS=OP=RH b: CS≠OP≠RH c: CS≠OP≠RH					
	2	***a: CS=OP=RH*** ***b: CS ≠(OP=RH)*** ***c: CS ≠ OP ≠ RH***	***CS/OP/RH:*** *4.73 ± 0.085*	***CS:*** *4.35 ± 0.087* ***OP/RH:*** *1.00 ± 0.095*	***CS:*** *8.59 ± 0.641* ***OP:*** *19.35 ± 1.847* ***RH:*** *22.49 ± 2.084*	Model 1 vs. 2	0.59
	3	b: CS=OP=RH				Model 2 vs. 3	<0.0001
	4	c: CS≠(OP=RH)				Model 2 vs. 4	0.048
	5	c: CS=OP=RH				Model 2 vs. 5	<0.0001
CEC ~ BC%, n= 101	1	a: CS=OP=RH b: CS≠OP≠RH c: CS≠OP≠RH					
	2	b: CS=OP=RH				Model 1 vs. 2	0.4184
	3	***a: CS=OP=RH*** **b: CS=OP=RH** **c: (CS=OP)≠RH**	***CS/OP/RH:*** *5.62 ± 1.470*	***CS/OP/RH:*** *2.51 ± 0.432*	***CS/OP:*** *3.94 ± 0.940* ***RH:*** *11.77 ± 3.133*	Model 3 vs. 2	0.317
Ca ~ BC%, n= 97	1	a: CS=OP=RH b: CS≠OP≠RH c: CS≠OP≠RH					
	2	***a: CS=OP=RH*** ***b: CS ≠(OP=RH)*** ***c: CS ≠ OP ≠ RH***	***CS/OP/RH:*** *0.50 ± 0.121*	***CS:*** *5.63 ± 0.366* ***OP/RH:*** *1.29 ± 0.151*	***CS:*** *16.10 ± 3.134* ***OP:*** *11.45 ± 2.905* ***RH:*** *62.93 ± 22.973*	Model 1 vs. 2	0.8823
	3	b: CS=OP=RH				Model 2 vs. 3	0.0001
	4	c: CS≠(OP=RH)				Model 2 vs. 4	<.0001
	5	c: RH≠(OP=CS)				Model 2 vs. 5	0.0044
Mg ~ BC%, n= 97	1	a: CS=OP=RH b: CS≠OP≠RH c: CS≠OP≠RH					
	2	***a: CS=OP=RH*** ***b: CS ≠(OP=RH)*** ***c: CS ≠ OP ≠ RH***	***CS/OP/RH:*** *0.19 ± 0.028*	***CS:*** *5.79 ± 0.508* ***OP/RH:*** *1.82 ± 0.499*	***CS:*** *23.60 ± 3.674* ***OP:*** *21.58 ± 11.284* ***RH:*** *50.71 ± 20.377*	Model 1 vs. 2	0.8479
	3	b: CS=OP=RH				Model 2 vs. 3	<0.0001
	4	c: CS≠(OP=RH)				Model 2 vs. 4	<.0001
	5	c: RH≠(OP=CS)				Model 2 vs. 5	<0.0001
K ~ BC%, n= 90	1	a: CS=OP=RH b: CS≠OP≠RH c: CS≠OP≠RH					
	2	***a: CS=OP=RH*** ***b: CS ≠ OP ≠ RH*** ***c: CS ≠(OP=RH)***	***CS/OP/RH:*** *0.05 ± 0.032*	***CS:*** *31.32 ± 4.253* ***OP*** *1.94 ± 3.565* ***RH:*** *3.94 ± 7.298*	***CS:*** *38.67 ± 6.757* ***OP/RH:*** *85.94 ± 187.342*	Model 1 vs. 2	0.0735
	3	b: CS≠(OP=RH)				Model 2 vs. 3	0.0002
	4	c: CS≠OP≠RH				Model 2 vs. 4	0.0132