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# Fading positive effect of biochar on crop yield and soil acidity during five growth seasons in an Indonesian Ultisol



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

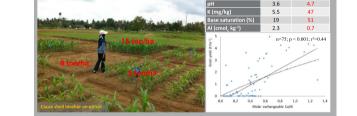
- Two biochars at 3 dosages over 5 seasons were studied in Indonesia.
- Cacao shell biochar showed a strong positive effect on maize crop yield.
  The effect was save by elleviation of
- The effect was cause by alleviation of soil acidity.
- After 3 to 5 seasons reapplication of the biochar was necessary.

### ARTICLE INFO

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#### ABSTRACT

Low fertility limits crop production on acidic soils dominating much of the humid tropics.

Biochar may be used as a soil enhancer, but little consensus exists on its effect on crop yield. Here we use a controlled, replicated and long-term field study in Sumatra, Indonesia, to investigate the longevity and mechanism of the effects of two contrasting biochars (produced from rice husk and cacao shell, and applied at dosages of 5 and 15 t ha<sup>-1</sup>) on maize production in a highly acidic Ultisol (pH<sub>KCI</sub> 3.6).

Compared to rice husk biochar, cacao shell biochar exhibited a higher pH (9.8 vs. 8.4), CEC (197 vs. 20 cmol<sub>c</sub> kg<sup>-1</sup>) and acid neutralizing capacity (217 vs. 45 cmol<sub>c</sub> kg<sup>-1</sup>) and thus had a greater liming potential. Crop yield effects of cacao shell biochar (15 t ha<sup>-1</sup>) were also much stronger than those of rice husk biochar, and could be related to more favorable Ca/Al ratios in response to cacao shell biochar (1.0 to 1.5) compared to rice husk biochar (0.3 to 0.6) and nonamended plots (0.15 to 0.6).

The maize yield obtained with the cacao shell biochar peaked in season 2, continued to have a good effect in seasons 3–4, and faded in season 5. The yield effect of the rice husk biochar was less pronounced and already faded from season 2 onwards.

Crop yields were correlated with the pH-related parameters Ca/Al ratio, base saturation and exchangeable K. The positive effects of cocoa shell biochar on crop yield in this Ultisol were at least in part related to alleviation of soil acidity. The fading effectiveness after multiple growth seasons, possibly due to leaching of the biochar-associated alkalinity, indicates that  $15 \text{ th} \text{ h}^{-1}$  of cocoa shell biochar needs to be applied approximately every third season in order to maintain positive effects on yield.

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#### 1. Introduction

Biochar amendment to soils offers a method to sequester carbon in soil with the co-benefits of waste management, pollutant immobilization, fertility increase and/or N<sub>2</sub>O emission reductions of degraded soils (Jeffery et al., 2015; Lehmann, 2007). The mechanism behind this fertility increase can be improved water retention (Bruun et al., 2014), improved soil structure (Bruun et al., 2014; Obia et al., 2017; Obia et al., 2016), improved nutrient retention (Biederman and Harpole, 2013; Hale et al., 2013; Laird et al., 2010; Martinsen et al., 2014), increased robustness towards pests (Harel et al., 2012; Mehari et al., 2015), improved nutrient transport by mycorrhizae (Warnock et al., 2007), alleviation of soil acidity (Biederman and Harpole, 2013; Jeffery et al., 2017; Martinsen et al., 2015; Yamato et al., 2006), or combinations of these mechanisms. For less degraded soils, enrichment of the biochar with nutrients by co-composting or mixing with urine or mineral nutrients can still result in positive biochar effects on crop yield, especially in those cases where nutrient availability of the main growth-limiting factor (Hagemann et al., 2017a; Kammann et al., 2015; Schmidt et al., 2017; Schmidt et al., 2015).

Large variations in biochar effectiveness on crop harvest in the tropics have been shown, from minor, generally insignificant effects to strongly positive effects, with the median effect (taken from a meta-analysis study) being an increase of about 20% (Jeffery et al., 2017). The effect of biochar is usually strong in tropical soil (Agegnehu et al., 2016; Asai et al., 2009; Jeffery et al., 2017; Jeffery et al., 2011; Major et al., 2010; Yamato et al., 2006) in comparison to soils in temperate zones where the effect of biochar on the yield and soil properties is usually low (Bonanomi et al., 2017; Jeffery et al., 2017; Jeffery et al., 2011). Soils of high fertility (high cation exchange capacity, water retention, neutral pH) have shown to benefit less from biochar addition (Bass et al., 2016; Cornelissen et al., 2013; Jones et al., 2012). Effects tend to be a bit more strongly positive for acidic (pH < 5) and weathered soils with coarse or medium/heavy texture which are characteristic of tropical soils (Crane-Droesch et al., 2013; Jeffery et al., 2017). The effect of biochar seems to be thus strongly connected to the soil properties and the climate, but thus far correlations with crop yield are not completely clear. Several authors (including meta-analysis studies) state the yield increases are related to an overall improvement of soil qualities (Agegnehu et al., 2016; Asai et al., 2009; Crane-Droesch et al., 2013; Jeffery et al., 2017; Jeffery et al., 2011), also in tropical soils (Biederman and Harpole, 2013; Gurwick et al., 2013; Jeffery et al., 2017), however pin pointing the exact mechanism behind the increase in yields can be challenging.

In extensive four-season field trials in Thailand and the Philippines with rice husk biochar, Haefele et al. (2011) observed increased yields of 16-35%, and hypothesized that the increase was a result of improvements in water retention and increased available K and P. Steiner et al. (2007) tested biochar effects over four planting seasons in an acidic soil in Brazil ( $pH_{H_{2}O} = 4.5$ ), and found positive effects of biochar that faded over time in multiple seasons. Major et al. (2010) studied biochar effects in an acidic oxisol in Colombia for 4 years, and did not find any effects in the first year, but maize yield increases in the three subsequent seasons. Griffin et al. (2017) investigated the amendment of walnut shell biochar over four years in a field experiment, and found a short-lived effect on maize crop yield in the second year. A long-term wheat/maize field experiment in a calcareous soil (pH 7.1-7.8) with extremely high biochar dosages (30, 60, and 90 t ha<sup>-1</sup>) revealed a slight increase in cumulative yield over four seasons (Liang et al., 2014), due to lower bulk density, improved soil moisture and K addition. Jones et al. (2012) did a three-year study of biochar on maize and grass yield, in pH-neutral (pH 6.6) sandy clay loam in Wales, UK. Biochar effects were stronger in year two than in year one. After three years in the field, biochar had caused beneficial changes in the microbial community.

Despite their merit of drawing general conclusions from a plethora of data, the meta-analyses on biochar effect on crop yield have necessarily pooled the available data without considering the time since biochar application or inter-season variation for studies carried out over multiple years. The reason is that there are too few studies carried out for longer time spans. A recent review reported that 60% of the 428 data points reviewed were based on one year trials or simply used data corresponding to the first year of multiple-year studies (Bach et al., 2016). Thus, there is a need for well-controlled, replicated and longer-term field studies on representative soils. Here, we contribute to closing this gap, as information will be obtained related to trends observed for yields from a highly acidic soil up to five seasons since biochar application, with two very different biochars. The mechanism explaining the soil enhancement effect of biochar will also be investigated, as well as and how often one would need to replenish the biochar in order to maintain the positive soil fertility effects.

Ultisols in the humid tropics such as the presently studied soil require significant liming or addition of organic matter to remediate Al toxicity, which is acknowledged as one of the major causes for crop failure (Bloom et al., 1979). Biochar often contains a major ash component, which is alkaline in nature, and may be used as an alternative for lime, with the co-benefits of carbon sequestration and other improved soil characteristics (Cornelissen et al., 2013; Kelly et al., 2014; Kimetu et al., 2008; Martinsen et al., 2015; Yamato et al., 2006). The two biochars tested for their effects on crop yield and soil properties were made from cacao shell and rice husk, strongly differing in acid neutralization capacity (ANC) and cation exchange capacity (CEC). A high ANC of a biochar can probably alleviate soil acidity and reduce available Al concentrations (Gruba and Mulder, 2015; Major et al., 2010; Martinsen et al., 2015; Steiner et al., 2007). Also P availability can be positively impacted by an increasing pH (Lajtha and Schlesinger, 1988; Martinsen et al., 2014).

The hypotheses for this study were 1) that the agronomic effects of biochar in this soil could be explained by reduced soil acidity, as expressed by reduced exchangeable  $Al^{3+}$  concentrations as well as increased pH, Ca/Al ratios, and base saturation. As a result it was also hypothesized that the biochar with highest ANC would give the strongest yield effects in a soil where crop growth is mainly limited by soil acidity, and 2) that the biochar effectiveness on crop yield would decline over time, due to continued nutrient leaching and rapid depletion of the alkalinity added via the biochar (Glaser et al., 2002; Lehmann and Rondon, 2006).

#### 2. Materials and methods

#### 2.1. General outline

To investigate the longevity and mechanism of biochar effects on maize production in highly acidic soils of the humid tropics, an extensive field trial was carried out over five cropping seasons, with two biochars and five replicates at an experimental farm in the Lampung district, South Sumatra, Indonesia. The soil was classified as a Typic Kanhapludult Ultisols with high levels of exchangeable aluminum (Al; around 2 cmol<sub>c</sub> kg<sup>-1</sup>) and very low pH (3.6 in KCl and 3.7 in water). The Lampung district has high rainfall (1796 mm) and temperatures (30 °C) throughout the year, and thus a high soil leaching and weathering potential.

Both biochars were applied in dosages of 0, 5 and 15 t  $ha^{-1}$  and mixed into the upper 10 cm of the soil. Soil bulk density was 1.30 g cm<sup>-3</sup>. Percent addition of biochars (w/w) was thus 0.4% and 1.2% for the 5 and 15 t  $ha^{-1}$  additions, respectively. Both soil chemical parameters and maize yields were monitored over the five growth seasons.

#### 2.2. Biochars

Biochars were prepared from rice husk and cacao shell, two common agricultural wastes in Indonesia. Pyrolysis was carried out in a simple kiln without a retort function, and the procedure and conditions for making the biochars have been extensively described in refs. (Alling et al., 2014; Martinsen et al., 2015; Obia et al., 2015), where the same biochars were studied. Pyrolysis temperatures, as determined via Thermogravimetric Analyses were between 400 and 500 °C (Obia et al., 2015). Biochar characteristics were reported in earlier work (Martinsen et al., 2015; Obia et al., 2015) and reported in Table S1.

#### 2.3. Soils

Field trials were carried out on a strongly acidic (pH<sub>KCl</sub> 3.6), sandy loam Ultisol (Typic Kanhapludults; Sand: 54%; silt: 22%; clay: 24%) in Lampung province (Southern Sumatra, Indonesia; 05°00.406' S; 105°29.405' E; annual rainfall 1796 mm; Fig. S1), over five planting seasons: season 1: July-October 2012; season 2: December 2012-April 2012; season 3: April-August 2013; season 4: November 2013-February 2014; season 5: April-July 2014. Precipitation occurs throughout the year in Lampung province, with relatively low rainfall in June-November (seasons 1; part of seasons 3 and 5). In the relatively dry seasons, the plots were irrigated when necessary, in order to keep the seasons as comparable as possible, and because the effect of biochar on moisture retention was neither the topic of the study nor a mechanism expected to be of importance for biochar effects in this humid region. In between cropping seasons, the land was tilled to 15–20 cm depth with a hand held mini-tractor, fertilized, weeded with a generic maize weed killer, and replanted. All plots were on flat terrain on the experimental farm Tamanbogo, belonging to the Indonesian Soil Research Institute. Selected soil properties are presented in Table S1.

#### 2.4. Field trial design

Five blocks were established in a completely randomized block design. A total of 30 experimental plots of  $4 \times 4$  m size were thus established (two biochars at three dosages each, in five blocks). Onemeter spaces were kept between the plots. Both biochars were applied in single dosages of 0, 5 and 15 t ha<sup>-1</sup> in each of the five blocks, prior to the first growth season only (July 2012), by manual mixing into the 0–10 cm soil layer. The biochars were not enriched with organic nutrients as this did not fit with common agricultural practice in the area due to limited availability of manure. Each treatment (6 levels) in each of the five blocks was sampled for 5 seasons (i.e. one sample per treatment, block and season).

#### 2.5. Field establishment

Maize (*Zea mays L.*) was planted at 20 cm  $\times$  75 cm spacing. Handweeding was carried out when required. Mineral fertilizer was applied three times per planting season: just before planting (NPK 30:30:30 kg ha<sup>-1</sup>), in the early vegetative growth stage (69 kg ha<sup>-1</sup> urea-N), and in the early generative growth stage (69 kg ha<sup>-1</sup> urea-N). Insecticide application was also carried out before planting. No lime was applied as one of the purposes of the experiment was to investigate the pH effect of biochar amendment.

#### 2.6. Yield data

Yield was normalized to grains dried overnight at 110 °C.

Statistical testing of effects of treatments or additions was done by the statistical package "R", version 3.4.4 (R-Core-Team, 2018). Linear mixed-effects models were fitted using the R extension package lme4 (Bates et al., 2015) to evaluate differences between biochar type, biochar dosage and season (fixed effects, Tables S5–S7 in the Supporting information). Variation in yield (and soil characteristics Ca/Al and pH, see below) between the different blocks was modeled by introducing random effects associated with each of the blocks. Likelihood ratio tests were used to simplify the fixed effects structure of the models. Model checking was based on visual inspection of residual and QQ plots. Differences between the management practices were assessed by means of pairwise comparisons (package "multcomp") using model-based approximate *t*-tests with adjustment for multiplicity (Hothorn et al., 2008). Variation between biochar types was assessed by comparing 5 t ha<sup>-1</sup> Cacoa shell vs. 5 t ha<sup>-1</sup> Rice husk per season (1–5) and 15 t ha<sup>-1</sup> Cacoa shell vs. 15 t ha<sup>-1</sup> Rice husk per season (1–5). Variation with biochar dosage was assessed by comparing 5 t ha<sup>-1</sup> vs. 15 t ha<sup>-1</sup> Cacoa shell per season (1–5) and 5 t ha<sup>-1</sup> vs. 15 t ha<sup>-1</sup> Rice husk per season (1–5). Changes with season were assessed by comparisons of seasons for each of the biochar type and dose combinations separately.

#### 2.7. Soil characterization after biochar amendment

After each planting season, soils from all individual plots were sampled and stored at 4 °C. Per individual plot, five 100 g soil samples, taken from 0 to 10 cm depth with a small spade, were pooled into one 500 g mixed sample per plot. Thus, five replicate samples per treatment were obtained (as there were five replicate plots per treatment). The following parameters were measured for the first three planting seasons: CEC (ammonium acetate 1 M, pH 7), pH<sub>H<sub>2</sub>O</sub> (pH in water, 1 g soil in 5 mL water) and pH<sub>KCI</sub> (pH in 1 M KCI), exchangeable base cations in the CEC extracts and base saturation, exchangeable H<sup>+</sup> (back-titration of the CEC extract with sodium hydroxide to pH 7) and Al<sup>3+</sup>, available P (Bray), and elemental composition [total N and H, organic C (catalytic combustion elemental analysis at 1030 °C after acidification of the soil with 50 µL 1 M HCl per 15 mg dry sample)], all using standard methods as described in refs. (Alling et al., 2014; Cornelissen et al., 2013; Martinsen et al., 2015) and in the footnote of Table S1. During season four and five, due to funding limitations soil analyses were restricted to  $pH_{H_2O}$ , exchangeable K, and CEC. Differences in molar exchangeable Ca/Al ratios and pH (three seasons for Ca/Al, five seasons for pH) were assessed as described above. In addition, 0 t ha<sup>-1</sup> was included for comparisons of biochar type, biochar dosage and season (Tables S3-S7). Linear regression was used for exploring relationships between grain yield and selected soil variables.

#### 3. Results

#### 3.1. Soil and biochar quality

The soil was a strongly acidic ( $pH_{H_2O} = 3.7$ ;  $pH_{KCI} = 3.6$ ) sandy loamy Typic Kanhapludult, with a low base saturation (15% of the CEC; Table S1). Associated with the low soil pH, the soil had a relatively high exchangeable  $Al^{3+}$  content (2.4 cmol<sub>c</sub> kg<sup>-1</sup>), low exchangeable  $Ca^{2+}$  (1.0 cmol<sub>c</sub> kg<sup>-1</sup>) and thus low Ca/Al molar ratio (0.63  $\pm$  0.05; average for control plots in planting seasons 1-3). The organic carbon content was low (0.87  $\pm$  0.01%). Cocoa shell biochar exhibited a higher pH than rice husk biochar (9.8 vs. 8.4, respectively), as well as a much higher CEC (197 vs. 18 cmol<sub>c</sub> kg<sup>-1</sup>, respectively, for exchangeable and soluble cations as measured by ammonium acetate extraction, and 37 vs. 26 cmol<sub>c</sub> kg<sup>-1</sup>, respectively, for truly exchangeable cations as measured by ammonium replacement by KCl extraction). Importantly, the cacao shell biochar exhibited a much higher acid neutralizing capacity than the rice husk biochar (ANC, 217  $\text{cmol}_{c}$  kg<sup>-1</sup> vs. 45  $\text{cmol}_{c}$  kg<sup>-1</sup>, respectively), resulting in a much higher alkalinity. The cacao shell biochar had a lower ash content (19 vs. 51%, respectively) and a higher organic C content (70 vs. 41%, respectively) than the rice husk biochar. This could be due to the high silicate content, yet small base cation content of the rice husk biochar (Chandrasekhar et al., 2003), resulting in a relatively high ash content, but low ANC of the charred material.

#### 3.2. Crop yield: effect of biochar feedstock

Cacao shell biochar and rice husk biochar were both used in two dosages, under maize cropping for five seasons. Without biochar amendment, hardly any emergence of maize plants occurred and thus no maize yield was obtained (Figs. 1 and S1). This is likely a result of the low soil pH and high levels of exchangeable Al (Ca/Al ratios between 0.15 and 0.6 with little variation between season 1 and 2; Fig. 2a). The smallest cacao shell biochar amendment (5 t ha<sup>-1</sup>) alleviated the deleterious effects of Al<sup>3+</sup> (see next section) and improved maize emergence and resulted in grain yields of 1.3 and 2.9 t ha<sup>-1</sup> in seasons 1 and 2, respectively. A dosage of 15 t ha<sup>-1</sup> resulted in significantly (p < 0.05) higher grain yields (1.9 and 4.3 t ha<sup>-1</sup> in seasons 1 and 2, respectively) than a dosage of 5 t ha<sup>-1</sup> in all seasons except the first one (Fig. 1 and Table S3). At a dosage of 15 t ha<sup>-1</sup> in season 2) than cacao shell biochar (4.3 t ha<sup>-1</sup> in season 2) in all seasons except season 1, and for a dosage of 5 t ha<sup>-1</sup> the same was observed for seasons 1, 2 and 3 (Table S3).

#### 3.3. Crop yield: effect of multiple seasons

The biochar trials (both 5 and 15 t ha<sup>-1</sup> dosages, both cacao shell and rice husk feedstocks) were continued for five seasons. Variation of biochar effectiveness in the seasons following application revealed an interesting pattern (Fig. 1, Table S3). The maize yield obtained with the 15 t ha<sup>-1</sup> cacao shell biochar peaked in season 2 at 4.3 t ha<sup>-1</sup>, continued to have a good effect in seasons 3 and 4 (Fig. 1); the production stimulating effect faded during season 5 at the 15 t ha<sup>-1</sup> dosage. For the lower 5 t ha<sup>-1</sup> dose of cacao shell biochar, a decline in maize yield was already seen from season 3 onwards. The less strong yield effect of 15 t ha<sup>-1</sup> rice husk biochar amendment was only significant (p < 0.05) in season 1 (1.4 t ha<sup>-1</sup> maize yield; Fig. 1), and already faded from season 2 onwards. Results for maize stover biomass showed similar trends as for grain yield (Fig. S2).

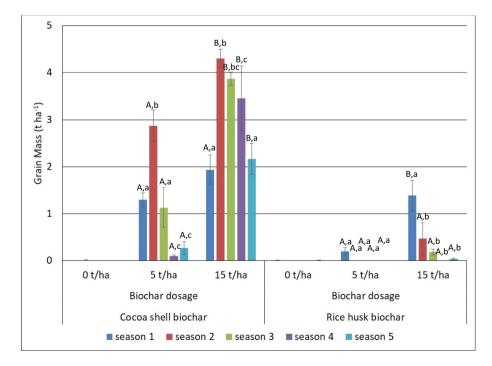
#### 3.4. Crop yield: effect of soil properties

Soil properties were measured after each planting season. The full soil data set can be found in Tables S8–S15. Soil  $pH_{H_2O}$  and base saturation increased significantly (p < 0.05) with the application of cacao shell biochar at both addition rates (Fig. 2). In contrast, the addition of rice husk biochar had a less pronounced effect on soil pH, Ca/Al ratios and

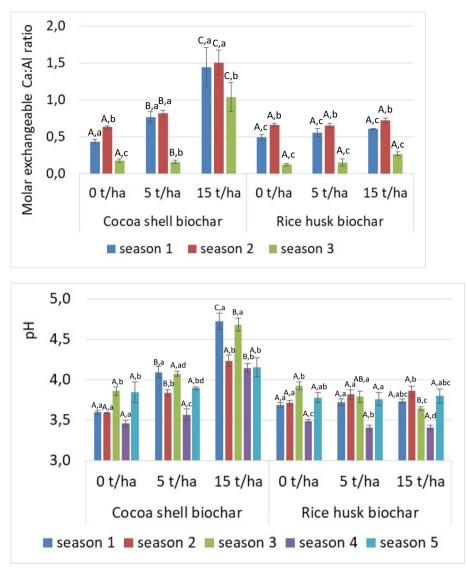
base saturation, which is probably the main explanation for the difference in crop yield effects between the two biochar types. Similar to the trends observed for maize yield, soil pH and base saturation gradually decreased with season in the cacao shell biochar treated plots (Fig. 2). This indicates that the fading effect of biochar on crop yield may be related to soil acidity. In particular, the exchangeable molar Ca/Al ratios, analyzed in field samples for seasons 1–3, showed largely similar patterns as grain yields, especially with biochar type and dosage, and to a certain extent with season (Fig. 2). That is, they were significantly higher for cacao shell biochar (around 1.5 at 15 t ha<sup>-1</sup> biochar dosage) than for rice husk biochar (around 0.6 at 15 t ha<sup>-1</sup> biochar dosage). Also, Ca/Al ratios were significantly higher at the high dosage of 15 t ha<sup>-1</sup> cacao shell biochar (around 1.5) than for the lower dosage of 5 t  $ha^{-1}$  of the same biochar (0.7–0.9) and higher for seasons 1 and 2 (around 1.5 at 15 t  $ha^{-1}$  cacao shell biochar) than for season 3 (around 1.0). For seasons 4 and 5 we do not have Ca/Al ratios, but as observed previously (Gruba and Mulder, 2015), pH and Ca/Al ratios are significantly correlated (Fig. S6), so here the measured pH values and their trends can be relied upon (Tables S14 and S15). The relation between pH and Ca/Al is related to the fact that base saturation (dominated by Ca, by far) is positively correlated with pH, whereas exchangeable Al is negatively correlated with pH (due to the decreasing solubility of Al in soils with increasing pH). This has been demonstrated explicitly (Gruba and Mulder, 2015).

The relation between crop yield and pH was not entirely straightforward across seasons, as e.g. pH (but not Ca/Al ratios) decreased from season 1 to 2 for both cacao shell biochar dosages (Table S4b), whereas crop yields showed a strong increase from season 1 to 2 (Table S3; Fig. 1). However, when comparing dosages, the pH effect of 5 t ha<sup>-1</sup> cacao shell biochar started to fade after season 2 (Table S4b; lowercase green letters), and the same was observed for crop yield (Fig. 1). Similar observations were made for the 15 t ha<sup>-1</sup> cacao shell biochar amendment, where the pH effect was significant for the first 4 seasons but no longer for season 5 (Table S4b), when also the crop yield effect faded and was back on the level observed during season 1 (Fig. 1).

In Fig. 3a the significant relationship between maize grain yield and Ca/Al ratios is shown (n = 75; p < 0.001;  $r^2 = 0.44$ ). The zero maize



**Fig. 1.** Maize grain yield (t ha<sup>-1</sup>) after amendment with 0, 5 or 15 t ha<sup>-1</sup> cocoa shell or rice husk biochar, over five seasons, at Lampung, Sumatra. Errors bars represent one standard error in five replicate blocks. Differences associated with biochar dosage (5 or 15 t ha<sup>-1</sup>; UPPERCASE LETTERS) and season (1–5; lowercase letters) is shown for the two biochars separately. Also see Table S3, where also differences between biochar types are indicated.



**Fig. 2.** (a) Ca/Al molar ratios and (b) pH after amendment with 0, 5 or 15 t ha<sup>-1</sup> cocoa shell or rice husk biochar, measured in field samples after planting seasons 1, 2 and 3 (Ca/Al) and planting seasons 1–5 (pH). Errors bars represent one standard error in samples from five replicate blocks. The figure shows statistical analysis of variation with biochar dosage (UPPERCASE LETTERS; 0 t ha<sup>-1</sup> vs. 5 t ha<sup>-1</sup> vs. 15 t ha<sup>-1</sup> Cacoa shell per season, and 0 t ha<sup>-1</sup> vs. 5 t ha<sup>-1</sup> vs. 5 t ha<sup>-1</sup> vs. 15 t ha<sup>-1</sup> cacoa shell per season, and 0 t ha<sup>-1</sup> vs. 5 t ha<sup>-1</sup> kice husk per season) and variation with season for the different biochar type and dose combinations separately (lowercase letters; season 1–3). Different letters indicate statistically different values (p < 0.05).

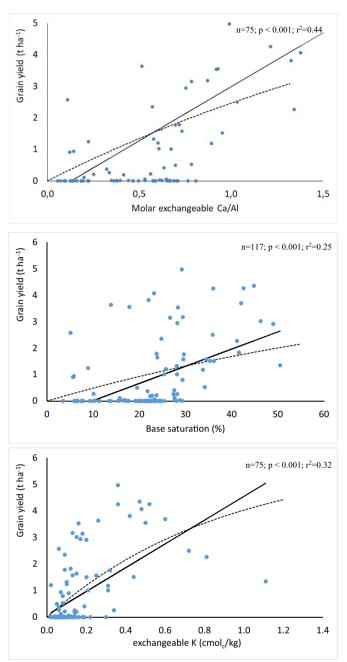
grain yield points were included as these data were actual results: as mentioned above, without biochar amendment no grain yields were obtained. p < 0.001 indicates a highly significant relationship, but, as typical for a field study, there was a lot of scatter in the observations, reducing  $r^2$ -values to 0.4–0.5 for crop yield vs. pH and Ca/Al. Similarly significant but slightly less strong relationships were observed between grain yield and the other acidity-related parameters: pH (Fig. S3), BS (Fig. 3b), and exchangeable K (Fig. 3c). Also here the zero grain yield values were included, as these were actual measurements. The same relationship was not observed for available P (Fig. S5), as P availability was not affected by pH in this low pH range between 3.5 and 5 (Fig. S7).

#### 4. Discussion

We observed a much stronger effectiveness in increasing crop yield for the cacao shell biochar than for the rice husk biochar, even though both were made in the same manner. This observation could be explained by its higher pH (9.8 vs. 8.4 for cacao shell and rice husk biochar, respectively) and ANC (217 vs. 45 cmol<sub>c</sub> kg<sup>-1</sup>; Table S1). The cacao shell biochar can thus improve the acidity-related soil properties more effectively than the rice husk one.

Recently, Gruba and Mulder (2015) showed that the exchangeable Al concentration in acid soils reaches maximum values at  $pH_{H_2O} \approx 4.2$ , while declining with pH increase. Thus, the pH for the unamended soil (3.7) was far below this threshold and thus explains the high Al availability. Soils treated with 5 t ha<sup>-1</sup> cacao shell biochar still had  $pH_{H_2O} \approx 4.2$ , still close to the Al release threshold. After addition of 15 t ha<sup>-1</sup> cacao shell biochars, pH was 4.5–5.0, well above the pH where extensive Al dissolution occurs. Since Ca concentrations increase sharply from near-zero above pH 4.2, this implies that Ca/Al ratios increase strongly with increasing pH, associated with the application of cacao shell biochar.

Our first hypothesis was that the biochars' agronomic effects could be explained by reduced soil acidity (reduced exchangeable  $Al^{3+}$ ), and that the biochar with highest ANC would give the strongest yield effects. This hypothesis was largely supported by our data, both with regard to acidity alleviation explaining the biochar effects on crop yield, and with regard to the higher-ANC biochar (cacao shell biochar) having stronger



**Fig. 3.** Maize grain yield (t ha<sup>-1</sup>) as a function of a) molar exchangeable Ca/Al ratio, b) base saturation (BS), and c) exchangeable K, for samples amended with 0, 5 or 15 t ha<sup>-1</sup> cocoa shell or rice husk biochar (five replicates), measured in field samples after planting seasons 1, 2 and 3 (Ca/Al and K) or seasons 1–5 (BS). The zero maize grain yield points were included in the graphs as these data were actual results: without biochar amendment no grain yields were obtained. Drawn lines: linear regression. Dashed lines: exponential fitting to  $y = a - e^{-bx}$ , where y is grain yield, x is x-axis variable (Ca/Al, K, BS), and a and b are fitting parameters optimized by minimizing the sum of squared residuals between measured and modeled parameters.

effects than the low-ANC one (rice husk biochar). Although confirmed in the large picture, not all individual pH observations were in agreement with the hypothesis, e.g. relatively low pH in season 2 for both cacao shell biochar dosages whereas crop yields were surging, and a significant increase in pH from season 4 to 5 for the 5 t ha<sup>-1</sup> cacao shell biochar trial. These exceptions in individual measurements are often encountered in field studies, and the differences between the two biochars, and the trends with biochar dosage, were fully in line with the acidity alleviation hypothesis.

## 4.1. Time trends and comparison with previous multiyear biochar field trials

Our most significant findings with regard to time trends of biochar effects on crop yield are i) that the biochar effect was stronger in the second season than in the first one, and ii) that the effectiveness started to fade after three to five cropping seasons, partly because the acidity alleviation effect started to fade (e.g., no significant effect on pH of either 5 or 15 t ha<sup>-1</sup> cacao shell biochar in season 5). Thus, our second hypothesis that biochar effectiveness on crop yield would decline over time, was supported by the observations. This could be due to continued nutrient leaching and rapid depletion of the alkalinity added via the biochar, and the effects varied with biochar type and dose.

We hypothesize that the first observation of initial increased effectiveness over time may be explained by "aging" of the raw, nonenriched biochar in soil, where biochar improves soil structure over time, leading to improved soil aggregation (Obia et al., 2016) as well as decreased soil density (Obia et al., 2017) and root penetration resistance (Obia et al., 2017). Spectroscopic evidence has indicated that an organic coating is formed on the biochar surface over time, resulting in improved nutrient retention and creating a more optimal habitat for soil microorganisms (Hagemann et al., 2017a; Hagemann et al., 2017b; Kammann et al., 2015). In addition, precipitation of the dissolved Al may be a slow process (Gruba and Mulder, 2008). Better crop yield effects in season 2 than immediately after application (season 1) have been observed in a few studies before (Griffin et al., 2017; Jones et al., 2012; Major et al., 2010). Major et al. (2010) studied biochar (20 t ha<sup>-1</sup>) effects in an acidic oxisol in the humid tropics of Colombia for 4 years, and did not find any strong effects in the first year, but maize yield increases of +28, +30 and +140% in the three following seasons (yields around 2 to 5 t  $ha^{-1}$ ). The authors attributed the greater crop yield primarily to increases in available Ca and Mg, but indeed the small but important increases in soil pH<sub>KCI</sub> (from 3.9 to 4.1) and corresponding increases in molar available Ca/Al ratios (from an extremely low 0.06 to around 0.12, mainly due to increased Ca but not decreased Al) probably also contributed to the observed biochar effects, similar to observations in the present study. Compared to the present study, where molar Ca/Al ratios around 0.2 resulted in almost no crop yield, the crop yields of Major et al. (2010) of >2 t ha<sup>-1</sup> in the absence of biochar, were surprisingly high. Also Griffin et al. (2017), studying walnut shell biochar over four years in a field experiment (10 t  $ha^{-1}$ ), under dry conditions in CA, USA, on a silty clay loam with pH 6.7 and CEC of 22.3 cmol<sub>c</sub> kg<sup>-1</sup>, observed positive yield effects in the second year (+8%) but not in the first one. The authors ruled out pH and nutrient retention effects, and attributed the yield effect to short-lived increases in available K through direct additions of these nutrients via the biochar (biochar often contains high K levels (Biederman and Harpole, 2013; Martinsen et al., 2014)), analogous to the present study (Fig. 3c) and other observations of higher K in both soil solution and plant tissue after biochar addition (Biederman and Harpole, 2013; Martinsen et al., 2014). In addition, moisture retention effects could have explained the biochar effect, and the best effect being in the second year could be explained by gradual improvements in soil structure (Bruun et al., 2014; Obia et al., 2016). Jones et al. (2012) investigated the 3-year effect of the amendment of 25 and 50 t ha<sup>-1</sup> biochar on maize and grass yield as well as on soil parameters, in pH-neutral (pH 6.6) sandy clay loam in Wales, UK. In accordance with our study, biochar effects were stronger in year 2 than in year 1.

The second important observation, the fading biochar effect over multiple seasons, can probably be attributed to leaching of the alkaline ashes in the humid tropical climate with high rainfall (Glaser et al., 2002; Lehmann and Rondon, 2006). Analogous to our study, Steiner et al. (2007) tested 15 different treatments over four planting seasons in an acidic soil in Brazil ( $pH_{H_2O} = 4.5$ ), and found that biochar doubled the grain yield in the presence of mineral fertilizers, but that the effect faded after multiple seasons. The authors hypothesized that the positive

vield effect resulting from the biochar amendment could partly be caused by significantly lower exchangeable Al concentrations in biochar-amended soil. However, the Ca/Al molar ratios that can be calculated for their untreated soil (around 20) were much larger than the ones observed presently (below 1), in accordance with their soil pH being above the limit of pH 4.2 where exchangeable Al contents are substantially increased (Gruba and Mulder, 2008). These results cast doubt on the hypothesis that reduced Al toxicity was the mechanism behind the biochar effect in the Brazilian study (Steiner et al., 2007). A fading effect of the biochar over time after season 2 was also observed by Jones et al. (2012): after 3 years in the field, the alkalinity associated with the biochar had been fully neutralized, similar to our observations, with the pH of the soil being back at 6.6, and the pH of the biochar itself having decreased from 8.8 to 6.6. However, the pH effect of the biochar probably was not of much importance in these near pH-neutral soils, and the authors hypothesized that the main effect of biochar was beneficial changes in the microbial community, stimulating fungal and bacterial growth as well as soil respiration.

On the other hand, in extensive four-season trials in Thailand and the Philippines with rice husk biochar on a poor, dry, nonacidic (pH 6) soil, Haefele et al. (2011) observed increased yields of 16–35%, although without any clear trends with season, and hypothesized that the increase was a result of improvements in water retention and increased available K and P, not of pH. Yamato et al. (2006) investigated the effect of biochar produced from *Acacia mangium* on maize and peanut yield and soil properties in South Sumatra (Indonesia). They observed a significant increase in yield, as well as the density of the rooting system, soil pH, total N, available P<sub>2</sub>O<sub>5</sub>, cation exchange capacity, and observed a strong reduction in soil Al<sup>3+</sup> concentration, analogous to the present study on the same island.

A meta-analysis undertaken by Jeffery et al. (2017) has shown the significant positive effect of biochar application on crop productivity, with a grand mean increase of +20-25% for tropical soils. This increase was attributed to a liming effect and improved water holding capacity of the soil, along with improved crop nutrient availability when biochar is added to soil. Biederman and Harpole (2013), in their meta-analysis reported a statically significant positive yield effect of biochar amendment of +20%, citing pH and soil quality increases (mainly N, P, K) as the main reasons for this. These conclusions have also been confirmed by a meta-regression analysis carried out by Crane-Droesch et al. (2013). They concluded that soil cation exchange capacity and organic carbon also were strong predictors of crop yield response.

Despite the fact that the previous meta-analyses have shown that biochar amendment leads to a significant effect on crop yield, individual literature studies are still quite variable, showing inconsistency between laboratory and field tests and among field studies (Gurwick et al., 2013). For example, Bass et al. (2016) investigated the effect of biochar amendment on banana and papaya crop yield and soil properties in Australia. Although there was a positive effect of biochar on soil properties, where cation exchange capacity, K<sup>+</sup>, Ca<sup>2+</sup>, soil C content and water retention all increased, there was a no effect (for papaya), or a negative effect (-18% for banana) on fruit yield. Further studies are thus necessary in order to fully understand the effect of biochar on soil properties and yield, despite the fact that increases in soil alkalinity and nutrient availability seem to be the most reasonable and statically significant explanations.

#### 5. Perspective

Comparing biochar amendment to conventional liming, the ANC of the cacao shell biochar (217 cmol<sub>c</sub> kg<sup>-1</sup>; Table S1) was about 0.2 CaCO<sub>3</sub>-equivalents (Sparks, 2012). In addition, the cacao shell biochar added 127 cmol<sub>c</sub> kg<sup>-1</sup> exchangeable + soluble K, 37 cmol<sub>c</sub> kg<sup>-1</sup> Ca and 32 cmol<sub>c</sub> kg<sup>-1</sup> Mg to the soil (Table S1). In comparison, dolomite would add much more of both Ca and Mg (around 1000 cmol<sub>c</sub> each (Sparks, 2012)), but not K. From the ANC of the cacao shell biochar it

can be calculated that minimally 3 t  $ha^{-1}$  calcite or dolomite would be needed for the same pH effect as  $15 \text{ t ha}^{-1}$  biochar. However, smallscale tests with various dolomite additions over 10 d in the same soil revealed that a higher dolomite dosage around 6 t ha<sup>-1</sup> was needed for a pH increase to 4.5–5.0, the pH after 15 t ha<sup>-1</sup> cacao shell biochar amendment. At a dolomite price of 250 to 500 US t<sup>-1</sup>, this is a major cost for the small-scale farmers in Lampung district. Biochar can be made for as little as 100 US\$  $t^{-1}$  by these smallholder farmers, especially when using clean, fast and free-of-charge flame curtain kilns (Cornelissen et al., 2016), and provides the farmer with the added advantages of K addition (Biederman and Harpole, 2013; Martinsen et al., 2014) and improvement of soil structure and microbiology (Jones et al., 2012; Obia et al., 2016), as well as the global advantages of carbon sequestration (Jeffery et al., 2015; Weng et al., 2017; Woolf and Lehmann, 2012) and nitrous oxide suppression (Lehmann, 2007; Obia et al., 2015; Weng et al., 2017).

#### 6. Conclusion

The main conclusion of our study is that the primary cause of increased crop production in an Ultisol of the humid tropics due to biochar addition was related to its acid neutralizing capacity. Thus, the role of biochar as a soil enhancer was mainly associated with its liming effect, causing a significant decline in toxic Al. Our hypothesis that this effect fades over time was supported, and thus multiple biochar amendments are necessary. In this case biochar would need to be applied approximately every third season, similar to conventional liming in the study area. Also, moderate additions of 5 t  $ha^{-1}$  biochar did not suffice for acidity alleviation, and high dosages of 15 t  $ha^{-1}$  were necessary. Such dosages are better amenable with intensive small-scale horticulture or kitchen gardening than more extensive maize farming (Torres-Rojas et al., 2011). In current controlled field trials in the study area we are comparing biochar amendment to liming and ash amendment in multi-season trials, also testing the longevity of the various amendment effects, in order to come to the best farmer recommendation for biochar implementation.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2018.03.380.

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