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Estimating potential dust emissions from biochar amended soils under simulated tillage

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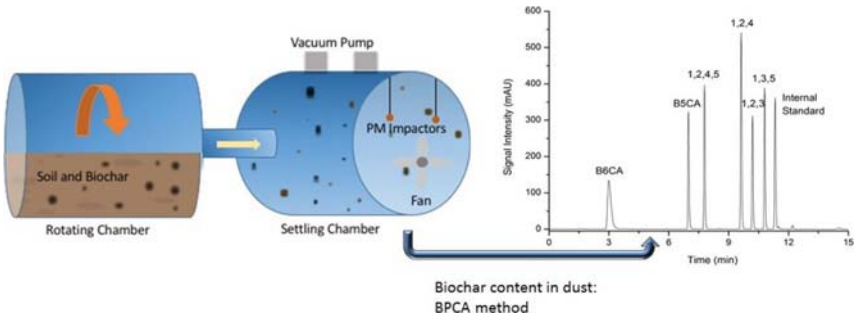
HIGHLIGHTS

- Addition of the walnut shell biochar led to markedly higher dust emissions than the pure soils.
- Increasing walnut shell biochar content in soil may enhance particle dispersion
- Maintaining high moisture content reduced biochar and soil emissions in dust.
- PM_{10} dust contains less biochar particles than PM_{100}.

GRAPHIC ABSTRACT



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ABSTRACT

Although biochars may provide agricultural benefits, the potential risks related to agricultural dust emissions have not been adequately investigated. This study examines the impact of biochar type (WS 900: walnut shell, 900 °C; PW 500, PW 700 and PW 900: pine wood, 500, 700, 900 °C), biochar application rate (0, 1, 2, 5% wt.) and soil water content (low, medium and high) on dust emissions in two different textured-soils (silt loam, sandy loam). Dust was produced via a dust generator simulating soil disturbance (e.g, tillage) and dust fractions with an aerodynamic diameter under 100 μm and 10 μm (PM_{100} and PM_{10}) were collected. The data indicate that the higher application rate of WS 900 led to higher PM_{100} and PM_{10} emissions while PW biochar treatments emitted equivalent amounts of dust as controls (non-amended soils). Dust emissions were exponentially reduced as soil water content increased, irrespective of biochar's presence. Specific markers for biochar, benzene polycarboxylic acids (BPCAs), were used to estimate the biochar content within dust. Results indicate that the increased dust emissions from WS 900 treatments mainly derive from soil particles due to the greater dispersion potential of WS 900 biochar. The collected data also reveal that PM_{10} dust contains less biochar particles than PM_{100}, attributed to biochars originally containing negligible amounts of particles <10 μm.

1. Introduction

Agricultural dust is the largest contributor to airborne particles in intensively farmed regions (Madden et al., 2008; Madden et al., 2010) and has been long recognized as an occupational health hazard for farmers, increasing the risk for respiratory symptoms (Lee et al., 2004; Schenker, 2000). Dust emission by standard farming operations, particularly tillage, can be four to six times greater than by natural wind erosion (Funk et al., 2008; Goossens, 2004; Goossens et al., 2001). Dust emission under different farming practices, therefore, has been extensively studied across a variety of cropping systems and environmental conditions, such as oilseed crops in winter wheat (*Triticum aestivum* L.) (Sharratt and Schillinger, 2014), cotton (*Gossypium hirsutum* L.)-tomato

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(*Lycopersicon esculentum* Mill.) rotation (Baker et al., 2005), or lettuce (*Lactuca sativa* L.) (Madden et al., 2009). Meanwhile, efforts have been devoted to improving methodology in estimating dust discharges from agricultural fields (e.g. Hagen et al., 2010). Among different studies, a unified standard for airborne particulate matter has been established, with three particle-size fractions related to public health that include: 1) the inhalable-particulate fraction (particle equal or smaller than 100 μm in diameter, or PM_{100}), representing all particles that could possibly enter the body through nose and mouth; 2) the thoracic fraction, which has an aerodynamic diameter equal or below 10 μm, or PM_{10}; and 3) the respirable-dust fraction with a size equal or under 4 μm. Mammalian health risks increase with decreasing particle size, as smaller particles can reach the alveolar region of the lungs and are difficult for the body to remove naturally (Clausnitzer and Singer, 1996).

Soil amendments have been seldom investigated individually regarding their contributions to agricultural dust emissions since they are usually applied at low rates to soils and therefore are not considered as major components of the soil. Although biochar, an amendment produced through biomass thermal conversion, has been gaining attention as it may offer some agricultural benefits, the potential risks towards public health are yet to be fully elucidated (Verheijen et al., 2010). Most biochars have low bulk densities and are highly porous with a wide range of particle sizes (Chia et al., 2015). These features render biochar susceptible to mechanical disturbance and release into ambient air as aerosols, while potentially carrying toxic chemicals adsorbed to the surfaces, including heavy metals, organics and other pollutant residues (Beesley and Marmiroli, 2011; Bushnaf et al., 2011; Cao et al., 2011; Chai et al., 2012; Chen and Yuan, 2011; Chen et al., 2011). Increased PM_{10} emission has been observed when adding biochar to sand and two soils using a wind tunnel experiment, which simulates the wind erosion process (Ravi et al., 2016). Two possible mechanisms are responsible for this accelerated emission including an emission of native fine biochar particles and an emission of fine particles by abrasion of large biochar particles. Therefore, it is of equal importance to investigate the influence of biochar on the amount of dust (soil and biochar borne) generated under tillage practices, and to better understand the possible risks to human health associated with these dust emissions.

Like other soil amendments, biochar is a class of materials with a variety of commercialized types (Glover, 2009). Generally, biomass feedstock and production technique are the most significant features influencing the physical and chemical characteristics of the resultant biochars. Particle size distribution of biochars, for example, exhibit a trend towards smaller particle sizes with increasing highest treatment temperature (HTT) when using sawdust and woodchips as feedstocks (Downie et al., 2009). Similarly, with consistent pyrolysis conditions, the use of different feedstocks results in variations in solid bulk densities of biochars, and a linear relationship between the bulk densities of biochars and densities of the corresponding feedstocks has been observed (Byrne and Nagle, 1997). Additionally, the quantity of biochar added to the soil can be varied to achieve optimal effects regarding crop yield increase, toxin remediation, soil microbial activity enhancement and other desirable benefits. Many studies suggest that higher biochar application rate can result in greater agricultural benefits (Jeffery et al., 2011; Major et al., 2010; Namgay et al., 2010; Solaiman et al., 2010), with application rates as high as 135 t h^{-1} (Jeffery et al., 2011).

Land applied biochar is also influenced by soil variables, such as soil moisture and soil texture, which have been suggested to play significant roles on dust emissions. For example, increasing soil moisture (characterized by soil gravimetric water content, or GWC) is an effective and simple method reducing dust emissions (Funk et al., 2008; Madden et al., 2010). However, the soil GWC threshold values, where small increases in soil moisture can cause a distinct reduction of dust emission, are highly dependent on soil texture (Carvacho et al., 2001; Madden et al., 2010). Although it has been suggested that biochar should be applied in slurries and mixed with soils to avoid dust formation (Sigmund et al., 2017), no studies, to the best of our knowledge, provide the evidence that wetting biochars is equally effective in different soil systems.

Overall, it is unrealistic to generalize the impact of biochar on agricultural dust emission without considering properties or conditions possessed by both biochar and soil. To address this important knowledge gap, the objectives of this study are to 1) determine whether the addition of biochar including its type and biochar application rate, influences the dust emission from different textured agricultural soils; 2) examine the effectiveness of increasing biochar moisture in different textured soils as a strategy to minimize the occupational hazards towards dust exposure.

2. Materials and methods

2.1. Soils and biochars

Soils used in this study included a fine-textured soil, Yolo (silt loam [SiL], Mollic Xerofluvent, Davis, CA) and a coarse-textured soil, Grangeville (sandy loam [SL], Fluvaquentic Haploxeroll, Lockeford, CA). The cropping system where SiL was sampled is an annual crop rotation of processing tomato (*Lycopersicon esculentum* Mill.) and corn (*Zea mays* L.); whereas, SL is a rangeland soil. Soils were sampled from 0 to 15 cm. Air-dried soils were ground and passed through a 2 mm sieve prior to use to prevent mechanical damage of the dust generator. Physical and chemical properties of both soils can be found in Table S1 (Supplementary Material).

A thermo-sequence of ponderosa pine wood biochar (HTT 500, 700, 900 °C, slow pyrolysis, 4 h) and walnut shell biochar (HTT 900 °C, gasification, 4 h) were used in this study (abbreviated as PW 500, PW 700, PW 900, WS 900, respectively). The physical and chemical characteristics of the biochars can be found in Table S2 and all characterization methods are described in Mukome et al. (2013). All biochars were passed through a 2-mm sieve without grinding and only the <2 mm fraction was used in this study.

2.2. Experimental design

Three individual experiments were included in this study and controls in these experiments were all non-amended soils. Experiment I examined the effect of four types of biochar, at the same application rate and soil moisture, on dust emissions. WS 900 markedly increased dust emissions when applied to both soils while this phenomenon was not observed for other biochar types (details in Section 3.1); therefore, subsequent experiments were conducted with WS 900. A biochar application rate gradient (four levels) was established in Experiment II. Experiment III investigated the effectiveness of increasing moisture as a dust control strategy in soils amended by WS 900 at a fixed application rate. Three soil gravimetric water contents (GWCs) were chosen to produce a range of moisture content (low, medium, high) determined via soil water potential curves (Fig. S1). The soil matrix potential of −1500 kPa (nominal permanent wilting point) is the boundary of moist and dusty/dry appearance of soils (Brady and Weil, 2008). In addition, a recent study found that the threshold friction velocity dramatically decreases when the matrix potentials are below −1 MPa (−1000 kPa) (Sharratt et al., 2013). Therefore, the GWC value at the permanent wilting point was chosen as the medium moisture level for this study. A GWC value less than this medium level (specific values listed in Table 1) was adopted as the low moisture level while the field capacity, or the GWC at −33 kPa (representing the maximal amount of capillary water held by soil pores) was used as the high moisture content. Moisture adjustments were achieved through spraying groundwater (sampled from Russell Ranch Sustainable Agricultural Research Facility) into air-dried soils (in plastic bags). Daily re-spraying (until target weight) with mechanical disturbance was performed to

Table 1
Summary of the experiments conducted in this study.

	Brief Description
Experiment I	–Treatments: WS 900, PW 900, PW 700 and PW 500 at 20 g kg^{-1} (2%) to both soils (SiL and SL) –Low moisture content maintained (where dust emissions were the greatest) for biochar-amended soils and corresponding controls (0 g biochar kg^{-1} soil) –SiL and SiL + biochar GWC: 60 g kg^{-1}; SL and SL + biochar GWC: 30 g kg^{-1}[a]
Experiment II	–Treatments: 0, 10, 20 and 50 g kg^{-1} (0, 1, 2, 5%) WS 900 to both soils (SiL and SL) –Low moisture content maintained and GWCs described in Experiment I
Experiment III	–Treatments: low, medium and high soil moisture level using WS 900 at 20 g kg^{-1} to both soils (SiL and SL) —At low moisture: SiL and SiL + biochar GWC: 60 g kg^{-1}; SL and SL + biochar GWC: 30 g kg^{-1}[a] —At medium moisture: SiL and SiL + biochar GWC: 170 g kg^{-1}; SL and SL + biochar GWC: 80 g kg^{-1}[a] —At high moisture: SiL GWC: 250 g kg^{-1} and SiL + biochar GWC: 290 g kg^{-1}[b]; SL GWC: 140 g kg^{-1} and SL + biochar GWC: 180 g kg^{-1}[b]

[a] Biochar, irrespective of its application rate (Fig. S1a and S1c) or type (Fig. S1b and S1d), did not increase the GWC at −1500 kPa compared to the soil controls. Therefore, neither the low nor medium moisture level was different between soil controls and the biochar-amended treatments.

[b] WS 900 (applied at 2% wt.) enhanced field capacities for both soils (Fig. S1b and S1d), compared to the corresponding controls; therefore, GWC values for the high moisture levels were different between the biochar-amended and non-amended soils. Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell; PW, pine wood.

promote even water distribution for at least seven days. Soil GWC was measured prior to dust sampling to ensure the desirable soil moisture content. A summary of the experimental design is in Table 1.

2.3. Dust collection

Dust samples were collected using a dust generator designed at the University of California, Davis (Domingo et al., 2010). The dust generator consists of a rotating chamber (diameter 46 cm × length 76 cm) and a settling chamber (diameter 60 cm × length 87 cm). Simulation of dust generation can minimize logistical problems resulting from spatial or temporal variability in soil properties, field operations, and meteorological conditions associated with field sampling and thereby provide an effective comparison basis. In addition, unlike dust generators that can only suspend a tiny amount of source materials (1 mg to 1 g) (Carvacho et al., 2001; Carvacho et al., 2004; Chow et al., 1994), this dust generator generates a large cloud of dust in continuous plumes, which is suitable for simulating tillage operations rather than natural wind erosion (Domingo et al., 2010). Operating conditions for this generator have been optimized (Domingo et al., 2010) and these parameters were adopted for the current study. Briefly, 300 g of soil or soil/biochar mixture, at desired moisture content, was placed in the chamber and tumbled for 3 min with a drum rotating speed of 16 rpm. The dust generator was cleaned before each sample run using a vacuum cleaner and compressed air, followed by wiping of all interior portions with 2-propanol (Madden et al., 2010). An ambient background was measured before the dust generation each day using the same operational parameters.

Only the inhalable (PM_{100}) and thoracic fraction (PM_{10}) of the dust were collected for each sample in this study since the respirable (or smaller size) fraction did not produce sufficient samples for subsequent analysis. The inhalable fraction was sampled using an Institute of Occupational Medicine (IOM) 100 μm median cut point sampler (Edinburgh, UK) operated at 2.0 L min^{-1}. The thoracic fraction was sampled with a Model 200 Personal Environmental Monitor (PEM) operated at 4.0 L min^{-1} (MSP Corporation, Shoreview, MN) Gilian pumps (Wayne, NJ) were used to achieve desirable vacuum flow rates during dust sampling and were calibrated before and after dust sample collection with an SKC Ultraflo electronic airflow meter (BIOS Inter. Corp., Butler, NJ) to ensure uniform flow throughout sampling. Samples were discarded and re-run if the difference between pre- and post-sampling flow rates was >0.3 L min^{-1}. Details regarding the dust samplers can be found in Mark and Vincent (1986) and Domingo et al. (2010).

2.4. Measurement of airborne dust mass concentration

Dust samples were collected on Teflon filters (with 2.0 μm openings, Pall Corp., Washington, NY) of appropriate size: 37-mm diameter filters for the PEM samplers and 25-mm filters for IOM samplers (Domingo et al., 2010). Filters from PEM samplers were weighed before and after dust collection using a microbalance (C-35; Cahn, Paramount, CA) with a sensitivity of 1 μg. Mass of dust collected on the filter was obtained and the airborne PM_{10} mass concentration was calculated based on this mass and the total sampling air volume. For the PM_{100} fraction, the cassette assembly inside the IOM sampler and the filter were weighed together and the airborne PM_{100} mass concentration was calculated in a similar way to PM_{10}. Following weighing, the dust was removed from Teflon filters via ultra-sonication (Bransonic B-12; Branson Ultrasonics Corp., Danbury, CT) in 2 propanol (Optima grade). The resulting 2-propanol containing dusts were frozen and freeze-dried (VirTis Freeze Mobile 25EL; SP Industries, Inc., Warminster, PA). Dry samples were collected into amber vials for storage at 4 °C prior to analysis.

2.5. Estimated quantification of biochar content in dust

2.5.1. Method selection and rationale

Determination of the relative biochar content in dust utilized a specific molecular marker method via measuring benzene polycarboxylic acids (BPCA) produced from nitric acid sample digestion (Glaser et al., 1998). Biochars do not have their own isolating, characterizing or quantifying techniques, but rather share methods used for quantifying pyrogenic carbonaceous materials in different matrices (Lehmann and Joseph, 2015; Poot et al., 2009). An extensive comparison of various techniques and matrices can be found in Hammes et al. (2007) where they concluded that no method is without some shortcomings. Nevertheless, the BPCA method was chosen for this study as our assessment of the literature revealed it to be the most advantageous. Additionally, our preliminary testing revealed this method was best at minimizing interferences from soil matrices, including the natural soil organic carbon (compared to the thermal/optical methods) and metal ions in clay minerals (compared to the nuclear magnetic resonance spectroscopy).

2.5.2. Method description

The BPCA method used in this study was adapted from Wiedemeier et al. (2013). Briefly, samples containing >1 mg total organic carbon (TOC) were weighed and digested directly with nitric acid (68–70%, 8 h at 170 °C). The resulting BPCA solution was filtered through ashless cellulose filter paper (Whatman 589/3) and further cleaned by a cation exchange resin column to remove polyvalent ions (Dowex 50WX8, 200–400 mesh). The clean solution was freeze-dried and the residue was redissolved in methanol/water (1:1) and eluted over a C18 solid phase extraction cartridge (Supelco Discovery, 3 mL). The final eluate was dried again and transferred to HPLC vials in 18.2 MΩ·cm water (Barnstead Nanopure) for analysis. Phthalic acid was used as the internal standard for quantification of all BPCAs. Chromatographic BPCA separation conditions and quality controls are provided in the Supplementary Material.

2.5.3. Correlation of BPCA-C with biochar content

Glaser et al. (1998) first proposed to correlate BPCA carbon mass to black carbon (BC) content by using a conversion faction of 2.27 because they found approximately 45% of BC carbon can be converted into BPCA carbon after digestion. Subsequently, this conversion factor has been shown to often under-estimate BC due to some BPCA artefact formation during the pre-treatment and a conversion factor of 4.5 has been suggested (Brodowski et al., 2005). However, recent studies indicate that the application of a single conversion factor for different types of charcoal is not appropriate (Schneider et al., 2010). Therefore, in our current study, we developed biochar specific conversion factors (Table 2) to estimate the relative biochar content for different biochar types. Each conversion factor is the reciprocal of the ratio of BPCA carbon to total organic carbon (TOC, before digestion), based on the measured data ($n = 3$).

2.6. Determination of the dispersive potential caused by biochar

Initial data revealed that some biochars may contribute to soil dispersion and thus lead to increased dust emissions. In order to explain this observed phenomenon the dispersive potential of the biochars (< 2 mm fraction) was evaluated in the context of their extractable (aqueous and exchangeable) salts. Aqueous cations (K^+, Ca^{2+}, Mg^{2+}, Na^+) in biochars were extracted using deionized water and biochars were separated from water extracts. Biochars were sequentially extracted by ammonium acetate solution buffered to pH 7.0 to obtain the exchangeable cations. Cations were quantified by inductively coupled plasma atomic emission spectrometry (ICP-AES) (Thomas, 1982). This method has detection limits of approximately 0.01 cmol kg^{-1} for each cation.

The presence of monovalent cations, i.e. sodium (Na^+) and potassium (K^+), in soil solution can cause clay dispersion effects (Goldberg et al., 1988; Levy and Torrento, 1995) while divalent cations, including calcium (Ca^{2+}) and magnesium (Mg^{2+}) flocculate clays in soil (Curtin et al., 1994; Yousaf et al., 1987). However, it is found that the dispersive effects of Na^+ and K^+ are not the same in soil, neither are the flocculating effects of Ca^{2+} and Mg^{2+} (Quirk and Schofield, 1955; Rengasamy, 1998; Rengasamy et al., 1986). In this study, the cation ratio of structural stability (CROSS), proposed as an index of soil stability for mixed cation combinations in soil solution, was used to quantify the dispersive potentials of different biochars.

$$\mathrm{CROSS} = \left(\mathrm{Na^+} + 0.56\,\mathrm{K^+}\right) / \left[\left(\mathrm{Ca^{2+}} + 0.6\mathrm{Mg^{2+}}\right)/2\right]^{1/2} \quad (1)$$

Coefficients for K^+ and Mg^{2+} are determined based on the differential flocculating powers of the four cations (Rengasamy, 2002). Briefly, CROSS assumes that Na^+ has 1.8-fold dispersive power than K^+ while Mg^{2+} only has 60% flocculating power of Ca^{2+}.

2.7. Biochar particle size distribution

The International Biochar Initiate (IBI) guidelines suggest a dry sieving method to determine biochar's size distribution (with a lower end of 0.25 mm) but a standardized method measuring biochar particle at micro-meter scale is lacking (IBI, 2014). Therefore, in the present study, biochar particles with diameter under 100 μm were separated from the <2 mm fraction using a sieve with an opening diameter of 100 μm. To determine biochar particle size distributions for the <100 μm fraction, scanning electron microscopy (SEM; FEI XL 30, USA) at 20 kV was used. The masses of <100 μm and 100 μm - 2 mm fractions were individually measured and the corresponding percentages (on a mass basis) were calculated. Diameters of biochar particles under 100 μm were determined by Fiji image processing package in ImageJ using the biochar particle images obtained by SEM. A minimum of 2000 particles were measured and counted for each biochar and size distributions were on a number basis.

Table 2
Conversion factors from BPCA carbon to biochar used in this study.

	WS 900 °C	PW 900 °C	PW 700 °C	PW 500 °C
TOC (%)	65.7 ± 1.7	75.9 ± 3.9	81.9 ± 2.8	70.1 ± 0.9
BPCA-C/TOC (%)	23.6 ± 1.2	19.0 ± 1.5	17.2 ± 1.2	16.3 ± 0.4
Conversion factor	4.24	5.26	5.81	6.13

Values represent the mean ± standard error (n = 3). Abbreviations: WS, walnut shell; PW, pine wood.

2.8. Statistical analyses

All data were analyzed with JMP (Version 11.1). Statistically significant differences were tested at a 5% significance level. The Shapiro-Wilks and Levene's tests were used to verify the normality and homogeneity of variance assumptions required of ANOVA. The transformation was performed if the original data failed to meet the assumption. For each response variable, ANOVA was performed testing soil texture and biochar treatment factors and their interaction. If there was a significant interaction between factors, ANOVA was performed for each soil separately (p values in Table S6, SI).

3. Results

3.1. Impacts of using different biochar types

Addition of 2% WS 900 biochar resulted in a pronounced increase in dust emissions compared to controls. Specifically, SiL amended with 2% WS 900 emitted 61% more ($p = 0.0121$) PM_{100} (Fig. 1a) and 45% more (but not significantly, $p = 0.2700$) PM_{10} (Fig. 1b) than non-amended SiL. For SL, addition of 2% 900 WS led to 59% more ($p = 0.0002$) PM_{100} (Fig. 1a) and 78% more ($p = 0.0027$) PM_{10} (Fig. 1b) than the control. In contrast, adding 2% PW biochars (regardless of different HTTs) to soils emitted statistically equal amount of PM_{100} and PM_{10} compared to soil controls. In addition to measuring the total dust emissions, it is important to consider the amount of biochar present in the dust fractions. Dusts generated from 2% biochar-treated soils, regardless of biochar type, showed pronounced higher BPCA levels (Fig. S3a and S4a) after digestion, and therefore contained higher estimated biochar contents (Fig. 2a and b), than controls. The biochar content in the PM_{100} fraction from SL treated with PW 500 biochar was 1.9-fold higher than that

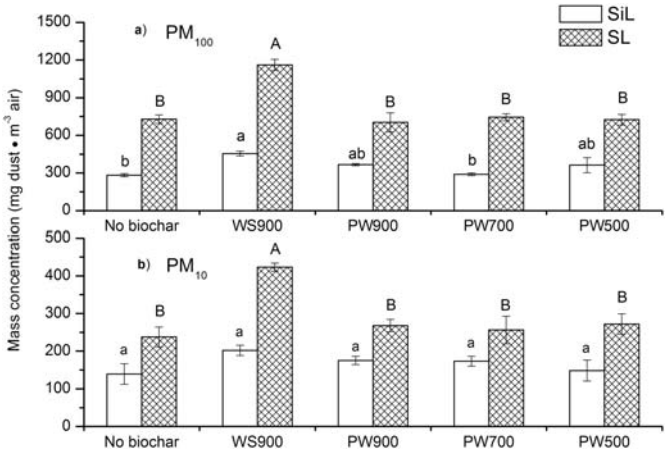

Fig. 1. Effect of biochar type (2% biochar, low soil moisture) on a) airborne mass concentration of PM_{100}; b) airborne mass concentration of PM_{10}. Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error (n = 3). Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell; PW, pine wood.

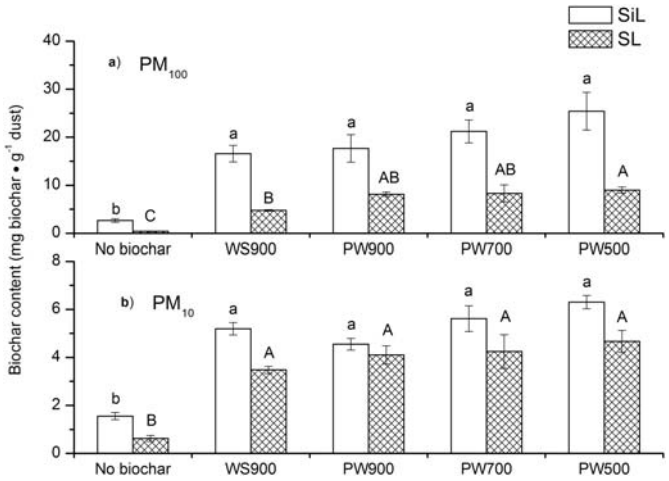

Fig. 2. Effect of biochar type (2% biochar, low soil moisture) on a) estimated biochar content in PM_{100}; b) estimated biochar content in PM_{10}. Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error ($n = 3$). Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell; PW, pine wood.

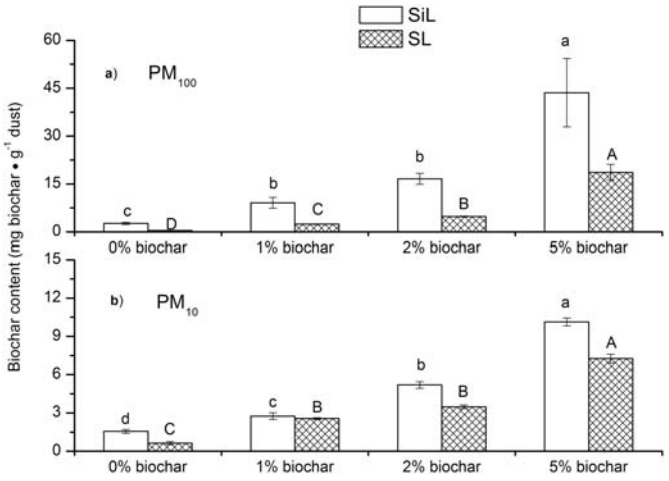

Fig. 4. Effect of application rate (WS 900 biochar, low soil moisture) on a) estimated biochar content in PM_{100}; b) estimated biochar content in PM_{10}. Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error ($n = 3$). Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell.

from the WS 900 treatment (Fig. 2a). However, biochar contents in PM_{10} were statistically the same, irrespective of biochar types, as long as the soil texture and the application rate in soil were fixed (Fig. 2b).

3.2. Impacts of increasing biochar application rate in soil

In general, increasing the WS 900 biochar application rate to soils resulted in greater amounts of PM_{100} and PM_{10} emissions (Fig. 3a and b). However, difference in dust emissions between biochar treatments and corresponding controls were not overt at a relatively low application rate. Specifically, application rates of 1% WS 900 biochar, regardless of soil texture, emitted equivalent amount of dust to SiL or SL controls. Increase in dust emissions became pronounced at the 2% application rate (one exception: PM_{10} from SiL mixed with 2% WS 900). WS 900 at the 5% application rate led to the greatest total emissions: 2.9-fold PM_{100} and 3.9-fold PM_{10} for SiL and 2.3-fold PM_{100} and 3.1-fold PM_{10} for SL. BPCA analyses (Figure 3Sb and 4Sb) indicate that higher application rates of WS 900 resulted in higher estimated biochar content within both PM_{100} (Fig. 4a) and PM_{10} (Fig. 4b).

3.3. Impacts of increasing soil moisture content

Increasing soil moisture content from 'low' to 'medium' for SiL treatments reduced 96–98% PM_{100} or PM_{10} emission, irrespective of the presence of biochar (Fig. 5a and b). For the SL treatments, at the medium moisture level, 90% PM_{100} (Fig. 5a) and 80% PM_{10} (Fig. 5b) can be reduced compared to the low moisture level. However, dust emissions were still detectable for SL treatments at high moisture contents. In contrast, emissions for the SiL treatments were negligible when soil moistures were at the medium and high levels and insufficient dust samples were collected to perform biochar content analyses. For SL treatments, biochar content in PM_{100} at high moisture level was 1.4-fold greater than the low moisture level (Fig. 6a) and it was 1.3-fold in PM_{10} fraction (Fig. 6b).

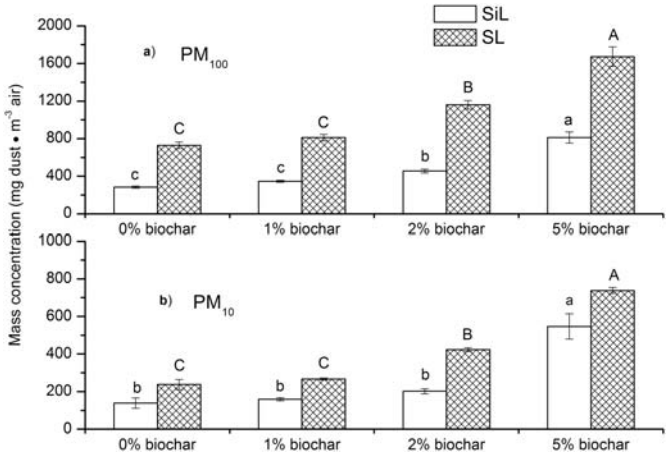

Fig. 3. Effect of application rate (WS 900 biochar, low soil moisture) on a) airborne mass concentration of PM_{100}; b) airborne mass concentration of PM_{10}. Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error ($n = 3$). Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell.

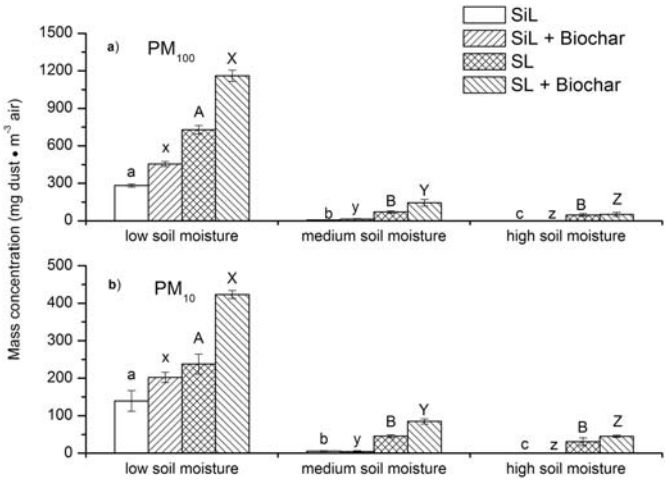

Fig. 5. Effect of soil moisture content (2% WS 900 biochar) on a) airborne mass concentration of PM_{100}; b) airborne mass concentration of PM_{10}; Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error ($n = 3$). Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell.

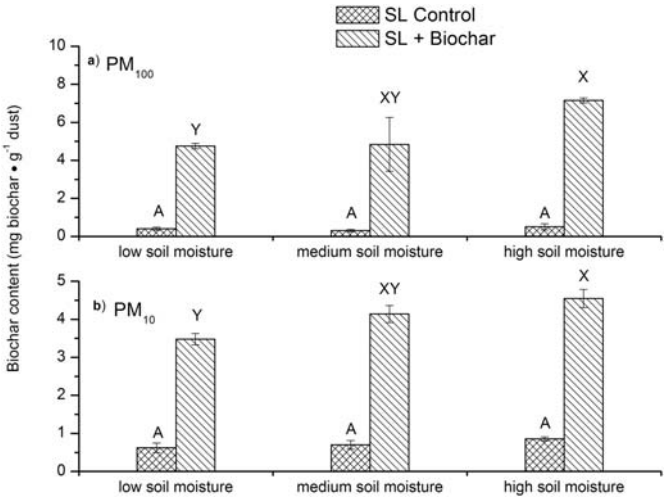

Fig. 6. Effect of soil moisture content (2% WS 900 biochar) on a) estimated biochar content in PM_{100}; b) estimated biochar content in PM_{10}. Statistical comparisons can only be made between the same letter case (significance level at $P \leq 0.05$). Values represent mean ± standard error ($n = 3$). Abbreviations: SL, sandy loam; WS, walnut shell.

4. Discussion

4.1. Detection of BPCA in the silt loam used in this study

Low levels of BPCA were detected in the dust derived from the SiL control (Fig. S3a and S4a), indicating that the SiL contains trace amounts of biochar and/or other non-biochar black carbon (BC) sources (false positive interference). The SiL was sampled from Russell Ranch (Davis, CA) where some of the plots received WS 900 for four years prior to sampling (Griffin et al., 2017). Although the SiL was sampled from an area that was not previously amended with biochars intentionally, nor directly adjacent to amended plots, mellitic acid (B6CA) was widely detected (BPCA chromatograms not shown) in different areas of Russell Ranch. It is possible that biochar particles were suspended and transported (e.g. via wind) from biochar-applied zones and re-deposited on other non-amended sites. Non-biochar BC sources (e.g. soot particles) are also possible either due to in-situ vegetation fires and/or fossil fuel burning or via atmospheric deposition (Eckmeier et al., 2013).

4.2. Dispersive effect of WS 900 biochar in soils

The mass concentrations, for each experiment, of airborne biochar particulate matter (PM) and those of soil (mineral) PM were calculated by utilizing the overall dust emissions and estimated biochar contents within dust samples (Table 3). The increase in total dust emissions, for the 2% WS 900 treatment, was mainly attributed to soil particles (contributions were 96–99% depending on soil texture and dust size fraction) while the contribution of biochar particles was negligible. Addition of PW biochars (regardless of HTTs), however, did not cause a significant increase in dust emissions from soil compared to controls. Both soils used in this study contain minimal extractable (aqueous and exchangeable) Na^+ of K^+ while WS 900 contains 48.1 g kg^{-1} extractable K^+, which is 121-fold higher than SiL and 261-fold higher than SL (Table 4). Therefore, it is hypothesized that WS 900 introduces substantial amounts of clay dispersants (mono-valent cations) and enhances soil microaggregate dispersion, generating more PM_{100} or PM_{10} mineral particles compared to the PW biochar treatments or controls (Kumari et al., 2017). To compare the dispersive potentials of different biochars, Eq. (1) is applied. Also, we artificially define CROSS BC_A and CROSS BC_{EX} to differentiate between the dispersive potential of aqueous cations and exchangeable cations since the former is more readily released as soil solution ions than the later (existing on biochar surfaces). Results indicate that WS 900 has both higher CROSS BC_A and CROSS BC_{EX} values than PW biochars (Table 4); whereas, the CROSS BC_A or CROSS BC_{EX} values for three PW biochars are similar. In addition, CROSS BC_A to CROSS BC_{EX} ratios range from 7.2 to 14.7 for PW biochars (high exchangeable Ca^{2+}) while the ratio for WS 900 is only 1.4 (high exchangeable K^+), suggesting that the flocculating potential for PW biochars can gradually increase with time. Therefore, it is important to consider CROSS BC_{EX} since change in soil stability is a gradual, rather than an abrupt, process (Jayawardane et al., 2011).

Table 3
Separation of biochar and soil particle emission in dust.

PM_{100} fraction				
	Silt Loam (SiL)		Sandy Loam (SL)	
Experiment I: 2% biochar, low soil moisture				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
No Char	0.74 ± 0.08	282.5 ± 12.3	0.30 ± 0.08	728.5 ± 34.3
WS 900	7.54 ± 0.84	447.2 ± 19.1	5.51 ± 0.16	1155.3 ± 45.0
PW 900	6.52 ± 1.14	360.3 ± 7.8	5.65 ± 0.41	697.4 ± 75.8
PW 700	6.11 ± 0.45	284.4 ± 11.0	6.09 ± 1.07	738.2 ± 29.3
PW 500	8.79 ± 0.55	354.3 ± 59.5	6.56 ± 0.79	718.6 ± 41.5
Experiment II: WS 900, low soil moisture				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
0%	0.74 ± 0.08	282.5 ± 12.3	0.30 ± 0.08	728.5 ± 34.3
1%	3.16 ± 0.65	343.7 ± 11.2	1.96 ± 0.15	809.4 ± 34.2
2%	7.54 ± 0.84	447.2 ± 19.1	5.51 ± 0.16	1155.3 ± 45.0
5%	34.07 ± 5.58	777.2 ± 69.9	31.71 ± 6.11	1641.0 ± 98.1
Experiment III: 2% WS 900 biochar				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
Low Soil Moisture	7.54 ± 0.84	447.2 ± 19.1	5.51 ± 0.16	1155.3 ± 45.0
Medium Soil Moisture	n.a.	n.a.	0.69 ± 0.20	144.1 ± 27.2
High Soil Moisture	n.a.	n.a.	0.37 ± 0.12	51.0 ± 16.0
PM_{10} fraction				
	Silt Loam (SiL)		Sandy Loam (SL)	
Experiment I: 2% biochar, low soil moisture				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
No Char	0.21 ± 0.02	139.0 ± 27.4	0.15 ± 0.03	237.6 ± 26.7
WS 900	1.05 ± 0.08	200.8 ± 13.7	1.48 ± 0.10	421.6 ± 10.6
PW 900	0.79 ± 0.02	174.6 ± 11.0	1.10 ± 0.14	266.9 ± 16.5
PW 700	0.96 ± 0.03	172.3 ± 13.0	1.06 ± 0.13	255.2 ± 36.7
PW 500	0.95 ± 0.20	147.6 ± 27.7	1.25 ± 0.08	270.7 ± 27.5
Experiment II: WS 900, low soil moisture				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
0%	0.21 ± 0.02	139.0 ± 27.4	0.15 ± 0.03	237.6 ± 26.7
1%	0.44 ± 0.06	158.7 ± 8.7	0.68 ± 0.03	266.5 ± 5.2
2%	1.05 ± 0.08	200.8 ± 13.7	1.48 ± 0.10	421.6 ± 10.6
5%	5.58 ± 0.84	541.1 ± 66.9	5.36 ± 0.25	732.7 ± 16.1
Experiment III: 2% WS 900 biochar				
	Biochar	Soil	Biochar	Soil
	mg particles m^{-3} air			
Low Soil Moisture	1.05 ± 0.08	200.8 ± 13.7	1.48 ± 0.10	421.6 ± 10.6
Medium Soil Moisture	n.a.	n.a.	0.41 ± 0.06	84.3 ± 6.9
High Soil Moisture	n.a.	n.a.	0.18 ± 0.00	44.7 ± 3.1

Values are represented as mean ± standard error ($n = 3$). Abbreviations: WS, walnut shell; PW, pine wood; n.a., not available due to insufficient sample collected.

Table 4
Major mono- and di-valent cations in soils and biochars used in this study and corresponding soil stability parameters based on measured cations.

	Aqueous (meq/100 g)				CROSS BC_A	Exchangeable (meq/100 g)				CROSS BC_{EX}
	K^+	Na^+	Ca^{2+}	Mg^{2+}		K^+	Na^+	Ca^{2+}	Mg^{2+}	
SiL	0.13	0.34	0.00	0.93	–	0.10	0.68	10.43	15.73	–
SL	0.02	0.11	0.04	0.04	–	0.01	0.36	7.92	1.50	–
PW 500	4.59	2.72	1.22	2.47	4.55	1.99	0.11	27.93	0.11	0.31
PW 700	4.18	2.09	1.21	1.24	4.48	1.97	0.30	23.39	0.03	0.41
PW 900	3.53	1.69	1.18	0.79	4.02	2.57	0.18	16.44	0.00	0.56
WS 900	99.23	13.74	61.91	36.49	10.71	24.14	1.31	6.36	1.55	7.77

Abbreviations: SiL, silt loam; SL, sandy loam; WS, walnut shell; PW, pine wood; CROSS BC_A, cation ratio of structural stability value based on aqueous cations in biochars; CROSS BC_{EX}, cation ratio of structural stability value based on exchangeable cations in biochars.

The use of CROSS is explored as an approach for explaining observed results, and although satisfying results were obtained, application of Eq. (1) to a biochar-soil system requires further evaluation. A major limitation is that the concentrations of the ions measured (Na, K, Ca and Mg) are expressed in $mmol_c/L$ solution in the original equation (Rengasamy and Marchuk, 2011), while results obtained in our study are expressed in $mmol_c/100$ g biochar. Therefore, transfer rates of the four different cations from biochar substrate to soil solution are assumed equal, which may not be always valid due to the unknown interaction between cations in biochars and those in soils. Nevertheless, for the current framework, Eq. (1) provides a useful tool for estimating the dispersive or flocculating potential for different biochars.

4.3. Increased mutual repulsion among particles caused by higher application rate of WS 900

Neither the soil (mineral) dust particle nor the biochar particle emissions linearly increase with the higher application rate of WS 900. Instead, convex function correlations are observed between airborne WS 900 particle mass concentration (Fig. S5) or soil particle mass concentration (Fig. S6) and the biochar application rate. This non-linear increase in the instability among particles can be attributed to the increased pH with more WS 900 applied to the soils. When homo- or hetero-aggregation among mineral phases in soil and biochar surfaces occurs, the stability of aggregates (especially microaggregates, 20–250 μm in size (Six et al., 2004) is strongly influenced by solution pH (Petosa et al., 2010). In alkaline environments, dissociable surface groups, such as broken –OH edges at soil mineral or biochar surface sites, will be formed, leading to negatively charged surfaces (Tombacz et al., 2004). At higher pH, the net negative surface charge increases, producing stronger repulsive forces between like-charged particles (Gillman, 1974; Goldberg and Glaubig, 1987; Suarez et al., 1984). WS 900 (no soil) has a pH of 9.7 (1:2 biochar:water, Table S2) and pH increases more than one unit, irrespective of soil texture, when 5% WS 900 is applied to soils, compared to the 1% application rate (Table S7). Therefore, at 5% WS 900, the mutual repulsion among colloids (soil or biochar) is enhanced, resulting in higher sensitivity towards mechanical disturbance and more dust emissions.

4.4. Effectiveness of increasing soil moisture content to reduce dust emission

In this study, dust emissions decreased by at least one order of magnitude when the soil moisture content changed from the low to medium level, irrespective of soil texture or the presence of biochar. Furthermore, for SL treatments, the relatively constant biochar content in dust at different moisture levels implies a proportional reduction in both biochar and soil particle emissions as moisture content increases. Unlike SiL systems, dust emissions are still detectable at high moisture contents for SL systems, implying that maintaining soil moisture above field capacity (the high moisture level used in this study) for SL is necessary to minimize not only soil particle but also biochar particle emissions. Previous studies indicated that threshold values of gravimetric soil water content (GWC), where small increases in soil moisture can cause a distinct reduction of dust emission (Funk et al., 2008; Madden et al., 2010), are highly dependent on soil texture. Coarse-textured soils, in general, emit larger amounts of dust than fine-textured soils because they typically have lower electrostatic surface charges and smaller surface areas, which are undesirable properties for absorbing water (an important binding force between soil particles). Therefore, SL is more susceptible to disruption by tillage than SiL due to its weakly-held aggregates, which explains why a moisture level of almost water saturation is required for SL to achieve undetectable dust emissions no matter whether biochar is applied or not.

4.5. Biochar content in different dust size fractions

Among the total inhalable dust (PM_{100}), a considerable portion (31 to 92% by mass basis) is PM_{10}. This suggests that 8 to 69% of the dust has the potential to remain in the mouth or nose while the remainder can diffuse down the throat and into the lungs. BPCA analysis suggests that the biochar content in PM_{10} fraction was much less than that in the corresponding PM_{100} fraction, which was also much lower than the original biochar application rate in soil. Based on the particle size distribution (Fig. 7), pure biochars, irrespective of type, contain <60% char particles under the diameter of 10 μm on a number basis. Assuming that for the same biochar type, biochar density remains the same for different particle sizes and all particles under 100 μm are spherical, char particles <10 μm consist of only 0.03% to 0.4%, on a mass basis. Therefore, unlike carbon nanomaterials or soots (Kong et al., 2013; Sahu et al., 2014), biochars are much larger with very minimal submicron size particles, explaining the relatively low biochar content detected in PM_{10} samples.

Knowing the biochar content in dust is important for assessing occupational hazards. Inorganic dusts (e.g. soil clay minerals) tend to result in nonallergic reactions in human body while organic dusts (plant or animal sources) can cause allergic diseases (Schenker, 2000). Dust from biochars may be more hazardous than typical organic dusts as they can carry carcinogenic toxins, such as polycyclic aromatic hydrocarbons (PAHs) (Hale et al., 2012; Hilber et al., 2012; Quilliam et al., 2013). In this sense, lower biochar content in smaller size dust is desirable as the smaller sized dust is more difficult to remove naturally.

5. Conclusion

For the first time, this study simulated the mechanical disturbance of biochar-amended soils and quantified the contribution of biochar particles emitted as dusts from two contrasting soil textures. The data from this study suggest that biochar type, especially its feedstock, should be evaluated prior to use in agricultural systems as some of them (e.g. WS 900) enhance soil instability, causing greater potential of dust emissions. Meanwhile, high application rates (e.g. 50 g kg^{-1} used in this study) of such biochar should be avoided, for the dust emissions nonlinearly increase due to the enhanced mutual repulsion among particles. Tillage practices are most likely to be performed when field plots are dry, while our collected data suggest that tilling after wetting the biochar-amended plots effectively reduces exposure to both soil and

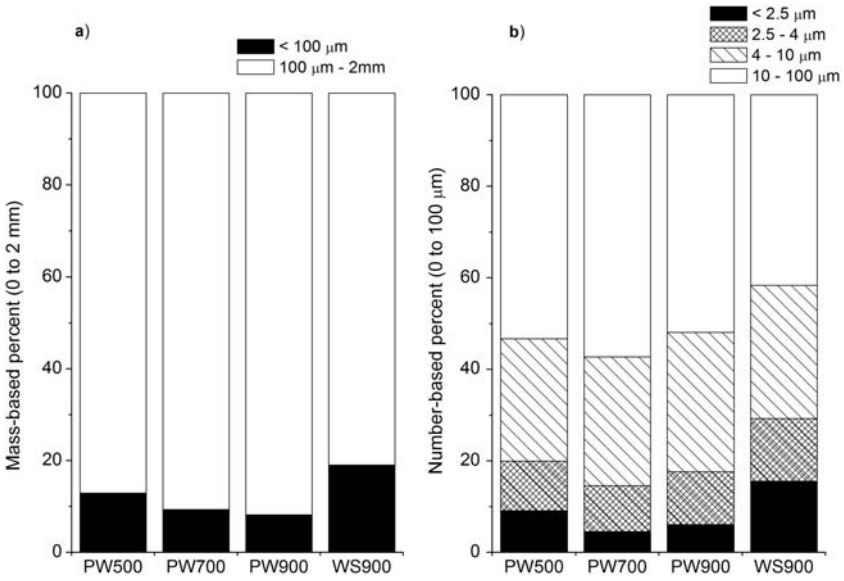

Fig. 7. Particle size distribution of biochars used in this study: a) 100 μm – 2 mm fraction was separated from <100 μm by sieving and expressed on a mass basis; b) < 100 μm fraction was expressed on a number-counting basis using scanning electron microscopy. Abbreviations: WS, walnut shell; PW, pine wood.

biochar particles. However, unlike fine-textured soils, maintaining a high moisture level, close to saturation, is necessary for coarse-textured soils to achieve the greatest dust reduction. It is important to note that in this study, the impact of tillage on biochar dust emissions following land application was investigated and, thus, does not provide data on biochar inclusion in soil aggregates and dust emissions from subsequent tillage events. Our future work is to assess toxicity of different biochars and biochar-rich dust because, from a public health perspective, the amount of toxicants carried by biochar surfaces is equally important as the overall amount and size distribution of dusts generated in a system.

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Appendix A. Supplementary data

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