



Article Impact of Biochar Formulation on the Release of Particulate Matter and on Short-Term Agronomic Performance

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Abstract: When applied in agriculture, the solid carbonaceous residue of anoxic thermochemical conversion of biomass (biochar) has variable effects on soil, crop yields, and climate mitigation. Biochar can be added to soil as powder or as pellets. While powdered forms have demonstrated effects on crop yields, they may release coarse and fine particulate that can be transported into the atmosphere during production, packaging, storage, transport, and distribution. Biochar weathering and wind erosion may also cause the release of particles. Particulate matter (PM) released from biochar may have negative effects on human health and increase the atmospheric burden of shortwave absorbing black carbon aerosols with non-negligible effects on atmospheric radiative forcing. Pelletizing feedstock before the thermochemical conversion and moistening of biochar are expected to reduce the emission of PM in the processing and post-processing phases while also increasing the mean residence time of Carbon in soils. The impact of biochar formulation (pellet and non-pellet) on the release of coarse and fine particulate in wet and dry conditions was assessed in a laboratory experiment. In parallel, the effects of pellet and non-pellet formulations on growth and yield of processing tomato plants were tested in a pot experiment. Results show that pelletization and moistening substantially reduce the amount of fine particles released and are therefore practices that should be adopted to maximize the mitigation potential of biochar. A reduction of tomato yield was observed in pellet treatment, suggesting that the higher interface area of powdered biochar may boost productivity in the short term. This work points to the existence of a tradeoff between the short-term maximization of agronomic benefits and the minimization of harmful effects due to particulate release.

Keywords: biochar formulation; pelletization; yield effect; particulate matter; processing tomato

1. Introduction

The incorporation in agricultural soils of biochar, a carbon-rich material obtained by biomass pyrolysis, is considered a promising strategy to steadily sequester carbon (C) in soils and improve agricultural yields [1–3]. Biochar application to soils has been reported to increase nutrients availability and water holding capacity [4,5], to decrease nutrients leaching [6,7] with generally positive consequences on yields and quality [8–10]. In addition, biochar contributes to a net reduction of greenhouse gases emissions [2,3] by means of soil C sequestration and of a partial offset in the use of fossil fuel based energy. Those benefits have been repeatedly confirmed by proper life cycle assessment

(LCA) [11]. Despite those positive aspects, the release of particulate from biochar is cause for concern because of the potentially harmful effects on health and the implications in terms of reduction of its mitigation potential [12]. Indeed, the production and post-production processes (packaging, storage, transport, and field application) can cause substantial losses of biochar, whose magnitude and fate depends on many factors. A fraction of the smallest biochar particles can be lost by percolation, runoff, and lateral migration or transported by turbulence into the atmosphere [13–15]. In particular, fine and ultra-fine particles may have long residence time in the atmosphere (days to weeks) with potentially negative impact on air quality [14] and on the earth radiative balance [12]. Anthropogenic Black Carbon aerosols (BCa), due to their shortwave absorption properties, are known to have both a direct and indirect climate warming effect [16], and their radiative forcing potential has been estimated to range from +0.16 to +0.80 W m⁻² [17]. The release of particulate matter from biochar can lead to the formation of BCa, potentially reversing the efficacy of biochar for climate change mitigation [12]. The intrinsic dimensional spectrum of biochar particles and the mechanisms of physical deterioration that may take place in the soil after application are therefore important [13]. The formulation of biochar that is applied to soil, i.e., the mesh size, and its susceptibility to fragmentation should be therefore taken into account as far as agronomic and climate mitigation potentials are concerned. So far, the use of powdered or granular biochar has been preferred to the use of biochar produced from pelletized biomass [18], as a small mesh size, a faster fragmentation, and rapid migration are supposed to favor the incorporation of biochar in the soil and with this the interaction with the soil matrix and the amelioration of soil physical properties [19]. On the other hand, the pelletization of biomass prior to pyrolysis is likely to reduce fragmentation and migration of biochar, leading at the same time to a reduced risk of particulate matter (PM) release [2].

Based on these assumptions, the aim of the present work was to investigate the impact of biochar formulation on the release of PM and on the short-term agronomic performances. This was made investigating in a laboratory experiment the relationship between biochar formulation and its release of fine particles. Moreover, the effect of pelletized and granular biochar on growth and yield of processing tomato (*Lycopersicon esculentum* Mill.) were compared in a pot experiment. The implications of the adoption of pelletization practice for biochar production are discussed, focusing the expected impacts for agronomic sustainability and the potential consequences on radiative forcing.

2. Materials and Methods

2.1. Particle Matter Experiment

Six biochar types from different feedstock were used in the laboratory experiment. Three out of six biochar were in form of pellets (PEL), while the remaining had different dimensional spectra (NO PEL). Characteristics of the feedstock, of pyrolysis process, and the chemical characterization of biochar are reported in Table 1. The PEL biochar size was ~2 cm long. The mesh class distribution was assessed by sieving and is reported in Table 2. The analysis of the mesh classes revealed a quite homogeneous distribution for PEL biochar, with the most represented class in the size >2 mm, even if the BP3 showed a different pattern of class distribution. Mesh classes were instead more evenly distributed in NO PEL biochar types.

Determinations of particle matter (PM) were made in the laboratory to assess the coarse (2.5 μ m < PM \leq 10 μ m) and fine (PM \leq 2.5 μ m) particles content of six different biochar types in both dry and wet conditions. Wet conditions (W) were defined as 50% moisture content on a weight basis while dry conditions (D) were obtained by oven-drying wet biochar at 80 °C for 24 h.

In order to quantify the release of fine and coarse particles, 20 g of each biochar type, for both W and D treatments, were placed in a clean airtight glass container connected to an aerosol spectrometer (Grimm Technologies Inc., Douglasville, GA, USA). Each glass container was manually shaken for 1 min, and 2 min after shaking, readings of the spectrometer were acquired. Spectrometric measurements were made in five replicates.

Feedstock	Acr.	Process	Temp (°C)	Formulation	C (%)	N (%)	Bulk Dens. (kg cm $^{-3}$)	pН
Kenaf	BP_1	pyrolysis	550	PEL	86.7	1.9	0.8	9.8
Poplar wood	BP_2	gasification	1200	PEL	90.2	1.5	0.6	7.8
Wheat bran	BP ₃	gasification	1200	PEL *	50.4	1.2	0.5	11.2
Almond husk	BNP ₄	pyrolysis	500	NO PEL	60.3	5.3	0.4	10.1
Maple tree	BNP5	pyrolysis	600	NO PEL	90.1	0.9	0.2	8.3
Wheat straw	BNP ₆	pyrolysis	700	NO PEL	48.6	1.1	0.6	10.8

Table 1. List of six biochar types used for the particulate matter content experiment, describing feedstocks, acronyms, thermochemical conversion process, temperature of the process, formulation, carbon and nitrogen content (%), bulk density (kg cm⁻³), and pH.

* This biochar is the same used in the pot experiment. PEL: pellet; NO PEL: non-pellet.

Table 2. Mesh classes (mm) distribution (%) of the biochar types used for the determination of coarse and fine particles content.

Mesh Classes (mm)	BP ₁ (%)	BP ₂ (%)	BP ₃ (%)	BNP ₄ (%)	BNP ₅ (%)	BNP ₆ (%)
>2	99.3	98.7	74.3	17.1	82.4	64.2
>1	0.4	0.6	10.0	16.0	2.9	14.2
>0.5	0.3	0.4	7.0	18.9	6.2	8.9
>0.25	0.0	0.3	5.3	26.5	3.3	6.8
< 0.25	0.0	0.0	3.4	21.5	5.2	5.9

2.2. Pot Experiment

A pot experiment was performed during the Summer of 2014 at the National Research Council in Sesto Fiorentino (Fi), Italy ($43^{\circ}49'04.12''$ N, $11^{\circ}12'00.42''$ E), using a typical Mediterranean processing tomato cultivar Pietrarossa (Pietrarossa F1). Tomato seedlings were transplanted in 50 cm³ pots at the beginning of May. The trial considered three treatments: control (C_O) without biochar, biochar pellet (PEL), and biochar non-pellet (NO PEL) in a randomized design with five replicates. Chemical characteristics of the biochar feedstock and of biochar are reported on Table 3.

Table 3. Chemical characteristics of the wheat bran feedstock and the biochar of wheat bran applied in the pot experiment.

Parameter	Unit	Wheat Bran Feedstock	Wheat Bran Biochar
pН	-	-	11.2
Ĉ	%	34.6	50.4
Ν	%	2.2	1.2
Ca	$ m g~kg^{-1}$	1.0	3.6
K	$ m g~kg^{-1}$	4.5	14.2
Mg	$ m g~kg^{-1}$	3.9	4.8
Na	$g kg^{-1}$	0.1	0.9
Р	$ m g~kg^{-1}$	9.1	9.8
Cu	${ m mg}{ m kg}^{-1}$	<100	<100
Fe	$mg kg^{-1}$	<120	<120

The biochar used was the BP3, described in Table 1. The choice of BP3 was made considering that this biochar has already been used in previous field experiments aimed at the assessment of its effect on the agronomic performance of processing tomato [5] and on the effect of the pellet formulation on soil hydrological properties [20]. NO PEL biochar was obtained by mechanical fragmentation and sieving of BP3. Particle mesh classes of PEL and NO PEL biochar were obtained by sieving and are reported in Table 4.

Mesh Classes (mm)	PEL (%)	NO PEL (%)
>2	74.3	48.2
>1	10.0	21.5
>0.5	7.0	13.5
>0.25	5.3	9.6
< 0.25	3.4	7.2

Table 4. Mesh classes of the PEL and NO PEL biochar used in the pot experiment.

The characteristics of soil used for the pot experiment were reported in Table 5. Both biochar types were applied at a rate of 22 t ha^{-1} in dry weight and were manually incorporated before tomato transplanting (16 May). The pots were irrigated and fertilized (with 110 N kg ha^{1}). Plant height was determined at the vegetative growth stage, at the appearance of the first flowers and at harvest. The cumulative number of flowers was determined and the total number of fruits was counted. N and C content of leaves were measured at harvest using a CHN elemental analyzer (Perkin Elmer 2400 series II CHNS/O elemental analyzer) together with plant and root dry biomass. The fruits were manually harvested, weighed, measured and visually classified as marketable (not damaged red fruits) and non-marketable (damaged and/or immature fruits). Quality parameters such as fresh fruit pH and the fresh fruit optical residue (°Brix) were also measured. Soil C and N content was measured at tomato harvest (end of experiment, 20 August).

Statistical analysis was performed using one-way and two-way analysis of variance (ANOVA) by the statistical program JMP software (version 10).

Table 5. Characteristics of soil used of the pot experiment. Total organic carbon (C) content was determined using a CHN auto-analyzer (CHN1500, Carlo Erba), Nitrogen (N) content was determined using a CHN auto-analyzer (CHN 1500, Carlo Erba). Cation exchange capacity (CEC) was determined using a NH4OAc method, the pH was measured in a 1:2.5 (mass/vol) soil solution.

Soil Characteristics	
Sand $(g kg^{-1}) 2 mm >> 0.05 mm$	501
Silt (g kg ⁻¹) $0.055 >> 0.02 \text{ mm}$	433
Clay (g kg $^{-1}$) < 0.02	67
Bulk density (Mgm ⁻³)	1.2
C%	2
N%	1.5
CEC (m equiv./100 g)	19
pH	5.5

3. Results

3.1. Particle Matter Experiment

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The distribution of dimensional spectra of coarse and fine particulate released after shaking measured by Grimm aerosol spectrometer for the dry biochar types and for the wet biochar types are shown in Figure 1. The numbers of particles are referred to 1 g in dry weight of each biochar types.

Particles counts were the highest for PM ≤ 2.5 class, and progressively decreased in the 2.5 < PM ≤ 10 range, and for PM > 10. Particles counts were substantially higher in NO PEL biochar in both dry and wet conditions. Wetting the biochar caused a reduction of fine particulate release as high as 93% in PEL and 84% in NO PEL. For each biochar, the sum of mesh classes size <2 mm detected by Grimm aerosol spectrometer (mod.1.109) for the dry biochar were correlated with the release of fine particles, and all NO PEL biochar (BNP4, BNP5, BNP6) showed a good correlation (Figure 2).



Figure 1. Dimensional spectra measured by Grimm aerosol spectrometer (mod.1.109) for the dry biochar (**A**) and for the wet biochar (**B**). Black lines are the average of the Pellet (PEL) biochar types (solid) and the average of the non-pellet (NO PEL) biochar types (dotted), grey lines are the confidence levels at 95%.



Figure 2. Correlation between the size of mesh of three no pelletized biochar BNP4, BNP5, BNP6 and the fine particulate counts detected by Grimm aerosol spectrometer (mod.1.109).

3.2. Pot Experiment

Soil pH, cation exchange capacity (CEC), soil carbon (C_S) and soil nitrogen (N_S) content, and leaves carbon (C_P) and nitrogen (N_P) content measured at tomato harvest are reported in Table 5.

Both biochar treatments (PEL and NO PEL) caused an increase of soil pH, CEC, and total organic C_S compared to the control at the end of experiment (Table 6).

Table 6. Chemical analysis in Control (C_O), PEL and NO PEL treatments in soil (s) and leaves (p). Each value is the average of five measurements; standard error is reported in brackets. Different letters denote differences along treatments by Tukey test (p < 0.05). "ns" indicates no significant differences on the studied variables.

Treatment	pН	CEC meq/100 g	C _S (%)	N _S (%)	C _P (%)	N _P (%)
CO	5.5 a (0.1)	19 a (0.5)	2.1 a (0.2)	1.0 ns (0.1)	32.4 ns (0.5)	2.2 a (0.5)
NO PEL	6.4 b (0.5) 6.4 b (0.4)	21 ab (0.1) 23 b (0.3)	2.7 b (0.4) 2.7 b (0.2)	1.2 ns (0.1) 1.2 ns (0.1)	33.6 ns (1.2) 36.3 ns (2.4)	2.1 a (0.1) 3.7 b (0.1)

Nitrogen soil content did not vary significantly among the treatments while nitrogen content on leaves was significantly higher in NO PEL compared to both PEL and C_O.

Plant height was slightly increased in PEL and NO PEL compared to the C_O (p = 0.069) and no significant difference was observed between PEL and NO PEL treatments (Figure 3).

Root biomass was increased by three months of biochar addition along the sequence NOPEL > PEL > C_O (Figure 4). Moreover, the analysis of fruits at end of experiment also reveal a significant increase of fresh fruit total production (TP^a) and fresh fruit marketable production (MP^b) by biochar treatments (Table 7) for NO PEL compared to the C_O .



Figure 3. Plants height (cm) during the tomato growing season in Control (C_O), pelletized biochar (PEL) and no pelletized biochar (NO PEL). All values are average of five measurements. Bars are standard deviation. The values are not statistically different.



Figure 4. Roots biomass box plot, the value are the average of five replicates. The values are statistically different at p < 0.05. Different letters denote significant difference between treatments.

Table 7. Harvest parameters in the control (C_O), biochar pelletized (PEL) and no pelletized (NO PEL) treatments. Each value is the average of five replicates; standard error is reported in brackets. Different letters denote differences between treatments (p < 0.05). "ns" indicates no significant differences on the studied variables. TP^a = fresh fruit total production, MP^b = fresh fruit marketable production, NMP^c = fresh fruit no marketable production, Fw^d = red fresh fruit weight, Fd^e = red fresh fruit diameter, OR^f = fresh fruit Optical Residue, pH^g = fresh fruit.

Treat.	TP ^a (g plant ⁻¹)	MP ^b (g plant ⁻¹)	NMP ^c (g plant ⁻¹)	Fw ^d (g fruit ⁻¹)	Fd ^e (cm fruit ⁻¹)	OR ^f (°Brix)	pH ^g
Co	460.8 a (22.4)	170.5 a (7.5)	276.3 ns (23.2)	20.3 ns (1.5)	1.4 ns (0.1)	4.2 ns (0.2)	4.5 ns (0.3)
PEL	500.3 a (18.9)	185.5 ab (6.3)	314.8 ns (25.8)	23.2 ns (2.5)	1.5 ns (0.1)	4.2 ns (0.2)	4.1 ns (0.1)
NO PEL	800.5 b (19.3)	470.1 c (8.3)	330.4 ns (32.4)	23.5 ns (1.9)	1.4 ns (0.1)	4.3 ns (0.2)	4.1 ns (0.1)

4. Discussion

The assessment of biochar mitigation potential requires a better understanding of the physical disintegration that unavoidably follows its production and handling and the quantification of the consequential release of fine particulate in the atmosphere. Spokas et al. [13] proved that a substantial amount (on average 10%) of biochar can be lost as dissolved black carbon as a consequence of physical fracturing of original particles, negatively affecting the C-sequestration potential of biochar by increasing the likelihood of its oxidation and C-loss [13]. On the other hand, fragmentation involves the release of fine and ultrafine particulate in atmosphere, something that has the potential to fully reverse its mitigation potential [12]. The release of particulate matter from biochar in the atmosphere was demonstrated to also occur after incorporation into soil in a wind-tunnel experiment [14,15,21]. In particular, the finest particles (\leq 2.5 µm) fall in the category of black carbon aerosol, which is defined as a refractory, water insoluble carbonaceous material strongly absorbing shortwave radiation at wavelength of 550 nm [16]. Atmospheric BCa is known to cause a positive direct Radiative Forcing (RF) by re-emitting the absorbed shortwave radiation as long-wave [16]. The estimation of the effective potential contribution of biochar to the atmospheric burden of BCa, and hence to the global radiative forcing, is a complex matter as it involves multiple causes and mechanisms of BCa release (the way biochar is produced, the incorporation method, its humidity, the soil type, soil cover, etc.). Genesio and coauthors [12] recently calculated that, in the unlikely case of the release of all the BCa contained in biochar in the global application scenario [3], the radiative forcing due to BCa could be of the order of 0.77 to 1.44 W m⁻², thus higher than the mitigation potential of biochar. Of course, as stressed by the same authors, those figures are an overestimation and provocatively point to the need of carefully considering this aspect in order to minimize unintended negative collateral effects of biochar use.

In the laboratory experiment, the pellet formulation of biochar substantially reduced (up to 85%) the release of particulate matter, thus indicating that this could be a practice to limit the climatic impact of particulate matter release. This result holds in the short term, but it is not known if the long-term soil incubation of biochar in pellet prevent subsequent fragmentation of matter that may be transported later into the atmosphere. Wet biochar can also lead in the short term to a reduction (up to ~80%) of release but may also favor a particulate release in this case long-term effect and drying-wetting cycles in soils [13].

A substantial and significant increase in tomato fruit yield in response to the addition of highly fragmented biochar particles (NO PEL) confirmed that, at least in the short term, the mesh size of biochar is of importance for plant growth and production. Indeed, the smaller the mesh, the larger the interface between biochar, soil, and the circulating solution due to the higher surface-volume ratio. This has obvious consequences on the overall soil CEC. Furthermore, the smallest biochar fragments can migrate faster into the soil deep layers [22] with consequences on the local amendment properties. On one hand, such migration may ameliorate the deeper soil horizons, favoring aeration, where the enhanced oxygen availability is known to drive mineralization of organic matter and enhance the availability of mineral nutrients for plants roots [23,24]. On the other side, downward migration might lead to substantial losses of biochar by its percolation [19] and to a reduction of its local amendment properties and to an increased exposition of the carbonized material to oxidation. This is unlikely to occur at the same pace when pellet formulations are used [25].

The results of the pot experiment substantially confirm the interpretation that the dimensional spectrum of the biochar formulation is inversely correlated to soil amelioration properties and to plant yield [25]. Accordingly, the use of powdered biochar is very likely to be the ideal choice to improve yields, as the use of pelletized biochar is unlikely to have similar effect, at least in the short term.

5. Conclusions

The present study revealed that the size of the biochar formulation has additional environmental implications that go beyond the effective and sustainable storage of recalcitrant organic C in the soil and that are related to the release of particulate matter. It highlights a conflict between the agronomic benefits of biochar, which are maximized by powder formulation, and environmental benefits, including climate mitigation and reduced impact for human health, which are maximized in the case of pellet formulation. Pelletization of biochar and moistening practices are already recommended in the guidelines of the International Biochar Initiative (IBI) and the European Biochar Certificate (EBC) [26], but this recommendation should better address the possible tradeoff between environmental and agronomic matters. The assessment of the use of biochar should better integrate the climate mitigation aspects with a full account of its mitigation potential including the impact of BCa release of radiation balance and other albedo issues with soil amelioration (yield enhancement).

In conclusion, the adoption of a biochar-based strategy should not be based only on the agronomic benefits it delivers, but should integrate the entire spectrum of environmental feedbacks, adopting those practices, such as pelletization and moistening of biochar, aimed at the reduction of the release of particulate in the atmosphere. The possibility to resolve the current apparent conflict between those functions depends on a better understanding of the entire spectrum of implications.

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