

Jeroen Zethof (Referee comment posted)

Dear authors,

I read with great interest your work on the use of *solid carbon derived from methane pyrolysis* as soil amendment and its effect on a wide range of soil quality indicators. You presented a large dataset, comparing them with graphite and biochar while giving many insights in the effect of these amendments. Although I encountered a few unclarities, given below, more importantly is the discussion, which is at this point more or less a summation of the results. Especially as you indicate in the introduction that you want to compare these novel materials with the more widely studied biochar and graphite, I had expected to find a more general discussion and conclusion on this point. Maybe it helps to create a two step comparison: (1) Do SC_{cat} and SC_{plas} differ from each other? (2) Does SC's have an advantage/disadvantage over biochar or graphite?

I would also encourage you to include some form of summary statistics, like for example principle components analysis, as it will create a better connection between the two experiments and identify the most important differences between the studied soil amendments. Here below some minor comments, which hopefully encourage you to publish the interesting results you collected.

Response: Thank you for this helpful general comment. We agree that the previous Discussion was too close to a summary of the Results and did not sufficiently compare the novel solid carbon materials with the reference materials. We therefore revised the Discussion to provide a clearer two-step comparison: first, whether SC_{cat} and SC_{plas} differ from each other, and second, whether they show advantages or disadvantages relative to biochar and graphite. We also added a general applicability section to synthesize the main benefits, limitations, and future research needs across soil physical properties, soil biology/ecotoxicology, and heavy metal mobility.

Regarding the suggestion to include additional summary statistics, we considered this carefully. However, the two experiments included different soils, endpoints, extraction methods, and response variables, which would make a combined statistical summary difficult to interpret and potentially misleading. Instead, we strengthened the synthesis in the Discussion to connect the two experiments and identify the most important material-specific differences.

Revision in manuscript: The Discussion was revised to provide a clearer comparison between SC_{cat} and SC_{plas} and their advantages and disadvantages relative to biochar and graphite. A new general applicability section was added to synthesize the results across the two experiments. Additional summary statistics were not included because the two experiments involved different soils, endpoints, and response variables, which would limit their interpretability.

Minor points encountered:

Line 50-51: "The fertility of soils is known to be positively related to carbon content" as you're discussing different carbon forms and inorganic carbon is not known to enhance soil fertility, I suggest to be more specific, i.e. "organic carbon content". Furthermore, your study is more into soil functioning rather than productivity, therefore the term "soil quality" might fit better.

Response: Corrected. We revised the sentence to specify soil organic carbon rather than carbon in general and replaced "soil fertility" with "soil quality and functioning," which better reflects the scope of the study. Based on this change, we also checked and updated the supporting references to ensure that they match the revised statement.

Revision: The sentence was revised to: “Soil quality and functioning are often positively related to soil organic carbon content (Giandon, 2015; Lal, 2004).” The corresponding references were updated accordingly.

Line 69: “Building on the well-documented benefits of biochar,” Do I understand it right you assume the observations of biochar can be directly translated to solid carbon derived from methane pyrolysis? If so, why did you test SC against biochar and graphite if no difference was expected? This can be stated more clearly, otherwise indicate where the materials likely differ.

Response: Thank you for this comment. We revised the introduction to clarify that we do not assume that biochar effects can be directly transferred to methane-derived solid carbon materials. We now explicitly state that SC_{cat} and SC_{plas} may differ from biochar in crystallinity, surface chemistry, porosity, and potential residual organics. We also clarified that biochar and graphite were used as reference amendments to test whether the novel solid carbon materials show similar or distinct effects.

Revision: The introduction was revised to clarify the rationale for comparing SC_{cat} and SC_{plas} with biochar and graphite and to state that material-specific, soil-dependent effects were expected.

“Building on the well-documented benefits of biochar, we do not assume that its effects can be directly transferred to solid carbon derived from methane pyrolysis because the materials differ in crystallinity, surface chemistry, porosity, and (for SC_{plas}) the potential presence of residual organics. We expected biochar to show stronger effects on water retention and heavy metal immobilization because of its higher specific surface area and porosity. In contrast, we expected SC_{cat} to be comparatively inert due to its lower specific surface area and more crystalline structure, resulting in smaller effects on soil hydraulic, biological, and chemical properties. For SC_{plas} , we expected stronger sorption-related effects than for SC_{cat} because of its higher specific surface area, but also potential negative effects on soil organisms and microbial processes due to residual organic compounds. We therefore hypothesize that the two solid carbon materials will exert distinct and potentially soil-dependent effects on (i) soil hydraulic properties (water retention and conductivity), (ii) soil biological and microbiological processes and (iii) the mobility and bioavailability of heavy metals. ”

Line 73-75: Although you nicely state the focal points of your study, you don't give an expected direction of effect. This could have easily been your hypotheses as well. Considering that you test the SCs against biochar and graphite, it is better to state what differences (advantages/disadvantages) you expected to find.

Response: Thank you for this suggestion. We revised the hypothesis paragraph to state the expected direction of effects more clearly. We now explain that biochar was expected to show stronger effects on water retention and heavy metal immobilization, SC_{cat} was expected to behave more inertly, and SC_{plas} was expected to show stronger sorption-related effects than SC_{cat} but also possible negative effects on soil organisms and microbial processes.

Revision: The Introduction was revised to provide more directional hypotheses and clarify the expected differences among biochar, SC_{cat} , and SC_{plas} .

Line 88: “Fe@carbon catalyst,” Is this a typo or brand name?

Response: Regarding the name of the Fe@carbon catalyst, this is a common notation in materials science to indicate a certain structure. In this case, Fe@carbon means that it is a core-shell structure, with Fe in the center and carbon around it.

Line 99: Do you have some data on the C and N content/ratio for the biochar? This would be good for further data interpretation.

Response: Thank you for the clarification. We have this data and have included it in the publication.

Revision in manuscript: This information has been added to Table 1, “Characteristics of the studied solid carbon materials.”

	Solid carbon from catalytic pyrolysis (SC _{cat})	Solid carbon from plasma pyrolysis (SC _{plas})	Biochar	Graphite
Starting material	methane	methane	Straw (<i>Miscanthus x giganteus</i>)	graphite
Structure	crystallized	crystallized	amorphous	crystallized
Specific surface area, (m² g⁻¹)	5.9	58.8	346.4	17.4
Mesoporosity	2< pore diameter <50 nm	n.d.	2< pore diameter <50 nm	2< pore diameter <50 nm
Particles size	< 200 μm	<200 μm	125 and 1000 μm	< 50 μm
Metallic iron	1.7 wt.%	n.d.	n.d.	n.d.
Phenanthrene, (mg kg⁻¹)	0.09 ± 0.02	2.5 ± 0.05	n.d.	0.1 ± 0.02
Pyrene, (mg kg⁻¹)	0.01 ± 0.002	1.7 ± 0.2	n.d.	0.02 ± 0.004
C_{org} (%)	98	100	77.44	
N_{tot} (%)	n.d.	n.d.	0.59	
C/N(%)	n.d.	n.d.	131	

Line 222: “4.4% TOC” -> According Table 3 it is 5.0%, please check.

Response: Thank you for identifying this inconsistency. We checked the value and corrected the text to match the table.

Revision in manuscript: The TOC value was corrected to 5.0%.

Line 615: “, in contrast, had contrasting effects” Maybe change a “contrast” in a synonym or rewrite.

Response: Corrected. We rewrote the sentence to avoid the repetition of “contrast/contrasting” and to improve clarity.

Revision: The sentence was revised to: “ SC_{plas} showed texture-dependent effects on plant-available water.”

Table 2: How was C_{org} and N_t determined? Do you know if there are carbonates in the silty loam soil and if so, how much? Note that you also use N_t in the earthworm experiment, so please change the abbreviation.

Response: Thank you for this comment. We agree that the determination of C_{org} and N_{tot} needs to be described more clearly. We revised the Methods section to clarify that C_{org} and N_{tot} were determined separately from loss on ignition. We also revised the notation in the earthworm avoidance equation to avoid confusion with N_{tot} as total nitrogen.

Revision in manuscript: The Methods section was revised to clarify the determination of C_{org} and N_{tot} and to distinguish it from loss on ignition. The notation in the earthworm avoidance equation was revised to avoid ambiguity.

Table 3: Do you have data on the carbonate content? Would be good to add, if available.

Response: Thank you for this suggestion. Unfortunately, carbonate content was not available for all soils used in the study. To keep the soil-property table consistent across sites, we therefore did not add carbonate content. However, we clarified the available carbon measurements in the Methods section by specifying how total carbon, inorganic carbon, and organic carbon were determined where available.

Revision in manuscript: The Methods section was revised to clarify the determination of total carbon, inorganic carbon, and organic carbon where available. Carbonate content was not added to the soil-property table because comparable data were not available for all soils.