

## **Soil**

**Manuscript No.:** SOIL-2021-145

**Manuscript title:** The cooperative application of oyster shell and biochar efficiently enhanced in-situ remediation of cadmium contaminated soil around intensive industry

**Article type:** Research paper

**Authors:** Bin Wu, Jia Li, Mingping Sheng, He Peng, Dinghua Peng, Heng Xu

### **Reviewer 2:**

General comment:

The manuscript discusses the effects of biochar and oyster shell, added into rice-oilseed rape system, on the remediation of cadmium in contaminated soil and reduction of the element in the crops. This manuscript also evaluated the potential health risk from cadmium in the crops grown in the amended soils. This is quite a good piece of work; however, this paper largely lacks coherence with the objective(s), and needs more attention to its writing style and discussion in support of the results observed. This manuscript mainly emphasizes that the cooperative application of biochar and oyster shell is the most effective means for remediating cadmium in contaminated soil, in terms of reducing cadmium bioavailability in soil and cadmium uptake by plants as well as improving a number of soil quality parameters; however, these are indeed not reflected by the results (data/figures). Rather, the sole application of oyster shell, the cheaper one, appeared to be more effective compared to the joint application of biochar and oyster shell. Major revisions are necessary in many cases (please see the specific comments). The discussion section must be re-written, focusing on well constructive explanations; this part should be coherent and readable. In addition, all the references need to be corrected following the journal style. This manuscript may not be considered for publication; however, for a positive decision, significant improvements, and justification with the results in the highlighted contexts must be done, carefully considering the specific comments through major revisions.

**Response:** *On behalf of all authors, I would like to thank Referee 2 for his/her time, helpful and overall enthusiastic comments, which are very helpful for the improvement*

*of our manuscript. All comments have been taken into account and we attempted to answer all questions. In this study, we investigated that the effects of biochar and oyster shell on Cd uptake by crops and soil biochemical properties. This is an interesting and practical study, which revealed the cooperative application of biochar and oyster shell both reduced the Cd bioavailability, Cd uptake by crops and improved soil biochemical properties including pH, organic matter, available nutrients and enzyme activities. Maybe the cooperative effects were not clearly expressed in our manuscript. According to research results, oyster shell showed a better effect on reducing Cd bioavailability compared to biochar. For Cd immobilization, there was no significant difference between PA2 (15000 kg/ha oyster shell) and PA3 (7500 kg/ha biochar and 7500 kg/ha oyster shell). However, compared to signal biochar treatment, the cooperative application of oyster shell and biochar could significantly increase the Cd immobilization and decrease the remediation cost. Meanwhile, compared to signal oyster shell treatment, the cooperative application of oyster shell and biochar could significantly enhance the soil biochemical properties, such as the organic matter, Olsen-K, and the activities of dehydrogenase, acid phosphate and  $\beta$ -galactosidase. Therefore, the cooperative application of biochar and oyster shell might be a practical way to immobilize Cd and enhance the soil biochemical properties. In our revised manuscript, we will carefully revise the deficiency according to the Referee's comments. The results and discussion will also be carefully revised. In addition, all comments are easy to be revised in our revised manuscript.*

### **Specific comments**

#### **Abstract:**

Line 30-32. Please rephrase the sentence.

**Response:** *Thank you for your advice. This sentence will be revised in our revised manuscript.*

Lines 32-38. Please rephrase this part, with clear treatment combination. It seems that the treatments were applied twice for the two different crops, but this was not the case.

The repetition of “followed with” should be avoided.

**Response:** *Thank you for your kind advice. The treatment combination will be clearly rephrased in our revised manuscript.*

Line 38. Please indicate what the HOAc-extractable Cd means (like, bioavailable or so). Please use a single style for stating “HOAc-extractable Cd”. This is written in a different way at the line numbers 137 (acetic acid), 197, 315..... Also, what is the significance of only mentioning about Cd reduction in R-PA2 in the abstract?

**Response:** *Thank you for your kind advice. We will use a signal style for stating “HOAc-extractable Cd” in our revised manuscript.*

Line 38. ... compared to control.

**Response:** *Thank you for your kind advice. This sentence will be revised in our revised manuscript.*

Line 38-44. What is the significance of these observations – please link.

**Response:** *Thank you for your kind advice. The significance between different treatments will be clearly written in our revised manuscript.*

Line 45. Joint, not jiont. Please check this spelling in other places also.

**Response:** *We are sorry for our carelessness. This incorrect word will be revised in our revised manuscript.*

Line 47. How is the joint application more effective than the sole applications of biochar and oyster shell, while the results don't show any significant differences?

**Response:** *We are sorry for our unclear clarification. According to research results, oyster shell showed a better effect on reducing Cd bioavailability compared to biochar. For Cd immobilization, there was no significant difference between PA2 (15000 kg/ha oyster shell) and PA3 (7500 kg/ha biochar and 7500 kg/ha oyster shell). However, compared to signal biochar treatment, the cooperative application of oyster shell and biochar could significantly increase the Cd immobilization and decrease the*

remediation cost. Meanwhile, compared to signal oyster shell treatment, the cooperative application of oyster shell and biochar could significantly enhance the soil biochemical properties, such as the organic matter, Olsen-K, and the activities of dehydrogenase, acid phosphate and  $\beta$ -galactosidase. Therefore, the cooperative application of biochar and oyster shell might be a practical way to immobilize Cd and enhance the soil biochemical properties.

## **Introduction**

Line 52-53. Please rephrase the sentence, appropriately linking the second part.

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 56. ... has been ranked ....

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 66. In recent years, .....bio-wastes has been widely ....

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 68. Please use either 'and' or 'etc'.

**Response:** *Thank you for your kind advice. This sentence will be revised in our revised manuscript.*

Line 71. Limited or limits?

**Response:** *Thank you for your kind advice. This word will be revised in our revised manuscript.*

Line 72-74. Please rephrase this sentence. What is meant by "...not very significant"?

Please write acidic instead of acid.

**Response:** Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.

Line 76-78. Please rephrase this sentence. ‘that’ to be removed, and ‘which’ to be added between alkaloid and has.

**Response:** Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.

Line 81. There is little known....

**Response:** Thank you for your kind advice. This sentence will be rephrased to better understanding in our revised manuscript.

Line 83. ... are the main food...

**Response:** Thank you for your kind advice. This sentence will be corrected in our revised manuscript.

Line 85-87. Please give references for the previous studies. Please write “rice-oilseed rape”.

**Response:** Thank you for your kind advice. We will introduce more previous studies to highlight our research significance. The “rice-oilseed rape” will be corrected in our revised manuscript.

Line 87. Based on the above...

**Response:** Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.

Line 87. ... a field experiment, not filed.

**Response:** We are sorry for our carelessness. This word will be corrected in our revised manuscript.

Line 92. Alkeline-N or Alkaline-N? Please check this throughout the manuscript.

**Response:** We are sorry for our carelessness. This word will be corrected in our revised

*manuscript.*

Line 93. PCA is generally used for Principal Component Analysis. PCA here should be avoided.

**Response:** *Thank you for your kind advice. We will re-analyze the correlation of different factors in our revised manuscript.*

### **Materials and methods**

Line 103. Table S1 is missing.

**Response:** *We are sorry for our carelessness. This word will be corrected in our revised manuscript.*

Line 115 & 119. “passivators” may be replaced with a more appropriate word. However, the ‘passivators’ are not specified before.

**Response:** *Thank you for your kind advice. This word will be rephrased in our revised manuscript.*

Line 137. Please see the comment made before; compare with the line no. 38....

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 141. Alkeline-N or Alkaline-N? Please check the reference Liu et al. 2017 to confirm its content about the determination of alkaline-N.

**Response:** *Thank you for your kind advice. We will carefully check this reference.*

Line 142. TOC and OM should be indicated in full within (). (Walz et al., 2017) should be written as Walz et al. (2017). Please follow this style for other instances; e.g., in line no. 141.

**Response:** *Thank you for your kind advice. We will carefully check and revise the format of all references in our manuscript.*

Line 155. Human health risk....

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 155. Please use a single format – either ‘Cd’ or ‘cadmium’ throughout the manuscript.

**Response:** *Thank you for your kind advice. We will carefully check the format throughout the manuscript.*

Line 156. The health risks of Cd to adults and children in the crops... - this part of the sentence should be revised.

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 157. Please define Hazard Quotient – what it indicates actually.

**Response:** *Thank you for your kind advice. We will clearly explain the Hazard Quotient.*

Line 158. Please write HQ in full when it is in the beginning of a sentence. Please follow this for other cases throughout the manuscript.

**Response:** *Thank you for your kind advice. The full name of HQ will be explained at the beginning of the sentence in our revised manuscript.*

Line 159. calculated using the following equation:

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 160. (1) can be removed.

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 161. Please write “where,” beforehand.

**Response:** *Thank you for your kind advice. This sentence will be rephrased in our revised manuscript.*

Line 174. What is actually meant by “all statistics”? Should it be graphical interpretation or so...?

**Response:** *Thank you for your kind advice. Data statistical significance was analyzed using SPSS 18.0 package, and means values were considered to be different when  $P < 0.05$  using least significant difference (LSD). Figures were performed using Origin 8.0 (USA).*

Results

Line 175. ‘Results’ only.

**Response:** *We are sorry for our carelessness. This mistake will be correct in our revised manuscript.*

Line 180. OM is generally expressed in %.

**Response:** *Thank you for your kind advice. This expression of OM will be corrected in our revised manuscript.*

Line 182. Please rephrase. What is indicated by ‘reached’ here?

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 197. Please see the comment made before; compare with the line no. 38....

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 197-198. The second part of the sentence should be in the discussion section. Please use reference(s) for such statement.

**Response:** *This result will be adequately discussed according to references.*

Line 200. ... flooding irrigation... Please use one.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our*



*revised manuscript.*

Line 212-213. Not true. Please rephrase with actual observations from the figure.

**Response:** *we are sorry for our carelessness. We will carefully check and revise this sentence according to data.*

Line 213-214. Please rephrase “treatments without amendments”. Please check the Figure 3a to confirm the value 0.88 mg/kg.

**Response:** *We are sorry for our carelessness. We will carefully check and revise this sentence according to data.*

Line 216. ... oyster shell had the ....

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 220. Oilseed Cd content in RT-PA3 was not significantly lower than that in the Control. Please check and amend.

**Response:** *Thank you for your kind advice. We will carefully check and revise this sentence.*

Line 221. “The results indicated that biochar and oyster shell application...” – sole application or combine application?

**Response:** *We are sorry for our unclear statement. This sentence should be “The results indicated that the cooperative application of biochar and oyster shell could efficiently decrease Cd accumulation in brown rice and oilseed”.*

Line 224. Please write HQ in full as in the beginning of the sentence.

**Response:** *Thank you for your kind advice. The full name of HQ will be explained at the beginning of the sentence in our revised manuscript.*

Line 224-225. This sentence can be removed. Rather, ‘Figure 4’ can be written at the end of the sentence in line no. 227.

**Response:** Thank you for your kind advice. This sentence will be removed in the revised manuscript.

Line 226. .... adult female > adult male, which indicated that children were....

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 226. Please use a more appropriate word for 'sensitive'.

**Response:** Thank you for your kind advice. The word will be replaced using a more appropriate word in our revised manuscript.

Line 228. ... HQ values of consuming brown rice for adult male, adult female and children reached 5.46, 6.21 and 6.82, respectively. – What does it mean by “reached”? Is this the max value for a certain group? Moreover, the values given here do not match with the observations in the Figure 4a. Please check and amend clearly.

**Response:** We are sorry for our unclear writing. The values were the average mean. This sentence will be revised to better understand in our revised manuscript.

Line 229. ... HQ values for brown rice intake...

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 229-231. Please rephrase. The decrease in the HQ values for which group(s) – adult male female or children?

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 231-233. Please rephrase. What does the “significantly” mean here?

**Response:** Thank you for your kind advice. The sentence will be rephrased to better understanding in our revised manuscript.

Line 237. It should be Figure 5a, not 2a.

**Response:** *We are sorry for our mistake. This mistake will be corrected in our revised manuscript.*

Line 239. Please avoid using 'points' for indicating the pH values.

**Response:** *Thank you for your kind advice. This word will be replaced in our revised manuscript.*

Line 240. slightly, not slight.

**Response:** *We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.*

Line 241. CEC of soil....

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 242. Figure 5b, not 2b.

**Response:** *We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.*

Line 244-245. This is irrelevant here.

**Response:** *We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.*

Line 247. Table S2 is missing.

**Response:** *We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.*

Line 260-262. Please rephrase.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 262-264. Please rephrase.

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 265-266. Please rephrase.

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 267. .... biochar had no negative ...

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 269-270. Please make it clear whether it was individual application of the treatments or combined application.

**Response:** Thank you for your kind advice. The sentence will be rephrased to better understanding in our revised manuscript.

Line 274. Please rephrase “to experimental data”.

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 275. Person correlation value is expressed by ‘r’. Please check this throughout the manuscript.

**Response:** Thank you for your kind advice. We will carefully check the statement.

Line 278. The Figure 7b....

**Response:** We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.

Line 278. .... Cd uptake by oilseed rape ....

**Response:** Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.

Line 282. Acidic

**Response:** *Thank you for your kind advice. This mistake will be corrected in our revised manuscript.*

Line 287. (detailed information see Supplementary Information) – should be written in an appropriate way. There is no SI attached with the manuscript.

**Response:** *Thank you for your kind advice. We will revise this sentence in an appropriate way.*

Line 288. The dosage of application was 15 t/ha, i.e., 15000 kg/ha. So, please review this amount and also the calculated cost in line numbers 289 & 290.

**Response:** *Thank you for your kind advice. We will re-calculate the cost by the price and amount.*

Line 289-290. joint

**Response:** *Thank you for your kind advice. This mistake will be corrected in our revised manuscript.*

Line 291. Please amend the sentence as ‘... biochar and oyster shell was found to be more effective....’. Again, how this was more effective than the individual applications? The use of oyster shell alone had either similar effects or its effects were not significantly different than the joint application. Moreover, oyster shell is much cheaper than biochar, and thus, compared to the combined use.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript. Other indexes should be also considered. In our revised manuscript, we will explain this sentence more clearly.*

Line 292-294. Should be in the discussion section.

**Response:** *Thank you for your kind advice. The sentence will be replaced to the discussion section.*

## **Discussion**

Line 296. .... are the most ...

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 303-304. Please use a reference for this statement.

**Response:** *Thank you for your kind advice. We will add a reference for this statement.*

Line 305. .... have...

**Response:** *Thank you for your kind advice. This mistake will be corrected in our revised manuscript.*

Line 306. ... had ...

**Response:** *Thank you for your kind advice. This mistake will be corrected in our revised manuscript.*

Line 306. Please give references for the previous studies.

**Response:** *Thank you for your kind advice. We will add references for this statement.*

Line 307. Please rephrase the sentence.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 310. But the reductions in Cd were not statistically significant compared to the control treatment.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 311-314. Please rephrase.

**Response:** *Thank you for your kind advice. The sentence will be rephrased in our revised manuscript.*

Line 315. Please use an appropriate word for “has” here.

**Response:** *Thank you for your kind advice. We will use an appropriate word here.*

Line 315-326. Please rephrase with constructive discussion in the context.

**Response:** *Thank you for your kind advice. This part will be carefully rephrased in our revised manuscript.*

The discussions are mostly linked to the pH; however, the pH for oyster shell and combined treatment are not significantly different, and so the effects due to pH as well. How can this strongly support the joint application over oyster shell alone? The authors can also highlight the benefits of using biochar with oyster shell linking with different other factors.

**Response:** *Thank you for your kind advice. According to research results, oyster shell showed a better effect on reducing Cd bioavailability compared to biochar. For Cd immobilization, there was no significant difference between PA2 (15000 kg/ha oyster shell) and PA3 (7500 kg/ha biochar and 7500 kg/ha oyster shell). However, compared to signal biochar treatment, the cooperative application of oyster shell and biochar could significantly increase the Cd immobilization and decrease the remediation cost. Meanwhile, compared to signal oyster shell treatment, the cooperative application of oyster shell and biochar could significantly enhance the soil biochemical properties, such as the organic matter, Olsen-K, and the activities of dehydrogenase, acid phosphate and  $\beta$ -galactosidase. Therefore, the cooperative application of biochar and oyster shell might be a practical way to immobilize Cd and enhance the soil biochemical properties. In our revised manuscript, we will clearly state the benefits of using biochar with oyster shell with different factors.*

Line 337. Please write 'P' in full in the beginning of the sentence.

**Response:** *Thank you for your kind advice. The full name will be added at the beginning of the sentence.*

Line 345. Please avoid the use of "PCA".

**Response:** *Thank you for your kind advice. We will use another method to analyze the correlation of the different factors.*

Line 350-352. Please rephrase.

**Response:** *Thank you for your kind advice. This sentence will be carefully rephrased in our revised manuscript.*

Line 354. Please use appropriate words for 'obvious' and 'stimulation'.

**Response:** *Thank you for your kind advice. We will use appropriate words to replace to these words.*

Line 354-355. Please either remove this sentence or rephrase.

**Response:** *Thank you for your kind advice. This sentence will be carefully rephrased in our revised manuscript.*

Line 357. Please add year for the reference used, and remove the reference written at the end of the sentence in the line no. 358.

**Response:** *Thank you for your kind advice. We will revise the format of references correctly.*

Conclusions

Lines 372. Please use a more appropriate for 'extraordinary' here?

**Response:** *Thank you for your kind advice. We will use appropriate word to replace this word.*

Line 378. .... our, not Our.

**Response:** *We are sorry for our carelessness. This mistake will be corrected in our revised manuscript.*

**Others**

Figure 7. The values on the figures are not clear.

**Response:** *Thank you for your kind advice. We will revise the figures to be clear.*



## **References**

Please follow the journal style of referencing in the reference section. All the references need to be corrected following the journal style. So, please check and amend throughout the reference section.

**Response:** *Thank you for your kind advice. We will carefully revise the format of references according to the style of journal.*