Dear Editors and Reviewers, thank you very much for your critical comments and great support! We have answered all the comments you suggested, and we hope this revised manuscript can fit with the acceptable standard for Biogeosciences. Please see the attached point-by-point answers with the marked-up manuscript version for your further evaluation.

Sincerely yours,

Zhengqin (on behalf of all authors)

Editor comments:

(Remarks to the Author):

Your manuscript has now been seen by three referees, and all of them find your work of interest. I fully agree. I also appreciate that you have made several steps to address the suggestions and comments by the reviewers. However, upon closer inspection I believe several changes are still required before this manuscript can be accepted for publication.

Thank you very much for your great support and critical comments. Those comments are all valuable and very helpful for revising and improving our paper, as well as important guidance for our further researches. We have made corrections which we hope to meet with approval. Please see the following point-by-point answers.

1. Please have a very close look at your calculations. Table 2 seems to contain several inconsistencies. For instance, averaged across biochar treatments, the average N_2O fluxes show strong differences between soil types, with fairly low SD-values. If this is correct, then how can you explain the relatively small SD-values for the averages for the individual biochar treatments across soil types? After all, these are averages of numbers that show enormous spread. There might a part of the calculation that I am not aware of that can explain these results, but for now it seems that something might have gone wrong here. At the very least these calculations need a better explanation. Possibly calculations need to be redone. I have several other concerns about table 2 as well, please see the attached file for details.

   A: Thank you for your critical and constructive comment. You are right. It's not appropriate to analyze the mean effects of biochar among four different vegetable soils due to the strong differences between soil types. According to your suggestions, we have rechecked and revised our Table 2 to make it concise and clear. We deleted the mean effects of biochar and soil types and revised the related descriptions in the manuscript according to the Two-way analysis as a revised Table 2. According to the Two-way ANOVA analysis, both biochar and soil types and their interactions mostly had significant influences on these main parameters (N_2O, NO, NH_3, GNrE, Vegetable yield and GNrI) in our study. In addition, the multiple comparisons among the treatments and soil types for those main parameters were assessed in the revised Table 3. We hope that will be good to present the main results. Thank you very much for your comments.

2. Although the manuscript is generally easy to follow, it still contains quite a lot of grammatical errors. I made a few corrections in the attached file, but these are by no means complete. It might be an idea to involve a native speaker to check for language.

   A: Thank you! We asked once again the American Journal Expert for help to polish the manuscript.

3. On a minor note: I wonder if you can come up with a more informative title? "The effect of X on Y" is very common, but it doesn't tell potential readers what they want to know, and it doesn't invite them to find out either. Here's a possible suggestion (which you are of course free to ignore): "Biochar reduces yield-scaled emissions of reactive nitrogen gases from

vegetable soils across China"

A: Thanks for your nice suggestion. We have adopted this revised title on Page 1 lines 1-2.

Table 2

Two-way ANOVA for the effects of biochar (Bc) and soil (S) types on cumulative N_2O, NO and NH_3 emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) during the entire sampling period.

Factors	DF	N_2O emission			NO emission			NH_3 emission			GNrE			Vegetable yield			GNrI		
		SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P
Bc	2	271.9	65.1	***	46.4	174.7	***	0.5	0.8	n.s.	380.5	86.4	***	76.2	3.2	n.s.	0.1	7.9	**
S	3	1429.9	228.1	***	152.2	382.1	***	4.1	3.8	*	2322.6	351.5	***	4316.9	123.3	***	2.3	110.3	***
Bc×S	6	179.3	14.3	***	33.4	41.9	***	1.4	0.7	n.s.	234.5	17.7	***	230.4	3.3	*	0.1	1.6	n.s.
Model	11	4009.7	174.5	***	225.3	154.3	***	29.1	7.5	***	5290	218.3	***	15962.0	124.4	***	5.8	77.0	***
Error	24	50.1			3.2			8.5			52.9			280.0			0.2		

SS: the sum of squares.

F value: the ratio of mean squares of two independents samples.

P value: the index of differences between the control group and the experimental group. *, ** and *** indicate significance at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively.

n.s.: not significant.

Table 3

Cumulative gaseous nitrogen (N_2O, NO and NH_3) emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) under the different treatments across the four soils.

Treatments	Acrisol	Anthrosol	Cambisol	Phaeozem
(a) Cumulative N_2O emissions (kg N ha^{-1})				
N	30.59 ±3.15aA	7.83 ±0.60aB	2.52 ±0.37aC	7.10 ±1.91aB
N+Bw	19.45 ±2.43bA	3.20 ±0.28bB	1.97 ±0.21aB	3.45 ±0.86bB
N+Bm	31.56 ±1.35aA	3.63 ±0.62bB	2.26 ±0.58aB	4.01 ±0.68bB
(b) Cumulative NO emissions (kg N ha^{-1})				
N	8.99 ±1.01aA	1.27 ±0.15aB	0.20 ±0.08aC	0.97 ±0.11aBC
N+Bw	4.54 ±0.60bA	0.80 ±0.13bB	0.33 ±0.19aB	0.52 ±0.03bB
N+Bm	3.87 ±0.30bA	1.16 ±0.17aB	0.21 ±0.10aC	0.94 ±0.03aB
(c) Cumulative NH_3 emissions (kg N ha^{-1})				
N	4.72 ±0.27aB	5.79 ±0.54bA	6.34 ±0.51aA	5.67 ±0.42aA
N+Bw	5.09 ±0.38aB	6.83 ±0.74abA	7.35 ±0.75aA	6.24 ±0.49aAB
N+Bm	5.32 ±0.42aB	7.57 ±0.57aA	7.37 ±1.11aA	6.48 ±0.43aAB
(d) GNrE (kg N ha^{-1})				
N	44.30 ±3.13aA	14.89 ±1.33aB	9.06 ±0.80aC	13.74 ±1.67aB
N+Bw	29.08 ±2.21bA	10.82 ±1.14bB	9.64 ±0.88aB	10.21 ±0.92bB
N+Bm	40.76 ±1.66aA	12.36 ±0.74bB	9.84 ±0.49aC	11.42 ±0.27bBC
(e) Vegetable yield (t ha^{-1})				
N	35.20 ±2.52aB	25.29 ±3.90aC	39.09 ±2.03bB	75.65 ±5.84bA
N+Bw	29.05 ±2.35bC	23.57 ±1.74aC	44.53 ±3.74bB	76.95 ±4.04abA
N+Bm	34.93 ±2.87aC	26.30 ±2.63aD	51.00 ±3.18aB	85.89 ±3.29aA
(f) GNrI (kg N t^{-1} yield)				
N	1.27 ±0.18aA	0.59 ±0.08aB	0.23 ±0.02aC	0.18 ±0.04aC
N+Bw	1.01 ±0.12aA	0.46 ±0.05bB	0.22 ±0.04aC	0.13 ±0.02bC
N+Bm	1.17 ±0.15aA	0.47 ±0.04bB	0.19 ±0.01aC	0.13 ±0.01bC

Data shown are means ± standard deviations of the three replicates. See Fig. 1 for treatments codes. Different lowercase letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level. Different capital letters within the same row indicate significant differences among soil types within the same treatment at $p < 0.05$ level.

Specific revisions:

1. Thank you! We have changed the “mitigated” to “mitigating” on Page 3 line 13.
2. Thank you! We have split the lengthy sentence and revised it on Page 3 lines 14-17.
3. We have replaced “Additionally, …application” with “Finally,” on page 4 line 10. Thank you!
4. We have deleted the sentence “Consequently, …systems” on Page 4 line 17. Thank you!

5. We have modified the description that “Therefore, the reduction of reactive N loss is key to meet the joint challenges…” on Page 4 line 28. Thank you!
6. We have revised “would” by “have been suggested to” on Page 5 line 3. Thank you!
7. We have revised “from different soils” by “for various soils” on Page 5 line 6. Thank you!
8. We have substituted “Still” with “Besides” on Page 5 line 6. Thank you!
9. We have revised “system” by “systems” on Page 5 line 14. Thank you!
10. We have revised “intensified” by “intensively cropped” on Page 5 lines 16-17. Thank you!
11. According to your suggestion, we have split long sentence into several shorter and easier sentences on Page 6 lines 4-7. Thanks for your suggestion.
12. We have added verbs for the sentence on Page 6 line 15. Thank you!
13. We have changed “further explained” into “assessed” on Page 9 line 14. Thank you!
14. We have revised the description that Bm amendment increased SOC and TN by 5.8-20.5% and 9.5-14.2% ($p < 0.05$) on Page 10 line 7. Thank you!
15. We have revised the description that Bm increased MBC relative to Bw in all soils to make it clearer on Page 10 line 11. Thank you!
16. We have deleted “remarkable” on Page 10 line 16. Thank you!
17. We have revised the description that “with an average reduction of 45.8%” on Page 11 line 12. Thank you!
18. We have revised the description that “biochar effects differed between soils” on Page 11 line 12. Thank you!
19. Sorry for the inconvenience! We have rechecked and revised the data on Table 2 on Page 25. Thank you!
20. According to the Two-way ANOVA in Table 2 on Page 25, we got that biochar factor (Bc), without the consideration of its species, did not have a significant influence on vegetable yield. We have improved the description on Page 11 line 25. Thank you!
21. We have replaced the “neutrality” with “pH neutral” on Page 13 line 7. Thank you!
22. We have modified the description “probably by stimulating denitrification enzyme activity” on Page 13 line 11. Thank you!
23. We have modified the description “the liming effects of biochar may have prevented the chemical decomposition…” on Page 13 line 12. Thank you!
24. Sorry for the inconvenience. Different from the rest soils, no one significant relation was found between N_2O/NO and PNR or DEA, Which indicated some other processes might occur in Cambisol. We have modified the description on Page 13 line 13. Thank you!
25. Thank you! We have improved the description that compared with Bw, Bm had more the contents of the TN and DOC by 80% and 40%, respectively” on Page 13 line 25. Thank you!
26. We have corrected “Intensive” by “Intensively” on Page 14 line 9. Thank you!
27. Sorry for the inconvenience. We have modified the description to make it clear on Page 15 lines 4-6. Thank you!

28. We have improved the date format in Figs 2 and 3 on Pages 30-31 and added the description of the inserted panels on Page 28 lines 8-9. Thank you!

Reviewer #1 (Remarks to the Author):

The manuscript tries to assess the combined effects of biochar application and soil types on N_2O, NO, NH_3 and crop productivity. The results can provide useful information, however, the language need some final check by a professional and the manuscript also suffers from some major and minor problems.

Major comments:

1. Many results confused me in this paper. i.e. the effect of N_2O mitigation induced by biochar was probably due to the decreased DEA in SX and HLJ (fig.1b), it means the denitrification is the main process for the N_2O production, however, the highest N_2O emission occurred in HN with the lowest DEA (table 3), the result is in contradiction?
2. Line 264, the authors suggested that N_2O nor NO emissions were neither influenced by nitrification nor by denitrification, but by other process. Then what are the other processes? I think it should be more clearly discussed.

A: 1.Thank you for your nice comments! The main reason is that N_2O production and mitigation in different soil type was governed by different processes. It's applicable to SX and HLJ but not HN soil. There were no significant relations between N_2O emissions and DEA in HN soil (Table 4), which indicated denitrification was not the main process for the N_2O production. Many researchers had reported that some other processes such as heterotrophic nitrification (Zhu et al., 2011; Cai et al., 2010), nitrifier denitrification (Zhu et al., 2013) are the main pathways of N_2O emissions especially in the soil with low pH, low carbon content and high mineral N content (Wrage et al., 2001), which greatly match the soil properties of the vegetable soil from HN. Thus, due to the complex potential pathways in HN soil, the lowest DEA activity might influence but not determine the magnitude of N_2O emissions in HN soils.

Cai, Y.J., Ding, W.X., Zhang, X.L., Yu, H.Y., Wang, L.F., 2010. Contribution of heterotrophic nitrification to nitrous oxide production in a long-term N-fertilized arable black soil. Communications in Soil Science and Plant Analysis 41, 2264-2278.

Wrage, N., Velthof, G., Van Beusichem, M., Oenema, O., 2001. Role of nitrifier denitrification in the production of nitrous oxide. Soil Biology and Biochemistry 33, 1723-1732.

Zhu, T., Zhang, J., Cai, Z., 2011. The contribution of nitrogen transformation processes to total N_2O emissions from soils used for intensive vegetable cultivation. Plant and Soil 343, 313-327.

Zhu, X., Burger, M., Doane, T.A., Horwath, W.R., 2013. Ammonia oxidation pathways and nitrifier denitrification are significant sources of N_2O and NO under low oxygen availability. Proceedings of the National Academy of Sciences of the United States of America 110, 6328-6333.

2. Thank you for your comments! The other processes that related to the N_2O or NO emissions might be nitrifier denitrification and heterotrophic nitrification. We discussed more about the other processes that related to the N_2O or NO emissions on Page 13 line 14-18.

Specific comments:

1. The NH_3 volatilization result affected by biochar and soil types is not mentioned in the abstract.

A: Thank you! Biochar amendments stimulated the NH_3 emissions (highest in SX), and Bm resulted in slightly higher NH_3 emissions than Bw did in all types of soils. We added these results on Page 3 line 13-14.

2. Line 19, "Bm improved yield. . .except for HN," but the increment in SX is also not significant.

A: Yes, you are right. Bm improved yield by 13.5–30.5% (except for HN and SX).We have revised it on Page 3 line 13. Thank you!

3. Line 30, According to IPCC 2013, the global warming potential of N_2O is 265 times of CO_2 on a 100-year horizon. Please correct the data. Line 393-394, please modify.

A: Thank you! We have corrected the data 298 by 265 on Page 4 line 6 and modified the corresponding citation on Page 19 line 19-20.

4. Line 111, the experiment was conducted in the greenhouse experimental station, so how to use completely random design?

A: Sorry for the inappropriate descriptions! Before the trial, we labeled all the pots, and then distributed them by casting lots in the experiment region. We have also deleted the world "completely" to make it more appropriate on Page 7 line 9.

5. Line 255-257, could you maybe give some explanation for why a neutrality pH soil will cause mitigation effects of N_2O emission?

A: Thank you for your comments! As reported before, N_2O is produced during several N_2O production pathways and its release to the atmosphere is almost entirely controlled by microbial activities. Among all the pathways, denitrification has been approved to be a main process in upland fertilized soils (Cheng et al., 2015), especially in vegetable field (Qu et al., 2014). As was shown in Fig 1b, Biochar amendments significantly decreased DEA in neutrality pH soils (SX and HLJ), which cause mitigation effects of N_2O emission. However, biochar did not reduce the N_2O emissions in acid and alkaline soil. Soil pHs lower than 5 can adversely affect the activity of nitrous oxide reductase (Liu et al., 2010) and biochar application could not consistently alleviate the adverse effect of such acid pHs. Additionally, nitrification would be the main N_2O production pathways, and biochar amendment stimulated nitrification which could increase the N_2O emissions in alkaline soil (Sánchez-García et al., 2014). Therefore, biochar amendments might have promising mitigation effects through altering the DEA in neutrality pH soils, in which denitrification tends to be the main N_2O pathway.

Cheng, Y., Wang, J., Zhang, J. B., Müller, C., & Wang, S. Q. (2015). Mechanistic insights into the effects of n fertilizer application on N_2O-emission pathways in acidic soil of a tea plantation. Plant and Soil, 389(1), 45-57.

Liu, B., Mørkved, P.T., Frostegård, Å., Bakken, L.R., 2010. Denitrification gene pools, transcription and kinetics of NO, N_2O and N_2 production as affected by soil pH. Fems Microbiology Ecology 72, 407-417.

Sánchezgarcía, M., Roig, A., Sanchezmonedero, M.A., Cayuela, M.L., 2014. Biochar increases soil N_2O emissions produced by nitrification-mediated pathways. Frontiers in Environmental Science 2, 25.

Qu, Z., Wang, J., Almøy, T., & Bakken, L. R. (2014). Excessive use of nitrogen in chinese agriculture results in high $N_2O/(N_2O+N_2O)$ product ratio of denitrification, primarily due to acidification of the soils. Global Change Biology, 20(5), 1685–1698.

6. Line 293-299, please only discuss significant effects. No significant reductions of NH_3 volatilization were found in this study, NH_3 volatilization increased after biochar applied though the effect did not significantly. So I think the discussion of how the biochar reduce NH_3 volatilization is not necessary. And your interpretation of the results includes a lot of over speculations that cannot be logically derived from the results.

A: Yes, you are right! We had deleted some speculations about the mitigation of NH_3 emissions induced by biochar on Page 14 line 17. Thank you!

7. Line 304-310 and Line 311-318, should change place.

A: Thank you! We have exchange lines on Page 14 line 22-30 and Page 15 line 1-7.

8. Line 324-326, this is a lengthy sentence that could be maybe divided into two parts. Please split the sentence between "Additionally. . .vegetable yield".

A: Thank you! We have split the lengthy sentence and revised it on Page 15 line 13-15.

9. Line 326-328, the two sentences are dispensable.

A: Thank you! We have deleted the two sentences on Page 15 line 15.

10. Line 331-332, the conclusions of this study are either flawed. i.e. N_2O and NO in SD show no significant changes among all treatments, and the conclusion cannot be drawn from your results only. Please modify.

A: Yes, sorry for the inconvenience! We have modified the descriptions that biochar amendments mostly reduced N_2O and NO emissions in conclusion on Page 16 line 2.

11. Page 19-22, all the tables should be three-line tables.

A: We have revised all the tables on Page 24-27 and Page 3 in the supplementary material. Thank you!

12. Page 24-27, it is better to use the same y-axis scales in the same figure.

A: We have revised the figures on Page 30-32.

Reviewer #2 (Remarks to the Author):

The study "Effects of two contrasting biochars on gaseous nitrogen emissions and intensity in intensive vegetable soils across mainland China" is a relevant piece of research. It shows N_2O, NO and NH_3 emissions from a greenhouse experiment with 4 vegetable soils during 5 consecutive crops. Apart from the high value of the data itself, the results are interesting and open new research questions that the authors could follow in future works. The differences found in N_2O mitigation in the different soils could be linked to different N_2O formation pathways. Strong points: 1) It analyses several N gases. This is quite unique, since most studies just focus on N_2O emissions. 2) It uses 4 types of soil (with contrasting properties) and it follows gas emissions for a whole year with 5 crop rotations. Weak points: 3) Only 3 replicates are used. This is a bit limited for pot studies. A minimum of 5-6 replicates should be used. Of course using more replicates limits the number of treatments that can be included, but it would give statistically stronger results.

1) The writing could be improved. The language is mostly correct, but the story line is sometimes missing, making it hard to follow. There's a lot of "biochar increases in this treatment and this soil and it decreases in this other soil …" Please summarize and integrate results. This would make the paper much more attractive. It is not necessary to comment on all the results, they are shown in the figures and tables.

2) I do not totally agree with summing up NH_3, N_2O and NO and naming it "gaseous N emissions". This misleads to think that these are all the N gas losses and the fact is that N_2 emissions have not been contemplated in the study and could be substantial.

A:

1. Thanks for your nice comments! However, it is relatively hard to get general results on all the parameters as affected by biochar in all the vegetable soil, which is largely depending on soil and biochar type. Thus, we have tried our best to summarize and integrated the results on Page 10 lines 16-17, Page 11 lines 7 and 12.
2. Yes, you are right. It is true that N_2 emissions could be substantial and were not contemplated in the present study. Therefore, we defined the NH_3, N_2O and NO emissions as GNrE short for

"gaseous reactive N emissions", which would be more accurate and appropriate. In addition, we have changed the "GNE" and "GNI" into "GNrE" and "GNrI", respectively, throughout the manuscript.

Specific comments:

1. The title could be improved. It should state the main results. For instance: "Biochar mostly decreases NO and N_2O emissions but slightly increases NH_3 emissions in intensive vegetable soils across mainland China". Or something similar. What is your main general conclusion? That should be your title.

A: Thank you for your nice comment! Actually, general conclusions could not be drawn easily for all the parameters due to the complexity of soil and biochar properties. Besides, vegetable yield is also another important index we concerned about. Taking into all the parameters (NH_3, N_2O and NO emissions and yield) into account, GNrI would be a better indicator evaluating biochar effects on various soils. Therefore, we revised the title as "Biochar can decrease the yield-scaled emissions of gaseous reactive nitrogen from vegetable soils across China". Thank you so much!

2. The abstract should also be better developed. For instance, it is not mentioned that wheat straw biochar performs better than the manure biochar regarding N_2O mitigation.

A: Yes, thank you! We have added the result that Bw performs better than Bm regarding N_2O mitigation on Page 3 lines 11-12.

3. Line 89. Please also include the amount of biochar added to each pot, not only the Kg/Ha.

A: Thank you! We have added the amount of biochar "282.6 g pot^{-1}" on Page 6 line 12.

4. Line 191: substitute "enhanced" for "increased".

A: Thank you! We have substituted "enhanced" for "increased" on Page 10 line 8.

5. Line 259-260. Please do not link your N_2O results with your DEA results. From Figure 1 we cannot know if biochar is decreasing total denitrified N or decreasing the N_2O/N_2 ratio.

A: Yes, you are right! We have deleted the sentence on Page 13 line 10. Thank you!

6. Line 303: There a spelling mistake (bicohar).

A: We are sorry for the inconvenience. We have corrected it on Page 14 line 21. Thank you!

7. Biochars should be characterized for elemental analysis (Corg, N, H, O). This is important since the atomic ratio H:Corg has been found to be a relevant index for N_2O mitigation.

A: Thank you for your nice recommendation. We have added the elemental analysis data (Corg, N, H, O) on Page 3 in the supplementary material and the corresponding measuring methods on Page 6 lines 22-25 in the materials and method section.

8. The X axis in Figures 2 and 3 must be wrong. They start in 1/15 and they finish in 1/15.

A: We are sorry for the inconvenience. We have corrected Figures 2 and 3 on Pages 30 and 31.

9. Does Figure 1 (DEA) only report N_2O? Why Is N_2 not included?

A: Yes, it does. Based on the method for DEA determination, acetylene (10%, v/v) was added to inhibit N_2O reductase activity (Yoshinari et al., 1977). Therefore, DEA indicated N_2O emissions from the processes " $NO_3^-\rightarrow NO_2^-\rightarrow NO\rightarrow N_2O$" not the final step "$N_2O\rightarrow N_2$". The reported N2O emissions should include the potential N_2 emissions. Thank you!

Yoshinari T, Hynes R, Knowles R. Acetylene inhibition of nitrous oxide reduction and measurement of denitrification and nitrogen fixation in soil[J]. Soil Biology & Biochemistry, 1977, 9(3):177-183.

Reviewer #3 (Remarks to the Author):

This article is well structured, well written and it seems that the experiment was well performed. Results are well described and discussed.

Some other articles on biochar and greenhouse gas emissions have put forward several hypotheses on the effect of biochar on greenhouse gas emissions. Some of them are discussed in the text, but all possible hypotheses could be discussed more systematically in the discussion.

I also would have used soil types instead of site names for the treatments. I think this is more relevant.

A: According to your recommendation, several hypotheses were put forward that: 1) biochar amendment could affect GNrEs, vegetable yield and GNrI in vegetable soils across mainland China, 2) those influences would vary among biochar and soil types. We have added those hypotheses on Page 5 line 17-19. Thank you!

Also, we have renamed the treatments with soil type instead of site names throughout the whole manuscript. Thank you so much for your suggestion!

Some other small remarks

1. Line 26 agriculture accountS

   A: Thank you! We have changed the “accounted” to “accounts” on Page 4 line 2.

2. Lines 78-line 83 please make shorter sentences. It also seems that the words ‘sites’ and ‘soils’ are mixed up.

   A: We have split the long sentence into shorter sentences on Page 6 lines 4-7. Thank you!

3. Line 86: what do you mean with ‘initial’ soil bulk density? Was that the bulk density of the site where soil was collected and does it mean that all treatments had different soil bulk densities?

   A: In order to simulate the biochar effects on different types of soil as much as possible, initial soil bulk density (same to the field condition) was set for the individual soil type. Thank you!

4. Line 90 add ‘were used’

   A: We are sorry for the inconvenience. We have added the “were used” on Page 6 line 15.

5. Line 192. It is mentioned that values were higher for Bm than for Bw amendments but this is only significant for HN; ie the soil with the lowest initial pH

   A: Yes, biochar can increase soil pH at different degree, and this elevation is more significant for acidic soil than alkaline soil (Chintala et al., 2014). We have made some revision about the description on Page 10 line 10. Thank you!

   Chintala R, Schumacher T E, McDonald L M, et al. Phosphorus sorption and availability from biochars and soil/biochar mixtures[J]. Clean–Soil, Air, Water, 2014, 42(5): 626-634.

6. Line 193-194: Bm performed only significantly better in HN, so it did not perform better in all soils.

   A: Yes, you are right! Although the MBC was slightly higher in Bm amendment than that in Bw in all soils, this difference reached significant level only in Phaeozem. We have revised it on Page 10 line 10-11. Thank you!

7. Line 208-209: they greatly lowered some peaks of N_2O emissions: how many occasions, what reduction %, was it significant?

   A: There were six and two times that biochar amendment significantly lowered peaks of N_2O emissions in Anthrosol and Phaeozem by 8.7-74.4% and 23.6-73.6%, respectively. We have already added some description about the reduction on N_2O peaks as affected by biochar on

Page 10 line 26-27. Thank you!

8. Line 243: lowered

   A: We are sorry for the inconvenience. We have corrected it on Page 11 line 28. Thank you!

9. Line 277: how is inorganic nitrogen being immobilized in biochar with higher C/N ratio? What is the presumed mechanism?

   A：Although biochar generally have high carbon contents, labile C fraction exist in biochar simultaneously. The assimilation of labile C by microorganism resulted in microbial demand for inorganic N in soil solution (DeLuca et al., 2006), which lead more N being immobilized. The Bw with higher C/N ratio possessed more labile C (such volatile matter, Bw vs Bm, 23.9% vs 16.3%, data not shown), which make Bw more suitable for N immobilization (Deenik et al., 2010).

   DeLuca T H, MacKenzie M D, Gundale M J, et al. Wildfire-produced charcoal directly influences nitrogen cycling in ponderosa pine forests[J]. Soil Science Society of America Journal, 2006, 70(2): 448-453.

   Deenik J L. Charcoal volatile matter content influences plant growth and soil nitrogen transformations.[J]. Soil Science Society of America Journal, 2010, 74(4):1259-1270.

10. Line 303: biochar is written wrong

    A: We are sorry for the inconvenience. We have corrected the spelling of biochar on Page 14 line 21. Thank you!

11. Line 306-307: how can soil microorganisms lead to unsustainable greenhouse vegetable production?

    A: That is a good question. As long as we know, soil microorganisms are critical to the maintenance of ecosystem service because of their contribution to soil structure; decomposition of organic matter; toxin removal; biogeochemical cycling of carbon, nitrogen, phosphorous, and sulphur; and suppresiveness of plant pathogens (Chaparro et al., 2012; Ferris and Tuomisto, 2015; Larkin, 2015). Additionally, soil salinity has become an essential issue in vegetable ecosystem (Shi et al., 2009; Han et al., 2014), which reduces microbial activity, microbial biomass and changes microbial community structure, and result in the unsustainable for greenhouse vegetable production.

    Chaparro J M, Sheflin A M, Manter D K, et al. Manipulating the soil microbiome to increase soil health and plant fertility[J]. Biology and Fertility of Soils, 2012, 48(5):489-499.

    Ferris H, Tuomisto H. Unearthing the role of biological diversity in soil health[J]. Soil Biology & Biochemistry, 2015, 85:101-109.

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12. Table 2. NH_3-emissoins. BC is not a significant factor but letters are different for the biochar treatments. How is this possible?

    A: Sorry for the inconvenience! We have rechecked and revised the data on Table 2 on Page

25 and modified the corresponding description on Page 11 lines 19-20. Thank you so much!

Thank you very much once again for your helpful comments!
Best Regards!
Zhengqin

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Biochar reduces yield-scaled emissions of reactive nitrogen gases from vegetable soils across China~~Effects of two contrasting biochars on gaseous nitrogen emissions and intensity in intensive vegetable soils across mainland China~~

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Highlights

1. Two contrasting biochars affected G~~aseous ~~Nr~~itrogen ~~I~~ntensity~~ across 4 major vegetable soils in China.
2. Biochar affects gaseous~~-~~ Nr or yield largely depending on soil types.
3. Both biochars decreased GNrI with Bw ~~mitigated ~~mitigating gaseous Nr whereas Bm ~~improved ~~improving yield.

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Abstract

Biochar amendment to soil has been proposed as a strategy for sequestering carbon, mitigating climate change and enhancing crop productivity, but few studies have demonstrated the general effects of different feedstock-derived biochars on the various gaseous reactive nitrogen emissions (GNrEs, N_2O, NO and NH_3) simultaneously across the typical vegetable soils in China. A greenhouse pot experiment with five consecutive vegetable crops was conducted to investigate the effects of two contrasting biochar, namely, wheat straw biochar (Bw) and swine manure biochar (Bm) on GNrEs, vegetable yield and gaseous reactive nitrogen intensity (GNrI) in four typical vegetable soils from ~~the main vegetable production regions~~ ~~(~~Acrisol (Hunan province ~~(AcrisolHN~~), Anthrosol (Shanxi province ~~(AnthrosolSX~~), Cambisol (Shandong province ~~(CambisolSD~~) and Phaeozem (Heilongjiang province ~~(PhaeozemHLJ~~) which~~)~~ ~~that~~ are representative of the intensive vegetable ecosystems across mainland China. Results showed that remarkable GNrE mitigation induced by biochar occurred in Anthrosol~~SX~~ and Phaeozem~~HLJ soils~~, whereas enhancement of yield occurred in Cambisol~~SD~~ and Phaeozem~~HLJ soils~~. Additionally, both biochars decreased GNrI through reducing N_2O and NO emissions by 36.4–59.1 % and 37.0–49.5 % for Bw (except for Cambisol), respectively, while through improving yield by 13.5–30.5 % for Bm (except for Acrisol and Anthrosol), ~~with~~ ~~Bw performed better than Bm regarding N_2O mitigation,~~ ~~with Bw mitigateding N_2O and NO emissions by 21.8–59.1 % and 37.0–49.5 % (except for SD), respectively, while Bm improved yield by 4.013.5–30.5 % (except for HN and SX).~~ Biochar amendments generally stimulated the NH_3 emissions with greater enhancement from ~~Futhermore,~~ Bm ~~performed better~~ than Bw~~regarding N_2O mitigation by 11.8–38.4 % and Bm promoted yield better than Bw by 11.6–20.2%~~. We can infer that ~~Biochar amendments stimulated the NH_3 emissions (highest in AnthrosolSX), and Bm resulted in slightly higher NH_3 emissions than Bw did in all types of soils.~~ ~~Since~~ the biochar's effects on the GNrEs and vegetable yield strongly depend~~ed~~ on the attributes of the soil and biochar. Therefore, in order to achieve the maximum benefits under intensive greenhouse vegetable agriculture, both soil type and biochar characteristics should be seriously considered before conducting large-scale ~~application of~~ biochar applications ~~in order to achieve the maximum benefits under intensive greenhouse vegetable agriculture~~.

Keyword: Biochar, Intensive vegetable soil, Gaseous reactive nitrogen emissions (GNrEs), Gaseous reactive nitrogen intensity (GNrI)

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1 Introduction

Agriculture accounted accounts for an estimated emission of 4.1 (1.7–4.8) Tg N yr^{-1} for nitrous oxide (N_2O) and 3.7 Tg N yr^{-1} for nitric oxide (NO), contributing 60 % and 10 %, respectively, to the total global anthropogenic emissions, largely due to increases of nitrogen (N) fertilizer application in cropland (Ciais, 2013). The concentration of atmospheric N_2O, a powerful, long-lived, greenhouse gas, has increased from 270 parts per billion by volume (ppbv) in the pre-industrial era to ~ 324 ppbv (Ussiri and Lal, 2013); it has 298 265 times the global warming potential (GWP) of carbon dioxide (CO_2) on a 100-year horizon (IPCC, 2013) and also causes depletion of the ozone layer in the atmosphere (Ravishankara et al., 2009). In contrast, NO_x, which is mainly emitted as nitric oxide (NO), does not directly affect the earth's radiative balance but catalyzes the production of tropospheric ozone (O_3), which is a greenhouse gas associated with detrimental effects on human health (Anenberg et al., 2012) and crop production (Avnery et al., 2011). Additionally, along with the high nitrogen (N) applicationFinally, ammonia (NH_3) volatilization is one of the major N loss pathways (Harrison and Webb, 2001) as well, with up to 90% coming from agricultural activities (Misselbrook et al., 2000; Boyer et al., 2002). As a natural component and a dominant atmospheric alkaline gas, NH_3 plays an important role in atmospheric chemistry and ambient aerosol formation (Langridge et al., 2012; Wang et al., 2015b). In addition to nutrient enrichment (eutrophication) of terrestrial and aquatic systems and global acidification of precipitation, NH_3 has also been shown to be a major factor in the formation of atmospheric particulate matter and secondary aerosols (Kim et al., 2006; Pinder et al., 2007), leading to potentially adverse effects on human and ecosystem health such as visibility degradation and threats to biodiversity (Powlson et al., 2008; Behera et al., 2013). Consequently, the release of various reactive N species results in lower N use efficiency in agricultural systems.

In China, vegetable production devotes an area of approximately 24.7 $\times 10^6$ ha, equivalent to 12.4% of the total available cropping area, and the production represented 52 % of the world vegetable production in 2012 (FAO, 2015). Intensified vegetable cultivation in China is characterized by high N application rates, high cropping index and frequent farm practices. Annual Nnitrogen fertilizer inputs for intensively managed vegetable cultivation in rapidly developing areas are 3–6 times higher than in cereal grain cultivation in China (Ju et al., 2006; Diao et al., 2013; Wang et al., 2015a). As a result, great concern exists about excess N fertilizer application, leading to low use efficiency in intensive vegetable fields in China (Deng et al., 2013; Diao et al., 2013; Li et al., 2016). Meanwhile, intensive vegetable agriculture is considered to be an important source of N_2O (Xiong et al., 2006; Jia et al., 2012; Li et al., 2015b; Zhang et al., 2015) and NO production (Mei et al., 2009). Moreover, NH_3 ammonia volatilization is another important N pathway in fertilized soil, resulting in large losses of soil-plant N (Pacholski et al., 2008; Zhang et al., 2011). Therefore, the reduction of reactive N loss becomes a central environmental challengeis the key to meet the joint challenges of high production and acceptable environmental consequences in intensive vegetable production (Zhang et al., 2013).

Biochar is the dark-colored, carbon (C)-rich residue of pyrolysis or gasification of plant biomass under oxygen (O_2)-limited conditions, specifically produced for use as a soil amendment (Sohi, 2012). The amendment of agricultural ecosystems with biochar has been proposed as an effective countermeasure for climate change (Smith, 2016). These additions would have been suggested to increase soil carbon storage (Mukherjee and Zimmerman, 2013; Stavi and Lal, 2013), decrease greenhouse gas GHG emissions (Li et al., 2016), and improve soil fertility and crop production (Major et al., 2010; Liu et al., 2013). However, some recent studies have reported no difference or even an increase in soil N_2O emissions induced by biochar application from differentfor various soils (Saarnio et al., 2013; Wang et al., 2015a). StillBesides, NH_3 volatilization was enhanced by biochar application in pasture soil (Clough et al., 2010), vegetable soil (Sun et al., 2014) and paddy soil in the wheat-growing season (Zhao et al., 2014). Additionally, crop productivity responses to biochar amendments differed among various biochars (Cayuela et al., 2014). These inconsistent results suggest that current biochar application to soil is not a “one-size fit-all paradigm” because of the variation in the physical and chemical characteristics of the different biochars, soil types and crop species (Field et al., 2013; Cayuela et al., 2014). Moreover, limited types of biochar (Spokas and Reicosky, 2009) and soil (Sun et al., 2014) were involved in the experiments in previous studies. Thus, the evaluation of the different types of biochar under the typical soils is imperative to gain a comprehensive understanding of potential interactions before the large-scale application of biochars in intensive vegetable cropping systems in China.

Therefore, a greenhouse pot experiment was conducted in an effort to investigate the effects of different types of biochar on gaseous reactive nitrogen emissions (GNrEs), namely, N_2O, NO and NH_3, simultaneously in four typical intensified intensively cropped vegetable soils across main vegetable production areas of mainland China. We hypothesized that: 1) biochar amendment could affect GNrEs, vegetable yield and yield-scaled gaseous reactive nitrogen emissions, namely, gaseous reactive nitrogen intensity (GNrI) in vegetable soils across mainland China, 2) those influences would vary among biochar and soil types. Overall, the objectives of this research were to gain a comprehensive insight into the effects of two contrasting the different types of biochars on the GNrEs, vegetable yield and gaseous nitrogen intensity (GNrI) in intensively managed vegetable production in China.

2 Materials and methods

2.1. Experimental soil and biochar

Four typical greenhouse vegetable cultivation sites with a long history (more than 10 years) of conventional cultivation were selected from Northeast, Northwest, Central and Eastern China (Fig. S1~~), ~~): 1. ~~namely,~~a Phaeozem from Jiamusi (46°48´N, 130°12´E) in the Heilongjiang province, 2. an Anthrosol from Yangling (34°18´N, 108°2´E) in the Shanxi province, 3. an Acrisol from Changsha (28°32´N, 113°23´E) in the Hunan province, 4. ~~and~~ a Cambisol from Shouguang (36°56´N, 118°38´E) in the Shandong province (FAO and ISRIC, 2012). Those four types of vegetable soil~~from Jiamusi (46°48´N, 130°12´E), Heilongjiang province (HLJ); Yangling (34°18´N, 108°2´E), Shanxi province (SX); Changsha (28°32´N, 113°23´E), Hunan province (HN) and Shouguang (36°56´N, 118°38´E), Shandong province (SD), respectively were collected and~~ represented a range of differences in physicochemical properties and regions (Table S1). Soil samples were manually collected from the cultivated layer (0–20 cm) after the local vegetable harvest in April, 2015. The samples were air-dried and passed through a 5 mm stainless steel mesh sieve and homogenized thoroughly. Any visible roots and organic residues were removed manually before being packed with the necessary amount of soil to achieve the initial field bulk density. Each pot received 15 kg of 105 ℃ dry-weight-equivalent fresh soil. For each of the biochar amendment pot~~s~~, 282.6 g pot^{-1} sieved biochar (2 mm) was mixed with the soil thoroughly before the experiment, which was equivalent to a 40 t ha^{-1} biochar dose (dry weight). No more biochar was added later in the experimental period.

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The ~~Two~~ two types of biochar that were used in this experiment are ~~,~~derived from two common agricultural wastes in China: wheat straw and swine manure, hereafter referred to as Bw and Bm, respectively (Table S1). The Bw was produced at the Sanli New Energy Company in Henan, China, by pyrolysis and thermal decomposition at 400–500 ℃. The Bm was produced through thermal decomposition at 400 ℃ by the State Key Laboratory of Soil Science and Sustainable Agricultural, Institute of Soil Science, Chinese Academy of Sciences. In accordance with Lu (2000), ~~the~~ soil organic carbon (SOC) was measured by wet digestion with H_2SO_4–$K_2Cr_2O_7$, total nitrogen (TN) was determined by semi-micro Kjeldahl digestion, and soil texture was determined with the pipette method. The soil pH and biochar pH were measured in deionized water at a volume ratio of 1:2.5 (soil to water) with a PHS-3C mv/pH detector (Shanghai Kangyi Inc. China). Biochar content of hydrogen (H) was measured by elemental analysis after dry combustion (Euro EA, Hekatech GmbH, Wegberg, Germany). The oxygen content of biochar was measured with the same device after pyrolysis of the sample at 1000 ℃ followed by reduction of the evolved O_2 to CO and quantifi~~ed~~cation by GC-TCD. The soil nitrate (NO_3^-–N) and ammonium (NH_4^+–N) were measured following the two-wavelength ultraviolet spectrometry and indophenol blue method~~s~~, respectively, using an ultraviolet spectrophotometer (HITACHI, UV-2900, Tokyo, Japan).

Electric conductivity (EC) was measured by using a Mettler-Toledo instrument (FE30-K, Shanghai, China) at a 1:5 (w:v) soil to water ratio. Cation exchange capacity (CEC) was determined using the CH_3COONH_4 method. Dissolved organic carbon (DOC) was extracted from 5 g of the biochar/soil with an addition of 50 ml deionized water and measured by a TOC analyzer (TOC-2000/3000, Metash Instruments Co., LTD, Shanghai, China). Ash content was measured by heating the biochars at 750 ℃ for 4 h. The specific surface area of the biochar material was tested using the Brunauer–Emmett–Teller (BET) method, from which the N adsorption–desorption isotherms at 77 K were measured by an automated gas adsorption analyzer ASAP2000 (Micromeritics, Norcross, GA) with + 5% accuracy. Scanning electron microscopy (SEM) imaging analysis was conducted using a HITACHI S-3000N scanning electron microscope.

2.2. Experimental set-up and management

The pot experiments were performed at the greenhouse experimental station of Nanjing Agricultural University, China. Five vegetable crops were grown successively in the four vegetable soils during the experimental period. For each type of soil, three treatments with three replicates were arranged in a ~~completely~~ random design: urea without biochar (N), urea with wheat straw biochar (N+Bw), urea with swine manure biochar (N+Bm). In addition, phosphate and potassium fertilizers in the form of calcium magnesium phosphate and potassium chloride, together with urea, were broadcasted and mixed with soil thoroughly prior to sowing the vegetables. No topdressing events occurred because of the frequent cultivation and short growth period for the leafy vegetables. Based on the vegetable growth, all pots received equal amounts of water and no precipitation. Detailed information on the pot management practices is provided in Table S2.

Each pot consists of a 30 cm × 30 cm (height × diameter) cylinder made of polyvinyl chloride (PVC). The top of each pot was surrounded by a special water-filled trough collar, which allowed a chamber to sit on the pot and prevent gas exchange during the gas-sampling period. Small holes (diameter of 1 cm) at the bottom of the pots were designed for drainage. To prevent soil loss, a fine nylon mesh (< 0.5 mm) was attached to the base of the soil cores before packing.

2.3. Measurement of N_2O, NO and NH_3

The NO and N_2O fluxes were measured simultaneously from each vegetable cultivation using a static opaque chamber method (Zheng et al., 2008; Yao et al., 2009). A square PVC chamber of 35 cm × 35 cm × 40 cm (length × width × height) was temporarily mounted on the pot for gas flux measurement. The chamber was coated with sponge and aluminum foil outside to prevent solar radiation heating the chamber. Gas samples for flux measurements were collected between 8 and 10 a.m. on each measuring day to minimize the influence of diurnal temperature variation. Gas fluxes were usually measured once a week and every other day for one week following fertilizer application. To measure the N_2O flux, four samples were collected from the headspace chamber using 20 ml polypropylene syringes at 0, 10, 20, and

30 min after chamber closure. The gas concentrations in the samples were analyzed within 12 h after sampling using an Agilent 7890A gas chromatograph equipped with an electron capture detector (ECD) for N_2O detection. Argon-methane (5 %) was used as t~~T~~he carrier gas ~~was argon-methane (50 %)~~ at a flow rate of 40 ml min^{-1}. The column and ECD temperatures were maintained at 40 and 300 ℃, respectively. The gas chromatography configurations described by Wang et al. (2013) were adopted for the gas concentration analysis. N_2O flux was calculated using the linear increases in gas concentration with time. Sample sets were rejected unless they yielded a linear regression value of $R^2 > 0.90$.

For each NO flux measurement, gas samples were collected from the same chamber that was used for the N_2O flux measurements (Yao et al., 2009). Before closing the chamber, an approximately 1.0 L gas sample from the headspace of each chamber was extracted into an evacuated sampling bag (Delin Gas Packing Co., LTD, Dalian, China), and this measurement was regarded as time 0 min for NO analysis. After 30 min under chamber enclosure conditions (i.e., after the N_2O sample collections were completed), another headspace gas sample with the same volume was extracted from each chamber into another evacuated bag. Within 1 h after sampling, NO concentrations were analyzed by a model 42*i* chemiluminescence NO–NO–NO_X analyzer (Thermo Environmental Instruments Inc., Franklin, MA, USA). The NO fluxes were derived from the concentration differences between the two collected samples. The NOx analyzer was calibrated by a model 146*i* dynamic dilution calibrator system at the end of each crop-growing season.

The mean flux of N_2O or NO during the experiment period is ~~was calculated as~~ the average of all measured fluxes~~,~~ ~~which were~~ weighted by the interval between ~~the~~ two neighboring measurements (Xiong et al., 2006). The cumulative N_2O was calculated as the product of the mean flux and the entire duration.~~–~~

The NH_3 volatilization was determined using the ventilation method (Zhao et al., 2010). The phosphoglycerol-soaked sponge was replaced every day after each fertilization event for approximately one week. The phosphoglycerol-soaked sponges used to collect the NH_3 samples were immediately extracted with 300 mL potassium chloride (KCl) solution (1 mol L^{-1}) for 1 h. The concentration of ~~ammonia nitrogen (~~NH_4^+–N~~)~~ was measured using the indophenol blue method at 625 nm (Sororzano, 1969) by ultraviolet spectrophotometry (HITACHI, UV-2900, Tokyo, Japan, with 0.005 absorbance of photometric accuracy). The cumulative seasonal NH_3 volatilization was the sum of the daily emissions during the measurement period.

2.4. Auxiliary measurements

Simultaneously with the determination of trace gas fluxes, the air temperature and the soil temperature at a depth of 5 cm were measured using thermally sensitive probes at each sampling date. Soil water content was also measured using a portable water detector (Mode TZS-1K, Zhejiang Top Instrument Corporation Ltd., China) by the frequency domain reflectometer method at a depth of 5 cm. Measured soil water contents (v/v) were converted to water filled pore space

(WFPS) with the following equation:

$$\text{WFPS} = \text{volumetric water content (cm}^3\ \text{cm}^{-3}) / \text{total soil porosity (cm}^3\ \text{cm}^{-3}) \quad (1)$$

Here, total soil porosity = [1 — (soil bulk density (g cm^{-3}) / 2.65)] with an assumed soil particle density of 2.65 (g cm^{-3}). The total soil bulk density was determined with the cutting ring method according to Lu (2000).

After each vegetable crop reached physiological maturity, the fresh vegetable yield was measured by weighing the whole aboveground and belowground biomass in each pot.

$$\text{GNrE} = \text{cumulative } N_2O + \text{cumulative NO} + \text{cumulative } NH_3 \text{ emissions (kg N ha}^{-1}) \quad (2)$$

$$\text{GNrI} = \text{GNrE} / \text{vegetable fresh yield (kg N t}^{-1}\text{ yield)} \quad (3)$$

After the one-year pot experiment, a soil sample from each pot was blended carefully. One subsample was stored at 4 ℃ for determination of microbial biomass carbon (MBC), potential nitrification rate (PNR) and denitrification enzyme activity (DEA) within 3 days. Another subsample was air-dried for analysis of SOC, TN, pH and EC. MBC was determined by substrate-induced respiration using a gas chromatography (Anderson and Domsch 1978). PNR was measured using the chlorate inhibition soil-slurry method as previously described (Kurola et al., 2005) with modifications (Hu et al., 2016). DEA was quantified as described by Smith and Tiedje (1979).

2.5. Data processing and statistics

~~One-way ANOVA was performed to test the effects of the treatments on cumulative N_2O, NO and NH_3 emissions,; GNE,; vegetable yield and GNI.~~ Two-way ANOVA was used to analyze the effects of the biochar type,; soil type,; and their interactions on soil properties, N_2O, NO and NH_3 emissions, vegetable yield, GNrE and GNrI throughout the experimental period. Multiple comparisons among the treatments were ~~further explained~~assessed using Tukey's HSD test. Significant differences were considered at $P < 0.05$. All statistical analyses were performed using JMP ver. 7.0 (SAS Institute, Cary, NC, USA, 2007). Pearson's correlation analysis was used to determine whether there were significant interrelationships between N_2O/NO and PNR or DEA in each soil, using SPSS window version 18.0 (SPSS Inc., Chicago, USA).

3. Results

3.1. Soil responses to biochar amendment

~~Obvious~~ Appreciable differences in all observed soil properties existed among soil types (Table 1~~, $p < 0.001$~~), suggesting the wide variations of soil characters across mainland China. Additionally, biochar amendments had significant influences on all the soil properties (Table 1, $p < 0.05$). Compared with N treatments, biochar amendments increased the SOC, TN and EC by 20.4–135.0 %, 0.5–21.2 % and 2.4–38.1 %, respectively, across all the soils. Compared with Bw, Bm amendment ~~resulted in higher contents of~~increased SOC and TN by 5.8–20.5 % and 9.5–14.2 % ($p < 0.05$), respectively, whereas EC values were higher by 3.3–21.5 % induced by Bw than Bm amendment over all soils. Additionally, biochar amendments significantly increased~~enhanced~~ soil pH by 0.27–0.64 and 0.08–0.10 units compared with N treatment in Acrisol~~HN~~ and Anthrosol~~SX~~ soils ($p < 0.05$), respectively, and ~~higher values were detected with~~ Bm performed better than Bw on increasing soil pH ~~amendment~~ in ~~all soils~~Acrisol. Furthermore, biochar amendments tended to increase MBC in Cambisol~~SD~~ and Phaeozem ~~HLJ soils~~, and Bm ~~performed better in~~increased MBC ~~enhancements than~~relative to Bw in all soils~~all soils~~.

As shown in Fig. 1, no consensus effects on PNR and DEA were observed with biochar amendments across all soils. Compared with N treatment, biochar amendments significantly increased PNR in Phaeozem~~HLJ~~ while exerted no influences on Cambisol~~SD soil~~ (Fig. 1a). Compared with Bw, Bm amendment significantly increased PNR in Acrisol~~HN~~ and Anthrosol~~SX soils~~. Moreover, compared with N, biochar amendments ~~significantly~~ reduced DEA in most soils, significantly in Anthrosol and Phaeozem by an average of 40.1 and 37.8 % ~~in SX and HLJ~~ (Fig. 1b, $p < 0.05$), respectively~~, while producing no influence in SD soils (Fig. 1b)~~. In comparison with Bw, ~~remarkable~~ enhancements in DEA were observed by 42.5 and 74.4 % with Bm amendment in Acrisol~~HN~~ and Anthrosol~~SX soils~~, respectively ($p < 0.05$).

3.2. Seasonal variations of N_2O and NO emissions

The dynamics of N_2O fluxes from all N-applied treatments in the four vegetable soils were relatively consistent and followed a sporadic and pulse-like pattern that was accompanied with fertilization, tillage and irrigation (Fig. 2). In addition, peak N_2O fluxes varied greatly. Most of the N_2O emissions occurred during the Amaranth and Tung choy growing periods, and there were several small emissions peaks during the Spinach and Coriander herb growing periods due to lower N application rate (Table S2), soil temperature and water content (Fig. S2). The highest peaks of N_2O emissions from Acrisol~~HN~~, Anthrosol~~SX~~, Cambisol~~SD~~ and Phaeozem~~HLJ~~ were 4133.7, 1784.0, 432.4 and 1777.2 μg N m^{-2} h^{-1}, respectively. Although biochar (Bw and Bm) application did not significantly alter the seasonal pattern of the N_2O fluxes, they greatly lowered some peaks of N_2O emissions in the Anthrosol~~SX~~ and Phaeozem by 8.7–74.4% and

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23.6–73.6%, respectively (Fig. 2b and d).

Clearly, the NO fluxes demonstrated similar seasonal dynamics to the N_2O fluxes (Fig. 3). Some relatively high peak NO fluxes were still observed in the Spinach and Coriander herb planting seasons even though relatively low temperatures occurred during these periods, primarily due to lower soil moisture which was suitable for NO production. The NO fluxes ranged from -44.6 to 377.6 μg N m^{-2} h^{-1} across all soil types. Furthermore, some NO peaks were significantly weakened with the Bw and Bm in the AcrisolHN soil (Fig. 3a).

3.3. Cumulative N_2O, NO and NH_3 emissions

Cumulative N_2O emissions varied greatly among soil types (Table 23a, p < 0.00105), from 1.97 to 31.56 kg N ha^{-1} across all the soils during the vegetable cultivation period (Table 3a). Biochar amendments had significant influences on the cumulative N_2O emissions, reducing N_2O emissions by 13.7–41.6 % (Table 2, p < 0.001). In comparison with the N treatment, biochar amendment resulted in no consistent effects on N_2O emissions over all soils decreased N_2O emissions by an average of 56.4 % and 47.5 % in SX and HLJ (Table 3a, p < 0.05), respectively, with no remarkable influence in SD soil, indicating significant interactions between biochar and soil types (Table 2, p < 0.001). Additionally,Compared with Bm, Bw amendment decreased N_2O emissions by 11.8–38.4 % across all the soils in relation to Bm, which indicatinged that Bw performed better mitigation effects than Bm which decreased N_2O emissions by 11.8–38.4 % across all the soils, significantly in AcrisolHN soil (Table 3a, p < 0.05). In comparison with N_2O emission, tThe values of cumulative NO emissions was were much smaller than those of N_2O emissions, with a remarkable variation of 0.20–8.99 kg N ha^{-1} across all soils (Table– 3b). Though Biochar amendments had pronounced effects on NO emissions with a reduction by average of 45.8 % (Table 2, p < 0.05001), but biochar their amendments had no consensus effects across soilseffects differed between vegetable soils, reducing NO emissions in HN soil (Table 3b, p < 0.05) and producing no remarkable influence on SD soil, which suggested significant interactions between biochar and soil types (Table 2, p < 0.001). Compared with Bm, Bw amendment significantly reduced NO emissions in AnthrosolSX and PhaeozemHLJ soils (Table 3b, p < 0.05). Moreover,As shown in Table 4, N_2O emissions had positive relationships with DEA both in AnthrosolSX and PhaeozemHLJ soils, and were affected positively withby PNR in AcrisolHN soil (Table 4). Additionally, NO emissions had positive correlations with both PNR and DEA in AnthrosolSX soil. However, neither N_2O nor NO emissions were influenced significantly by PNR and DEA in CambisolSD soils.

As is shown in Table 3c, the cumulative NH_3 emissions fluctuated greatly from 4.72–7.57 kg N ha^{-1}across all the soils. Though significantly enhancing NH_3 emissions (Table 2), Bbiochar amendments produced no significant influences on the NH_3 emissions relative to N treatment in most soils (Table– 3c). A tendency was found for the cumulative NH_3 emissions in N+Bm to be higher than those in the N+Bw treatment, although this difference was not remarkable within

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each soil. Additionally, stimulation effects were consistently present after the first fertilization event in each type of soil (Fig. 4).

3.4. Vegetable yield and gaseous reactive N ~~emissions~~ intensity during the five-vegetable crop rotation

The vegetable yields for the five consecutive vegetable crops are presented in Table 3e. Pronounced differences existed among all soils (Table ~~2~~3e, p < 0.~~001~~05). Additionally, b~~B~~iochar amendments exerted no significant effects on vegetable yield (Table 2). Compared with the N treatment, biochar amendments were prone to increase vegetable yield in Cambisol~~SD~~ and Phaeozem~~HLJ soils~~ against Acrisol~~HN~~ and Anthrosol~~SX soils~~ (Tables 3e), denoting pronounced interactions between soil and biochar (Table 2, p < 0.05). Compared with Bm, Bw amendment lowered total yield over all the soils (Table 3e), significantly in Acrisol~~HN~~ and Cambisol~~SD soils~~ (p < 0.05).

Table 3f presents the GNrI during the whole experiment period, with a pronounced variation among soils (p < 0.05) ~~(Table 2, p < 0.001)~~. The GNrI was greatly affected by biochar amendment during the whole experiment period (Table 2, p < 0.01). Compared to N treatment, biochar amendments reduced the GNrI by 4.3–27.8 % across all soils, significantly in Anthrosol~~SX~~ and Phaeozem~~HLJ soils~~ (Table 3f, p < 0.05). Moreover, there were no remarkable differences between Bw and Bm throughout all soils.

4. Discussion

4.1. Biochar effects on GNrEs across different soil types

The effects of biochar amendment on the N_2O and NO emissions may be positive, negative or neutral, largely depending on the soil condition and the inherent characteristics of the biochar (Spokas and Reicosky, 2009; Nelissen et al., 2014). In our study, effects of two biochars on the N_2O and NO emissions did not follow a consensus trend across the four typical vegetable soils (Table 3a, b). In agreement with Cayuela et al. (2014), who reported that the role of biochar in mitigating N_2O emission was maximal in soils close to ~~pneutrality~~H neutral, remarkable mitigation effects were observed in Anthrosol~~SX~~ and Phaeozem~~HLJ~~ with the biochar amendments (Table 3a). These findings potentially resulted from the effects of the biochars on soil aeration, C/N ratio and pH, which affected the N dynamics and N cycling processes (Zhang et al., 2010; Ameloot et al., 2015). ~~Moreover, mitigation of N_2O emissions induced by biochar was probably due to the decreased denitrification in SX and HLJ soils (Fig.1b and Table 4).~~ In line with Obia et al. (2015), biochar decreased NO emissions in low-pH Acrisol~~HN soil~~ (Table 3b), probably by ~~inducing~~ stimulating denitrification enzyme~~s~~ activity ~~with higher activity~~, and then resulted in less NO accumulation relative to N_2 production. Moreover, the liming effects of biochar may have prevented the chemical decomposition of NO_2^- to NO (Islam et al., 2008), leaving only enzymatically produced NO to accumulate. However, different from the rest soils, neither N_2O nor NO emission was significantly influenced by PNR or DEA, suggesting other processes might play vital roles in Cambisol~~SD soil~~. Besides nitrification and denitrification, nitrifiers denitrification (Wrage et al., 2001) and heterotrophic nitrification (Zhu et al., 2011) can be important processes for producing N_2O/NO as well, especially in vegetable soils with low pH, low carbon content and high N content (Wrage et al., 2001). Ma et al. (2015) speculated that nitrifier denitrification was the main process producing N_2O in the North China Plain (Cambisol~~SD soil~~ within this region). In addition, surplus N input in vegetable systems probably masked the beneficial effects of the biochar addition on the N transformation (Wang et al., 2015a). Therefore, the underlying mechanism of how biochar affect those processes needs to be illustrated in the further research.

On the other hand, different biochars may not produce universal influences on N_2O emissions for the same soil due to the distinct properties of the biochar (Spokas and Reicosky, 2009). In the current study, overall, in comparison with Bm, the Bw amendment had more effective mitigation effects on N_2O and NO emissions (Table 3a, b), largely due to the following reasons. First, compared with Bw, ~~the contents of the TN and DOC in Bm were 1.8- and 1.4-fold~~Bm had more the contents of the TN and DOC by 80% and 40% (Table S1), respectively, which might supply extra N or C source for heterotrophic nitrification in the acidic Acrisol~~HN soil~~, ~~which made~~leading Bm to being ineffective for reducing the N_2O emissions (Table 3a). This result was in accordance with Li et al. (2015a), who observed that biochar amendment had no significant influence on the cumulative N_2O emissions, and even higher N_2O emissions occurred when biochar was input.

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Additionally, as shown in Fig.1, Bm was more prone to stimulate PNR and DEA, thus displaying lower mitigation ability than Bw. Second, compared with Bm, the C/N ratio was approximately twofold in Bw (Table S1), presumably leading to more inorganic nitrogen being immobilized in biochar with a higher C/N ratio (Ameloot et al., 2015), decreasing the available N for microorganisms. Last, as presented in Fig. S3 and Table S1, Bw had more pores and surface area, having a better advantage over Bm in absorbing NO accordingly. Others have found that the lower mitigation capacity of high-N biochars (e.g., manures or biosolids) is probably due to the increased N release in the soil from the biochar (Schouten et al., 2012). To our knowledge, very few studies have investigated biochar effects on NO emissions (Nelissen et al., 2014; Obia et al., 2015), and the mechanisms through which biochar influence NO emissions are not elucidated yet. Therefore, more research is needed to clarify the underlying mechanisms of biochar on NO emission.

Intensively managed soils receiving fertilizer such as urea or anhydrous NH_3 and ruminant urine patches are potential hot spots for NH_3 formation, where the use of biochar is expected to retain NH_3–N in the soil system (Clough and Condron, 2010). Actually, the effects of biochar amendments on NH_3 volatilization largely depend on soil characteristics, biochar types~~ and duration time~~. Soil texture is an important factor impacting NH_3 transfer and release. More clay contents were present in the Anthrosol ~~SX soil~~ (Table S1), which was limited in large soil pores, thus, the addition of porous biochar could enhance the soil aeration, promoting NH_3 volatilization (Sun et al., 2014). Additionally, it was worthy to note that cumulative NH_3 emissions were slightly higher in soils with the Bm than those with the Bw amendment (Fig. 4 and Table 3c) and that difference could presumably be attributed to less surface area and the much higher pH of Bm (Fig. S3 and Table S1), resulting in weak adsorption and great liming effects. ~~Overall, compared with previous studies (Ro et al., 2015; Mandal et al., 2016), no significant reductions were found in cumulative NH_3 volatilizations over the whole observation period when biochar was added to current vegetable soils. In general, freshly produced biochar typically has very low ability to absorb ammonium (Yao et al., 2012). Over time, biochar surfaces are oxidized and increase adsorption (Wang et al., 2016). Moreover, the recorded increase in CEC by Cheng et al. (2006) indicated that biochars that are sufficiently weathered over a period would increase their ability to retain cations such as NH_4^+–N. Further, relatively long-term experiments are required to elucidate the mechanism and duration of effect.~~

4.2. Biochar effects on vegetable yield and GNrI across different soil types

The application of biochar is usually intended to increase crop yields, and evidence suggests this may be successful (Schulz et al., 2013; Li et al., 2016). Due to its liming effect, biochar helps to improve the supply of essential macro- and micronutrients for plant growth (Chan and Xu, 2009; Major et al., 2010). Enhancement of vegetable yield with bi~~e~~ochar amendment occurred in Cambisol~~SD~~ and Phaeozem~~HLJ soils~~ (Table 3e). Additionally, the effects of Bm and Bw on vegetable yield ~~was~~were ~~mixed~~inconsistent, which probably due to~~ performance of biochars as an amendment is related~~

~~to~~ the wide diversity of physicochemical characteristics of biochar that translates into variable reactions in soil (Novak et al., 2014). First, compared to Bw, ~~more DOC content was in the~~ Bm has a higher DOC content (Table S1), through which more nutrients may be directly introduced to the soil (Rajkovich et al., 2012). ~~In addition,~~Secondly, besides their large amount of plant-available nutrients (Hass et al., 2012), ~~manure~~ biochars produced with manure have been generally considered significant for improving soil fertility by promoting soil structure development (Joseph et al., 2010), with the result that Bm was found superior to Bw in vegetable production enhancement in our case (Table 3e). As biochar effects on vegetable yield were variable, both biochar properties and soil conditions and crop species ought to be taken into account comprehensively before applying biochar to a certain soil condition.

However, no promotion of yield was observed with biochar amendments in Acrisol~~HN~~ and Anthrosol~~SX~~. This could be attributed to exacerbated soil salinity, which inhibited the uptake of nutrients and water (Ju et al., 2006; Zhou et al., 2010) and the growth of the soil microorganisms (Setia et al., 2011), leading to unsustainable greenhouse vegetable production. Compared with other biochar (Jia et al., 2012), the higher amounts of ash in Bw and Bm may contain high salts, which ~~causing~~ would result in soil salinity (Hussain et al., 2016). After the addition of the two salt-rich biochars, the EC values of Acrisol~~HN~~ and Anthrosol~~SX~~ vegetable soils increased, which might ~~and~~ reach~~ed~~ the limits to tolerance for the leafy vegetables (Shannon and Grieve, 1998). ~~Additionally, the mixed performance of biochars as an amendment is related to the wide diversity of physicochemical characteristics that translates into variable reactions in soil (Novak et al., 2014). First, compared to Bw, more DOC content was in the Bm (Table S1), through which more nutrients may be directly introduced to the soil (Rajkovich et al., 2012). In addition, besides their large amount of plant-available nutrients (Hass et al., 2012), manure biochars have been generally considered significant for improving soil fertility by promoting soil structure development (Joseph et al., 2010), with the result that Bm was found superior to Bw in vegetable production enhancement (Table 3e). As biochar effects on vegetable yield were variable, both biochar properties and soil conditions and crop species ought to be taken into account comprehensively before applying biochar to a certain soil condition.~~

Here, we assessed two feedstock-derived biochar effects on GNrI in typical cultivated vegetable soils across mainland China. Overall, biochar amendments reduced GNrI over all the soils, with the magnitude largely depending on soil type. Remarkable reduction in GNrI had been detected due to the efficient mitigation induced by biochar in Anthrosol~~SX~~ and Phaeozem~~HLJ~~ (Table 3f). However, despite enhanced vegetable yield, no significant decreases in GNrI were observed in Cambisol~~SD~~, mainly because of the absence of mitigation effects on N_2O, NO and NH_3 emissions of biochars (Table 3a, b and c) ~~Additionally~~Overall, ~~divergent influences on GNE and yield were determined with different biochars that~~ Bw was superior to Bm in mitigating the GNrE while Bm performed better in vegetable yield enhancement

(Table 3d and e). .Therefore, the mitigation efficacyies on GNrI were not notably different between Bw and Bm amendments across the four soils.,largely due to the divergent influences on GNE and yield that Bw was superior to Bm in mitigating the GNE while Bm performed better in vegetable yield (Table 3d and e). Furthermore, from our perspective, economic effectiveness/feasibility, such as the net ecosystem economic budget, should be considered synchronously in intensive vegetable production before large-scale biochar applicat

5. Conclusion

The study demonstrated that biochar amendments mostlygenerally reduced N_2O and NO emissions (except for CambisolSD soil) without influencingand slightly increased the NH_3 emissions, while produced no consensus influences on yield though those effects were largely both biochar- and soil-specific. Additionally, biochar amendments did decrease GNrI in intensive vegetable soils across mainland China. Furthermore, Bw was superior to Bm in mitigating the GNrE whereas the Bm performed better in crop yield throughout all soils. Consequently, both soil type and biochar characteristics need to be seriously considered before large-scale biochar application under certain regions of intensive vegetable production.

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Table legends

Table 1

Soil organic carbon (SOC), soil total nitrogen (TN), soil pH, electric conductivity (EC) and microbial biomass carbon (MBC) as affected by different treatments across the four vegetable soils.

Soil	Treatment	SOC (g kg^{-1})	TN (g kg^{-1})	pH	EC (ds m^{-1})	MBC (mg kg^{-1})
Acrisol~~HN~~	N	8.0 ±0.8c	1.37 ±0.12b	4.37 ±0.04c	1.76 ±0.21b	1353 ±119a
	N+Bw	15.6 ±0.5b	1.47 ±0.07b	4.64 ±0.04b	2.43 ±0.31a	1173 ±49b
	N+Bm	18.8 ±0.6a	1.64 ±0.04a	5.01 ±0.03a	2.00 ±0.32ab	1234 ±50ab
Anthrosol~~SX~~	N	9.7 ±0.7c	1.55 ±0.04b	7.53 ±0.02b	1.74 ±0.27b	490 ±9a
	N+Bw	15.6 ±0.8b	1.62 ±0.06b	7.61 ±0.05a	2.25 ±0.22a	495 ±16a
	N+Bm	17.5 ±1.1a	1.79 ±0.03a	7.63 ±0.01a	1.96 ±0.06ab	504 ±18a
Cambisol~~SD~~	N	7.9 ±0.1b	1.13 ±0.04b	7.70 ±0.08a	0.85 ±0.03b	535 ±13b
	N+Bw	14.2 ±0.6a	1.20 ±0.04b	7.66 ±0.03a	0.92 ±0.04a	554 ±10ab
	N+Bm	15.5 ±1.4a	1.37 ±0.06a	7.71 ±0.03a	0.87 ±0.02ab	573 ±12a
Phaeozem~~HLJ~~	N	29.9 ±0.5b	2.19 ±0.04b	6.91 ±0.05a	0.83 ±0.03b	921 ±44b
	N+Bw	36.0 ±1.5a	2.20 ±0.03b	6.92 ±0.06a	0.95 ±0.03a	988 ±56b
	N+Bm	38.1 ±1.8a	2.41 ±0.01a	6.94 ±0.04a	0.92 ±0.06a	1242 ±196a
ANOVA results						
Biochar		***	***	***	***	*
Soil		***	***	***	***	***
Biochar ×Soil		*	n.s.	***	n.s.	**

Data shown are means ± standard deviations of three replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level.

***Significant at $p < 0.001$; **significant at $p < 0.01$; *significant at $p < 0.05$; n.s. not significant.

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Table 2

Two-way ANOVA and meanfor the effects of biochar (Bc) and soil (S) types on cumulative gaseous nitrogen (N_2O, NO and NH_3) emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) during the entire sampling period.

Factors	DF	N_2O emission			NO emission			NH_3 emission			GNrE			Vegetable yield			GNrI		
		SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P
Bc	2	271.9	65.1	***	46.4	174.7	***	0.5	0.8	n.s.	380.5	86.4	***	76.2	3.2	n.s.	0.1	7.9	**
S	3	1429.9	228.1	***	152.2	382.1	***	4.1	3.8	*	2322.6	351.5	***	4316.9	123.3	***	2.3	110.3	***
Bc×S	6	179.3	14.3	***	33.4	41.9	***	1.4	0.7	n.s.	234.5	17.7	***	230.4	3.3	*	0.1	1.6	n.s.
Model	11	4009.7	174.5	***	225.3	154.3	***	29.1	7.5	***	5290	218.3	***	15962.0	124.4	***	5.8	77.0	***
Error	24	50.1			3.2			8.5			52.9			280.0			0.2		
biochar effect (n = 12)					—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
N mean			12.01±1.44a			2.86±0.24a			5.92±0.24b			20.50±1.60a			43.81±5.82a			0.57±0.05a	
N+Bw mean			7.01±0.58b			1.55±0.14b			6.65±0.27a			14.94±0.84b			43.53±6.31a			0.45±0.04b	
N+Bm mean			10.37±0.56a			1.55±0.10b			7.01±0.25a			18.60±0.65a			49.53±6.91a			0.49±0.03ab	
Soil effect (n = 9)					—	—	—	—	—	—			—	—	—	—	—	—	—
HN mean			27.20±1.85a			5.80±0.50a			5.31±0.16c			38.04±1.90a			33.06±1.65c			1.15±0.11a	
SX mean			4.89±0.45b			1.08±0.13b			12.69±0.46a			12.69±0.46b			25.05±1.11d			0.51±0.01b	
SD mean			2.25±0.26c			0.25±0.09c			9.51±0.55b			9.51±0.55c			44.88±0.49b			0.21±0.01c	
HLJ mean			4.48±0.68b			0.81±0.04b			11.79±0.71a			11.79±0.71b			79.50±2.41a			0.15±0.01c	

SS: the sum of squares.

F value: the ratio of mean squares of two independents samples.

P value: the index of differences between the control group and the experimental group. *, ** and *** indicate significance at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively.

n.s.: not significant.

带格式的：左侧： 2.54 厘米，右侧： 2.54 厘米，顶端： 1.9 厘米，底端： 1.9 厘米，宽度： 29.7 厘米，高度： 21 厘米

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Data shown are means ± standard deviations of the nine replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments at $p < 0.05$ level.

Table 3

Cumulative gaseous nitrogen (N_2O, NO and NH_3) emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) under the different treatments across the four soils.

Treatments	Acrisol~~HN~~	Anthrosol~~SX~~	Cambisol~~SD~~	Phaeozem~~HLJ~~
(a) Cumulative N_2O emissions (kg N ha^{-1})				
N	30.59 ±3.15aA	7.83 ±0.60aB	2.52 ±0.37aC	7.10 ±1.91aB
N+Bw	19.45 ±2.43bA	3.20 ±0.28bB	1.97 ±0.21aB	3.45 ±0.86bB
N+Bm	31.56 ±1.35aA	3.63 ±0.62bB	2.26 ±0.58aB	4.01 ±0.68bB
(b) Cumulative NO emissions (kg N ha^{-1})				
N	8.99 ±1.01aA	1.27 ±0.15aB	0.20 ±0.08aC	0.97 ±0.11aBC
N+Bw	4.54 ±0.60bA	0.80 ±0.13bB	0.33 ±0.19aB	0.52 ±0.03bB
N+Bm	3.87 ±0.30bA	1.16 ±0.17aB	0.21 ±0.10aC	0.94 ±0.03aB
(c) Cumulative NH_3 emissions (kg N ha^{-1})				
N	4.72 ±0.27aB	5.79 ±0.54bA	6.34 ±0.51aA	5.67 ±0.42aA
N+Bw	5.09 ±0.38aB	6.83 ±0.74abA	7.35 ±0.75aA	6.24 ±0.49aAB
N+Bm	5.32 ±0.42aB	7.57 ±0.57aA	7.37 ±1.11aA	6.48 ±0.43aAB
(d) GNrE (kg N ha^{-1})				
N	44.30 ±3.13aA	14.89 ±1.33aB	9.06 ±0.80aC	13.74 ±1.67aB
N+Bw	29.08 ±2.21bA	10.82 ±1.14bB	9.64 ±0.88aB	10.21 ±0.92bB
N+Bm	40.76 ±1.66aA	12.36 ±0.74bB	9.84 ±0.49aC	11.42 ±0.27bBC
(e) Vegetable yield (t ha^{-1})				
N	35.20 ±2.52aB	25.29 ±3.90aC	39.09 ±2.03bB	75.65 ±5.84bA
N+Bw	29.05 ±2.35bC	23.57 ±1.74aC	44.53 ±3.74bB	76.95 ±4.04abA
N+Bm	34.93 ±2.87aC	26.30 ±2.63aD	51.00 ±3.18aB	85.89 ±3.29aA
(f) GNrI (kg N t^{-1} yield)				
N	1.27 ±0.18aA	0.59 ±0.08aB	0.23 ±0.02aC	0.18 ±0.04aC
N+Bw	1.01 ±0.12aA	0.46 ±0.05bB	0.22 ±0.04aC	0.13 ±0.02bC
N+Bm	1.17 ±0.15aA	0.47 ±0.04bB	0.19 ±0.01aC	0.13 ±0.01bC

Data shown are means ± standard deviations of the three replicates. See Fig. 1 for treatments codes. Different lowercase letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level. Different capital letters within the same row indicate significant differences among soil types within the same treatment at $p < 0.05$ level.

Table 4

The correlations between N_2O or NO emission and PNR or DEA in each soil.

Item	Acrisol~~HN~~		Anthrosol~~SX~~		Cambisol~~SD~~		Phaeozem~~HLJ~~	
	PNR	DEA	PNR	DEA	PNR	DEA	PNR	DEA
N_2O	0.75*	0.66	0.49	0.76*	-0.10	0.16	-0.82**	0.70*
NO	0.62	-0.29	0.79*	0.69*	-0.54	0.01	-0.63	0.22

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Asterisks indicated 0.05 level significances (*$p < 0.05$) and 0.01 level significances (**$p < 0.01$), n = 9.

Figure legends

Fig. 1 Potential nitrification rate (PNR) and Denitrification enzyme activity (DEA) under different treatments in Acrisol~~HN~~, Anthrosol~~SX~~, Cambisol~~SD~~ and Phaeozem~~HLJ soils~~. The three treatments with each soil were urea without biochar (N), urea with wheat straw biochar (N+Bw) and urea with swine manure biochar (N+Bm). Bars indicate standard deviation (mean + SD, n = 3). Different letters above the bars indicate significant differences among the different treatments within the same soil, at $p < 0.05$.

Fig. 2 Temporal dynamics of soil N_2O (μg N m^{-2} h^{-1} ± SD, n = 3) fluxes under different treatments in Acrisol~~HN~~ (a), Anthrosol~~SX~~ (b), Cambisol~~SD~~ (c) and Phaeozem~~HLJ~~ (d) ~~vegetable soils~~ with five consecutive vegetable crops. The inserted panels describe the N_2O fluxes during the last two cropping seasons. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

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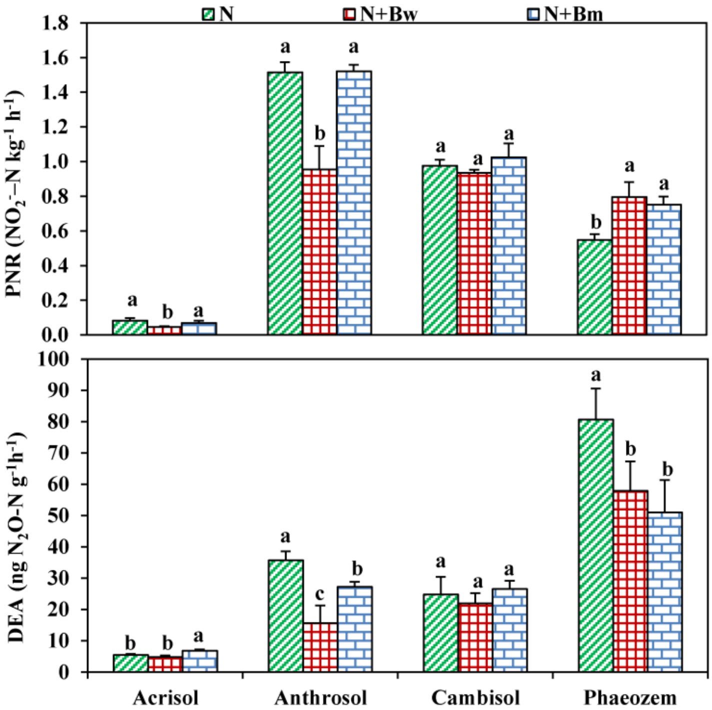
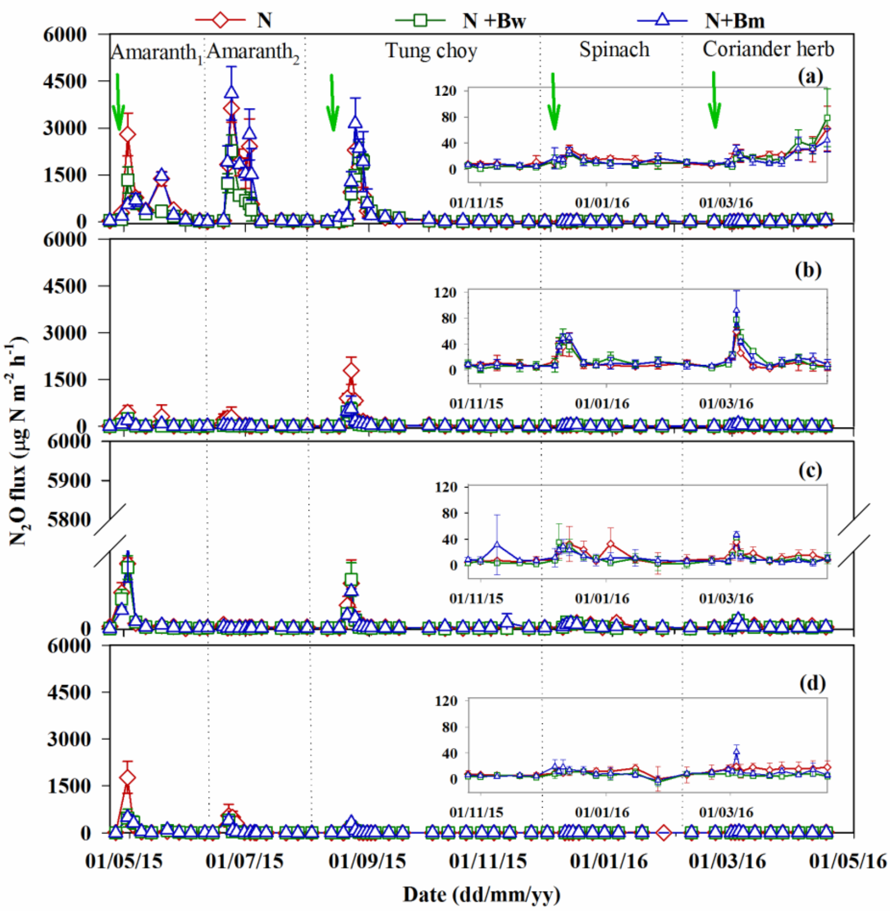
Fig. 3 Temporal dynamics of soil NO (μg N m^{-2} h^{-1} ± SD, n = 3) fluxes under different treatments in Acrisol~~HN~~ (a), Anthrosol~~SX~~ (b), Cambisol~~SD~~ (c) and Phaeozem ~~HLJ~~ (d) ~~vegetable soils~~ with five consecutive vegetable crops. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

Fig. 4 Cumulative ammonia (NH_3) emissions from the Acrisol~~HN~~ (a), Anthrosol~~SX~~ (b), Cambisol~~SD~~ (c) and Phaeozem~~HLJ~~ (d) ~~soils~~ during the four nitrogen fertilization events F: every N fertilization event. The bars indicate the standard deviation of the mean (kg N ha^{-1} ± SD, n = 3) of each treatment for the sum of the four N fertilization events. See Fig. 1 for treatments codes. Different letters above the bars indicate significant differences among the different treatments for each soil, at $p <$ 0.05.

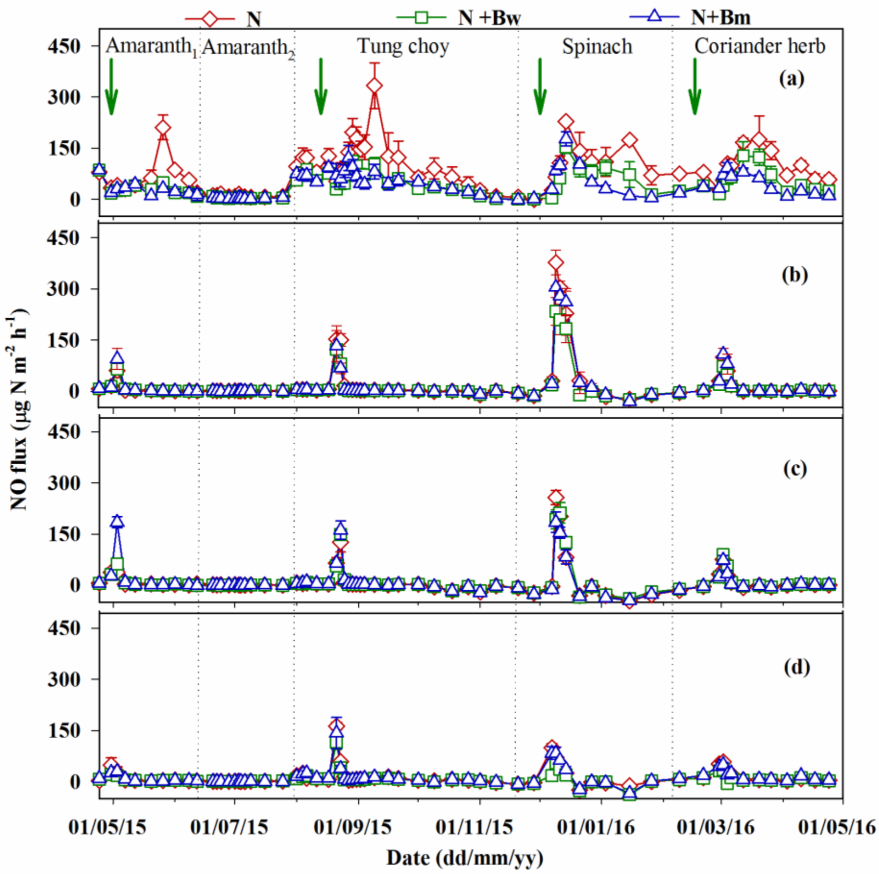

N
N+Bw
N+Bm
PNR (NO2−-N kg-1 h-1)
DEA (ng N2O-N g-1 h-1)
Acrisol
Anthrosol
Cambisol
Phaeozem

1

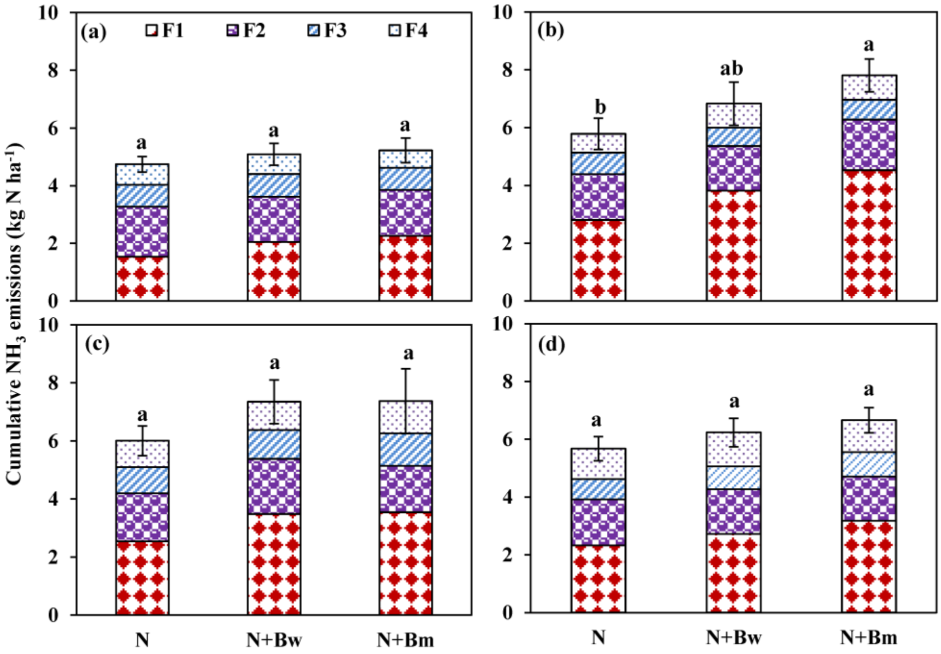

N
N +Bw
N+Bm
Amaranth1
Amaranth2
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
01/11/15
01/01/16
01/03/16
N2O flux (μg N m-2 h-1)
01/05/15
01/07/15
01/09/15
01/11/15
01/01/16
01/03/16
01/05/16
Date (dd/mm/yy)

1

1



N
N +Bw
N+Bm
Amaranth1
Amaranth2
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
450
300
150
0
NO flux (μg N m-2 h-1)
01/05/15
01/07/15
01/09/15
01/11/15
01/01/16
01/03/16
01/05/16
Date (dd/mm/yy)



Cumulative NH_3 emissions (kg N ha^{-1})
(a)
F1
F2
F3
F4
(b)
(c)
(d)
N
N+Bw
N+Bm
a
ab
b

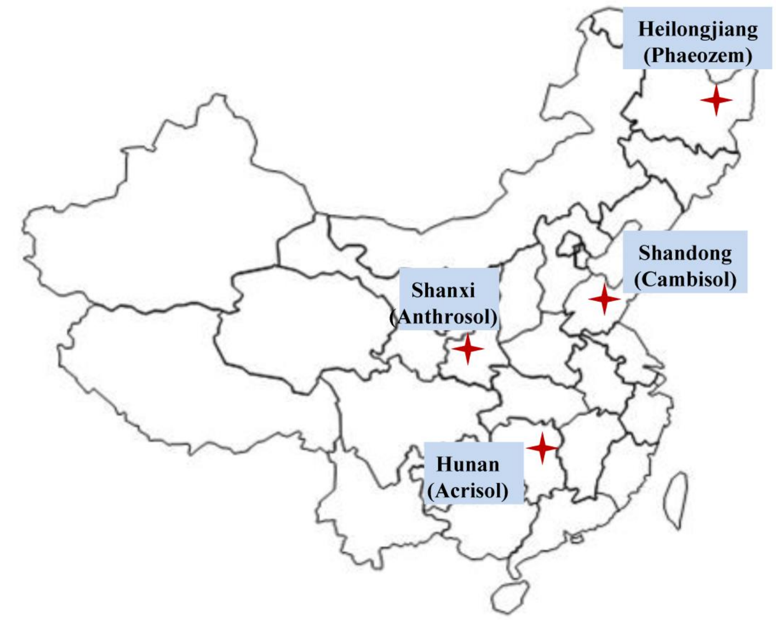
1

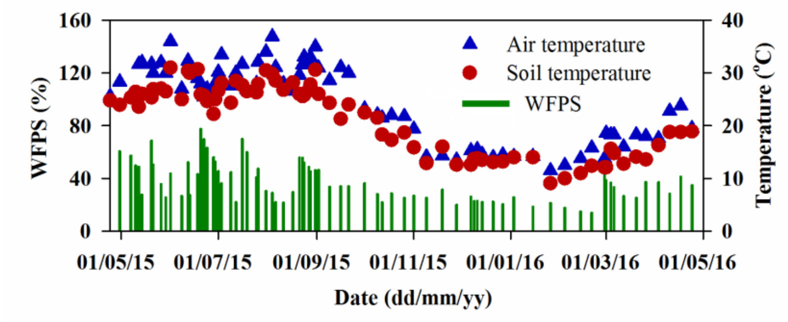
Supplementary information

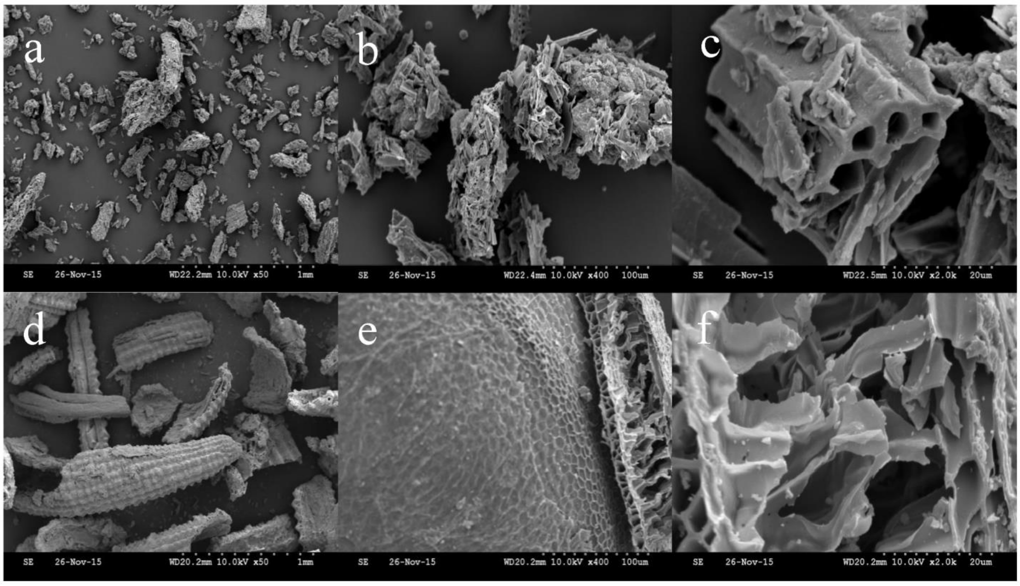
Fig. S1 Map showing the sampling sites in China.

Fig. S2 Dynamics of water filled pore space (WFPS), air temperature and soil temperature during the vegetable cultivation period.

Fig. S3 Scanning electron microscope (SEM) images of the biochars derived from Bw (a, b and c) and Bm (d, e and f). Same magnification for a and d (× 50), b and e (× 400) and c and f (× 2000).






a
b
c
d
e
f
SE 26-Nov-15 WD22.2mm 10.0kV x50 1mm
SE 26-Nov-15 WD22.4mm 10.0kV x400 100um
SE 26-Nov-15 WD22.5mm 10.0kV x2.0k 20um
SE 26-Nov-15 WD20.2mm 10.0kV x50 1mm
SE 26-Nov-15 WD20.2mm 10.0kV x400 100um
SE 26-Nov-15 WD20.2mm 10.0kV x2.0k 20um

1

Table S1

Characteristics of the vegetable soils and biochars used in the experiment.

Iterm	Vegetable soil				Biochar	
	HN	SX	SD	HLJ	Bw	Bm
Texture	sandy loam	silt (sandy) clay loam	silt (sandy) loam	silt (sandy) loam		
sand, %	47.1	17.7	24.7	31.6		
silt, %	40.0	59.6	60.4	52.8		
clay, %	12.9	22.7	14.9	15.6		
total C (g kg^{-1})	7.6	9.8	8.2	26.8	449.1	461.2.
total N (g kg^{-1})	1.2	1.4	1.0	2.1	6.5	12.0
C/N	6.3	7.0	8.2	12.8	69.1	38.4
H (g kg^{-1})					10.5	16.1
O (g kg^{-1})					52.4	96.7
H/Corg					0.3	0.4
pH	5.6	7.6	8.2	7.6	9.7	10.0
EC (ds m^{-1})	1.8	1.1	0.2	0.2	10.6	3.3
DOC (g kg^{-1})	0.5	0.4	0.2	0.7	0.9	1.3
CEC, cmol kg^{-1}	6.1	13.2	15.3	20.3	22.1	22.7
WHC, %	41.6	50.1	54.4	59.6	362.0	304.1
NH_4^+–N (mg kg^{-1})	105.3	32.2	28.4	31.6	4.3	4.0
NO_3^-–N (mg kg^{-1})	415.8	307.6	21.2	30.8	6.1	3.2
Bulk density (g cm^{-3})	1.2	1.4	1.1	1.1		
Surface area (m^2 g^{-1})					21.3	9.3
Ash content, %					29.1	38.6

EC: electronic conductivity; DOC: dissolved organic carbon; CEC: cation exchange capacity; WHC: water holding capacity

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Table S2

Crop rotation, tillage practices, and fertilizer application from April 2015 to April 2016.

Crop	Date	Agricultural activity	Fertilizer N rate (kg N ha^{-1})	Fertilizer P rate (kg N ha^{-1})	Fertilizer K rate (kg N ha^{-1})
$Amaranth_1$	04/22/2015	Tillage			
	04/29/2015	Fertilizer application and planting	240	240	240
	06/13/2015	Harvesting			
	06/14/2015	Tillage			
$Amaranth_2$	06/19/2015	Fertilizer application and planting	0	0	0
	07/31/2015	Harvesting			
	08/01/2015	Tillage			
Tung choy	08/20/2015	Fertilizer application and planting	200	200	200
	11/27/2015	Harvesting			
	11/28/2015	Tillage			
Spinach	12/06/2015	Fertilizer application and planting	150	150	150
	01/28/2016	Harvesting			
	01/09/2016	Tillage			
Coriander herb	02/28/2016	Fertilizer application and planting	180	180	180
	04/29/2016	Harvesting			
	04/30/2016	Tillage			