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<b>Submisi awal</b>	Studies on the performance of bentonite and its composite as phosphate adsorbent and fertilizer
<b>Judul setelah revisi</b>	Studies on the performance of bentonite and its composite as phosphate adsorbent and phosphate supplementation for plant

## Submission Confirmation for HAZMAT-D-20-03525R1

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From: Journal of Hazardous Materials (em@editorialmanager.com)

To: sheila\_p5@yahoo.com

Date: Friday, May 15, 2020 at 12:36 PM GMT+7

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Ms. Ref. No.: HAZMAT-D-20-03525R1

Title: Studies on the performance of bentonite and its composite as phosphate adsorbent and fertilizer  
Journal of Hazardous Materials

Dear Dr. Santoso,  
Research Paper

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## Decision Notification on HAZMAT-D-20-03525

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From: Yong Sik Ok (em@editorialmanager.com)

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CC: [dan.tsang@polyu.edu.hk](mailto:dan.tsang@polyu.edu.hk)

Ms. Ref. No.: HAZMAT-D-20-03525

Title: Synthesis of dual-purpose chitosan-rarasaponin-bentonite composite for phosphate adsorbent and soil fertilizer  
Journal of Hazardous Materials

Dear Dr. Santoso,

The review of your manuscript, Synthesis of dual-purpose chitosan-rarasaponin-bentonite composite for phosphate adsorbent and soil fertilizer, submitted to Journal of Hazardous Materials has been completed. The Reviewers recommend the need for major revisions.

Please revise and re-submit your manuscript according to the Reviewers' and Editor's comments appended below, and/or linked to your submission on the JHM website. To submit a revision, please go to <https://ees.elsevier.com/hazmat/> and login as an Author. On your Main Menu page is a folder entitled "Submissions Needing Revision" which displays your submission record. For guidelines on how to submit your revised manuscript please go the following address: [http://help.elsevier.com/app/answers/detail/p/7923/a\\_id/91](http://help.elsevier.com/app/answers/detail/p/7923/a_id/91)

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\* This should be a separate file labeled "Response to Reviewers" that carefully addresses, point-by-point, the issues raised in the comments appended below. Include each Reviewer's comments followed by your responses, and indicate where you have changed the manuscript in response to each comment, by providing the page and line number of the change in the manuscript. This will help me to compare the Reviewer comments with both the replies and the changes made in the manuscript.

\* Indicate the changes that you have made in response to Reviewers' comments in colour in the body of the revised manuscript.

\* Provide clear reasons in any cases where you have chosen not to make changes suggested by the Reviewers.

Revised Submission:

\* Ensure that the manuscript conforms to the format of the Journal and that all articles in your references include titles. Double check the author names provided in the submission; if your manuscript is accepted, any authorship change will involve approval from co-authors and the Editor handling the submission and may cause a significant delay in publishing your manuscript.

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Please submit your revised manuscript and Response to Reviewers according to the above instructions before May 18 2020 11:59:59:000PM. When I receive it, the manuscript will be sent to Reviewers and I will contact you soon after receiving their comments.

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Thank you for your interest in the Journal of Hazardous Materials. I look forward to receiving your resubmitted manuscript.

Yours sincerely,  
Daniel CW Tsang, PhD  
Editor  
Journal of Hazardous Materials

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Comments from Editors and Reviewers:

Reviewer #1: General comments:

This work dealt with the synthesis of modified bentonite and their application on phosphate adsorption and phosphorus-containing fertilizer. Organo-bentonite and acid-activated bentonite were produced and conventional characterization techniques, batch trial and seedling growth bioassay experiments were performed to evaluate their properties. The experiments were designed properly and reasonably, however, no novelty in relation to the mechanism, synthesis methods and results was found. Similar work could be seen from (Bernardo et al., 2018; Yao et al., 2013). In that case, I don't suggest the paper should be published. The questions and comments are given here:

1. Please emphasize the novelty in the introduction.
2. The word number of each highlight should be less than 85 (including blanks).
3. The material adsorption capacity is not better than most reported materials.
4. The plant experiment should include the treatment of conventional phosphorus fertilizer.
5. Line 88: Organo-bentonite (OrB) composite was prepared by bentonite in this part. However, the abstract mentioned that organo-bentonite was prepared by modifying the acid-activated bentonite (AAB).
6. Line 92: What are the concentration and volume the hot acetic acid solution?
7. What are the pH of adsorption isotherm and adsorption kinetics? Please add the contents of adsorbent doses and time interval.
8. Line 145: Please add the explain of Ce.

9. The models of the instruments should be added. State the software used to conduct the data analysis and statistical analyses.
10. The acid-activated bentonite can increase growth of the plants. However, the organo-bentonite shows unfavorable effects in plants grown. Thus, the chitosan-rarasaponin-bentonite composite is not used as soil fertilizer. Please adjust the title.
11. Fig. 4 and Fig. 5 should be combined, like Fig. 1, which may be more intuitive for comparative analysis
12. The font, size and position in the table can be adjusted and beautified;
13. The authors should mention the advantages of bentonite and bentonite-based materials in this field. Moreover, struvite is also widely used and studied in this field, what's the priorities of bentonite compare with struvite.
14. The mechanisms of adsorption and desorption need to be further explained.
15. The authors should describe how many seeds per treatment in the Arabidopsis thaliana growth experiment. According to Fig. 7, there are only two Arabidopsis plants in each treatment, which is not enough.

Bernardo, M.P., Guimarães, G.G.F., Majaron, V.F., Ribeiro, C. 2018. Controlled Release of Phosphate from Layered Double Hydroxide Structures: Dynamics in Soil and Application as Smart Fertilizer. *ACS Sustainable Chemistry & Engineering*, 6(4), 5152-5161.

Yao, Y., Gao, B., Chen, J., Yang, L. 2013. Engineered biochar reclaiming phosphate from aqueous solutions: mechanisms and potential application as a slow-release fertilizer. *Environ Sci Technol*, 47(15), 8700-8.

Reviewer #2: This paper is focused on synthesis of dual-purpose chitosan-rarasaponin-bentonite composite for phosphate adsorbent and soil fertilizer. Although there are very few points that are not clear and contradictory, but the overall innovation is enough to basically meet all the requirements of the adsorption experiment, the structure of the paper is complete. And I recommend that this paper can be published after major revision, and the questions are listed as follows:

1. From the perspective of the full paper, the title and the summary chart show inconsistencies and contradictions with the content of the paper. For example, although OrB can absorb phosphorus, it can also inhibit the growth of plants. How can it be used as soil fertilizer?
2. (L28 and L173) Pay attention to the difference between "... Pseudo-first-order..." and "... Pseudo-second order...".
3. Introduction:  
L34-38, the statement about the functions and importance of phosphorus should be also cited "Assembling biochar with various layered double hydroxides for enhancement of phosphate recovery, *Journal of Hazardous Materials*, 365(2019)665-673.; Efficient Phosphorus Recycling and Heavy Metal Removal from Wastewater Sludge by a Novel Hydrothermal Humification-Technique, *Chemical Engineering Journal*, 394(2020)124832."
- L52, "In physical treatment (i.e., sorption), removal of Pi involves the use of adsorbent and adsorptive media with potential sorption capacity" Please refer to related paper: Optimizing the synthesis of Fe/Al (hydr)oxides-biochars to maximize phosphorus removal via response surface model. *J. Clean. Prod.*, 2019, 237, 117770
4. Please explain why hot acetic acid is used to dissolve chitosan.
5. (L90) Reference seems to be needed for the time and power used in microwave ovens.
6. (L110) What reagent should be used to adjust the pH value?
7. (L117) Only one data of "... (1-50 mg) ..." is presented in this paper, and the adsorption dose of other models is 20 mg. Please explain why not used the other mass of adsorbent samples.
8. Please indicate the equipment, model and conductivity of deionized water.
9. (L123) The specific value of "...a certain time interval..." should be specified.
10. (L211-212) Obvious narrative error. Please avoid the occurrence of similar mistakes. The error is as follows: "It is observed that AAB has smaller pores than AAB."
11. In response to this phenomenon (L225-227), you can try to explore the ratio of functional groups and electrostatic adsorption on the surface of composite materials to phosphorus adsorption.
12. Adsorption isotherm and adsorption kinetics should be progressive rather than parallel. For relevant research, please refer to the following: High-dispersion zero-valent iron particles stabilized by artificial humic acid for lead ions removal, *Journal of Hazardous Materials*, 383(2020)121170.
13. Please add error bars to figures for adsorption isotherm and adsorption kinetics.
14. The conclusion is not accurate, and the adsorption mechanism is not only electrostatic adsorption.
15. In graphical abstract, inconsistent with the mechanism of action explained in the article.

Reviewer #3: This paper reported an economical dual-purpose organo-bentonite composite which act as a Pi-adsorbent and soil nutrient enrichment. It is very interesting to use modified bentonite as adsorbent and also fertilizer. However, there are still some problems with this paper. I suggest for major revision before publication:

1. In page 5, section 2.1: Please give the chemical composition and basic physical properties (phase composition, fineness etc) of unmodified bentonite, which is very important for next bentonite modification study.
2. In page 5, line 80: Please describe in detail the subsequent treatment process of acid-activated bentonite, whether it needs to be dried or pulverized as OrB.
3. In page 12, line 211-212: 'It is observed that AAB has smaller pores than AAB' is a wrong description, please correct it.
4. In page 18, line 314-315: The author said 'the value of  $q_S$  is very far different from  $q_e(\text{exp})$ ', however the value of  $q_S$  in Table 3 is very close to  $q_e(\text{exp})$ . Please explain this contradiction.
5. In page 20 and 22, line 335 and 357: '4.4 Adsorption kinetics' and '4.5 Root growth in Pi media' should be '3.4 Adsorption kinetics' and '3.5 Root growth in Pi media'.
6. The author emphasized that chitosan-rarasaponin-bentonite (OrB) was a dual-purpose material, for phosphate adsorbent and soil fertilizer. However, the results showed that OrB caused severe growth retardation in root grown. Therefore, OrB could not be considered as a fertilizer. Please amend the title and corresponding description of OrB as a fertilizer in whole paper.

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Ms. Ref. No.: HAZMAT-D-20-03525R2

Title: Studies on the performance of bentonite and its composite as phosphate adsorbent and phosphate supplementation for plant  
Journal of Hazardous Materials

Dear Dr. Santoso,

A final disposition of "Accept" has been registered for the above-mentioned manuscript.

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Kind regards,

Journal of Hazardous Materials

Comments from the Editors and Reviewers:

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# Journal of Hazardous Materials

## Studies on the performance of bentonite and its composite as phosphate adsorbent and phosphate supplementation for plant --Manuscript Draft--

<b>Manuscript Number:</b>	HAZMAT-D-20-03525R2
<b>Article Type:</b>	Research Paper
<b>Keywords:</b>	Organo bentonite; phosphate; adsorption; rarasaponin; root elongation.
<b>Corresponding Author:</b>	Shella Permatasari Santoso, Ph.D. Widya Mandala Catholic University Surabaya, INDONESIA
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<b>Abstract:</b>	<p>Organo-bentonite (OrB) was prepared by modifying bentonite with chitosan, and natural surfactant extracted from <i>Sapindus rarak</i> fruit. The physical alteration post-modification, performance of phosphates (Pi) adsorption, and possibility as a Pi-supplementation for plants of OrB were assessed and compared to acid-activated bentonite (AAB). The physical alteration due to modification of bentonite was characterized. SEM images were not indicating significant morphology differences between OrB and AAB. Existence of chitosan layers in OrB causes a decrease in basal spacing as characterized using XRD. The BET surface area of OrB was decreased compared to AAB due to pore coverage by chitosan. Adsorption studies reveal that OrB has a higher adsorption capacity towards Pi than AAB, which is 97.608 and 131.685 mg/g at 323K for AAB and OrB, respectively. The H-shape isotherm curve indicates that chemisorption is dominantly controlling the adsorption. The isotherm and kinetics adsorption were well fitted to Langmuir and Pseudo-second order models, respectively. Performance of AAB and OrB as Pi-supplementation was assessed based on growth phenotypes of <i>Arabidopsis thaliana</i>; seedlings show that supplementation of Pi@AAB and Pi@OrB (at half doses) can promote primary root extension. These results also demonstrate the safety of direct disposal of the materials into the soil.</p>