Dear Prof. Dr Bojana Obradović,

We appreciate Your and reviewers’ work on our manuscript entitled “Removal of herbicide 2,4-dichlorophenoxy acetic acid from water using of ultrahigh-efficient thermochemically activated carbon”. All the comments are valuable and very helpful. According to the comments and suggestions, we have revised our manuscript carefully and respond to each request. Corrections are blue marked in the revised manuscript. In any way, we wish to cordially thank reviewers for their useful suggestions. They influenced this manuscript to become much better. We hope that our manuscript in this form would be suitable for publication in your journal.

Thank you very much for your consideration. I am looking forward to hearing from you.

Sincerely,

Dr Miloš Kostić

ADDITIONAL COMMENTS

Please, be as specific as possible if major correction by the author(s) is recommended!:
The manuscript presents the adsorption of 2,4-D on activated carbon prepared from biomass Lagenaria vulgaris. A very similar article was published by the authors in the Arabian Journal of Chemistry (“Characterization of a low cost Lagenaria vulgaris based carbon for ranitidine removal from aqueous solutions”. Arabian Journal of Chemistry, 10, Issue 7, 2017, Pages 956-964). This paper has not been cited here! Therefore, I have thus doubt about the scientific up-to date state of the paper. So this is a clear shortcoming of this paper which calls for major revision even rejection! Authors should mention clearly the novelty in this work.

**Authors:** **The article in the Arabian Journal of Chemistry was primarily concerned with characterization, and less by application of materials. In these studies, different pollutants were used. In this manuscript, characterization of LVAC after sorption of 2.4D was performed. The possibilities of using this material in real waters and the possibility of its regeneration and reuse are examined in this manuscript. The article in the Arabian Journal of Chemistry is now cited.**

- line 47 – “2,4-D is non-volatile and highly soluble in water” - Is solubility really high?

**Authors: The sentence was corrected. Yes, 2,4-D has high solubility in water.** <http://npic.orst.edu/factsheets/archive/2%2C4-DTech.html>

- 115-117 – Use the same units, either Kelvin or Celsius degrees.

**Authors: Correction was done.**

- 119 – is “(mg/g1)” should be “(mg/g)”

**Authors: Correction was done.**

- 133 – is “the pseudo-first-order reaction rate equilibrium constant” should be “the pseudo-first-order adsorption rate constant”

**Authors: Correction was done.**

- 264 - How was the pHpzc determined?

**Authors: Your recommendations were accepted and applied in the section 2.4. The reference is added.**

- section 3.2.3 - Information about the running time of experiments should be added.

**Authors: Your suggestion is accepted. Samples were collected after 0, 0.5, 1, 5, 10, 20, 40, 60, 90, 120, 180 and 240 min and analyzed on the residual concentrations of 2,4D (this is added in section 2.4).**

- line 300 – is “kinetics constants” should be “kinetic constants”

**Authors: Your recommendation is accepted, and this sentence was changed.**

In my opinion, this manuscript should:

be published after major revision and additional review

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Reviewer

B:

ADDITIONAL COMMENTS

Please, be as specific as possible if major correction by the author(s) is recommended!:
The manuscript Removal of herbicide 2,4-dichlorophenoxy acetic acid from water using of ultrahigh-efficient 2 thermochemically activated carbon by Bojic et al. presents use of activated carbon obtained from biomass Lagenaria vulgaris for removal of the herbicide 2,4-dichlorophenoxy acetic acid from aqueous solutions. The activated carbon before and after adsorption studies was characterized by various techniques (SEM, EDS, FTIR, BET) while the adsorption process was investigated regarding kinetics and thermodynamic parameters. Also possibilities for reuse of the adsorbent were examined as well as the adsorption efficiency in a groundwater sample. The manuscript is clearly written and organized in a logical manner. However, activated carbon is largely used as a very efficient and available adsorbent in various adsorption processes for many years including wastewater treatment and water purification processes. It was also used in various studies for herbicide removal as also stated in the manuscript. Thus, the obtained results, although slightly better than those that were compared to in the manuscript, are largely expected while further analysis or hypotheses of reasons for such results is not provided. Therefore, the scientific contribution of the manuscript is not clear and should be clarified and included in the manuscript. Also there are some inconsistencies regarding presentation and discussion of kinetics modeling results that should be corrected.
Specific comments

P3 L53:

70 ug/dm should be 70 ug/dm3

**Authors: Correction was done.**

2.6. Recycling and reusing of LVAC sorbent

P9 L183:

“At this temperature, 2,4-D is decomposed and thus removed from the LVAC.”
Is this regeneration process feasible and could be scaled up? What are the resultant gases and would they be air pollutants?

**Authors: The separated gas which originates from 2,4-D can be caught in water, thereby concentrated in water, and subsequently be reused.**

2.7. Removal of 2,4-D from groundwater

P9 L189:

“2,4-D was added in a concentration of 10 mg/dm3.”
Why this concentration was chosen – arbitrarily or there was a specific reason?

**Authors:** **About 50 mg of 2,4-D is used per m2 of soil to remove weeds from it.** **The assumption is that only a lower concentration of 2,4-D can occur to groundwater.** **Since 50 mg/dm3 2,4-D is removed with an efficiency of 99.16%, lower concentration was chosen (certainly less than 50 mg/dm3 of 2,4-D will come in groundwater) to determine the removal efficacy at lower concentrations.**

Fig. 1 – What is the significance of the TG analysis? Is it different in any respect from other activated carbons?

**Authors: TG analysis was performed to determine stability of the LVAC at elevated temperatures, due to the selected regeneration process at elevated temperature. TG results may be different for diffrent chemically modified activated carbon.**

Fig. 2 – Is there a need for SEM micrographs? It is not expected that the herbicide will change in any respect the morphology of activated carbon.

**Authors: SEM micrographs can be confirmation of LVAC sorbent stability after sorption of 2,4-D. Conditions of application of a sorbent sometimes may change the sorbent morphology.**

3.2.1. Effect of initial concentration and contact time

P13 L239

“The sorbed amount at equilibrium was increased with the increase of the initial concentration of 2,4-D from 50 to 500 mg/dm3. The sorbed amount improved from 49.69 to 333.32 mg/g by increasing the initial 2,4-D concentration from 50 to 500 mg/dm3.”
These two sentences should be combined in one.

**Authors: Your recommendation is accepted, and this sentence was changed.**
P13 L253

“After equilibrium time, the sorbed amount of 2,4-D has almost no change with a further increase of the contact time.”

If the equilibrium is reached the sorbed amount does not change by definition. Please revise.

**Authors: This sentence was deleted because it was redundant.**

Fig. 3 – parameters of the experiment should be put in the legend for each graph; Fig 3b – please include error bars

**Authors: This has been changed according to your recommendation. The parameters of the experiment were added. Also, error bars were included in Fig 3b**

P14 L260

Please change “speed” to “rate”

**Authors: The “speed” to “rate” has been changed.**

P14 L262

“(about 33%, which is not great decrease)”

33 % is not insignificant. Please omit this description

**Authors: The sentence has been revised.**

P15 L263

“almost constant from the pH 2 to 7, an increase in the pH above 7 resulted in a small decrease in sorption (about 10%).”

This does not seem on the graph. Actually, if the point at pH 7 is omitted, a nice straight line of the decrease of qe with pH is obtained. The total decrease is 33 % and this cannot be described as an almost constant value up to pH 7 and a decrease of only 10 %. Please revise accordingly.

**Authors: This has been revised.**

3.2.3. Effect of sorbent dose and stirring speed

P15 L281

“while the sorption capacity of 2,4-D decreases”

The sorption capacity cannot decrease - it is the sorbed amount that is decreasing while the equilibrium capacity does not depend on the sorbent dosage. Please revise.

**Authors: The sentence have been modified according to your instructions.**

P16 L290

“Increasing of stirring rate ensure the higher availability of surface binding sites,”
How is that possible? Increase in stirring rate can just improve the convective mass transport by decreasing the liquid boundary layer. Please revise.

**Authors: The sentence has been modified.**

P16 L293

“interparticle space are accelerated”

The space cannot be accelerated. Please revise.

**Authors: The sentence has been revised.**

3.2.4. Kinetics study

P16 L298

“to determine the equilibrium time”

Equilibrium time is not determined by modeling. Please revise.

**Authors: The sentence has been revised.**

P16 L300

“The estimated kinetics constants are presented in Table 2.”

It is not sufficient to provide just table with parameters. Graphs with experimental data and best model predictions should be shown. The best models should be shown in the manuscript while the rest in the Supplementary material Models that do not fit the data should not be discussed.

**Authors:** **Because of the limitations of the journal, graphs with experimental data are shown in Supplement material, in Figure S1 and Figure S2.**

P17 L

“The r2 for the intraparticle diffusion model was ranged between 0.86 and 1, indicating that the intraparticle diffusion model can also fit the sorption process. The plot qt versus t0.5 presented a multilinearity relation and had three linear segments. The first linear segment on the plot is due to external surface sorption or instantaneous sorption. The slower sorption process observed in second linear stage came from of the intraparticle diffusion and last linear segment, indicating to the final equilibrium sorption process (the intraparticle diffusion starts to slow down due to the low solute concentration of 2,4-D in solution or the sorbent surface became saturated). The intraparticle diffusion is the rate-controlling step when qt versus t0.5 line passes through the origin, which is not the case here [34]. This indicates that the intraparticle diffusion was not the only rate-controlling step.”

Multilinear segments just show that the model is not applicable. The model is based on the approximation of the exact solution of the differential equation of diffusion through a spherical particle for short times (mt/minf <0.4; consider J. Siepmann, F. Siepmann, Modeling of diffusion controlled drug delivery, J. Controlled Release, 161 (2012) 351-362.). The external boundary layer could just delay adsorption and this would mean a negative constant C1. Here this constant is positive indicating instantaneous adsorption at time 0 (no resistances at all). So, this whole discussion is erroneous and should be omitted since this model is not applicable.

**Authors:** **Your suggestion is accepted. The intraparticle diffusion model has been omitted from manuscript and not discussed.**

P18 L328

“the legality of these three models (pseudo-second-order, Chrastil and intraparticle diffusion model).”
The intraparticle diffusion model is not applicable while from the graph, assumed adsorption mechanism and literature should be decided which of the other 2 models is best describing the adsorption process (each of these 2 models is based on a mechanism).

**Authors:** **The kinetic discussion have been modified according to your instructions**

3.2.5. Sorption isotherms

P19 L334

“calculated model parameters are presented in the Table 3.”

Again, model predictions with experimental data should be presented in graphs.

**Authors:** **The graph with experimental data are shown in Supplement material in Figure S2.**

Fig 4 – please include error bars

**Authors: The error bars were included in Fig 4**

3.5. Removal of 2,4-D from groundwater

Please show data. Without data presentation this section is not convincing and should be omitted.

**Authors:** **The graph with experimental data are shown in manuscript in Figure 6.**

P23 L395

“This and the previous results show”

Which previous results? Please state the reference.

**Authors:** **The sentence have been modified.** **It's about the results shown in this manuscript.**

In my opinion, this manuscript should:

be published after major revision and additional review