Response to Reviewer 3 Comments

**Point 1:** The title seems good, but the abstract seems to be fine. Please add one problem statement line in abstract to justify this sentence ``the long-term pilot-scale study is urged to be investigated.``.

**Response 1:** Thanks for the reviewer’s helpful suggestion. The objective statement has been added to the abstract to support the seriousness of the problem, and the revised sentences are as follows:

Biological activated carbon (BAC) biofilter coupling ultrafiltration (UF) is a promising process for the treatment of river water contaminated by pharmaceutical and personal care products (PPCPs). However, the pilot-scale study should be conducted to reveal the long-term removal performance and the respective contributions of BAC and UF.

**Point 2:** Research gap should be delivered on more clear way with directed necessity for the future research work.

**Response 2:** We agree with the suggestion of this reviewer. The research gap has been added in the Conclusion and is as follow:

To sum up, the results show that the proposed BAC-UF system can be effective in the treatment of river water polluted by PPCPs, conventional organic pollutants and ammonia nitrogen. The two-stage biofilms located in the activated carbon column and on the UF membrane synergistically, can be conducive to the removal performances. Besides, the results of this analysis can have significant implications for the conven-tional UF operation procedure and the ozone-activated carbon process, providing a simple decentralized approach to drinking water treatment for the areas where source water is contaminated with PPCPs. However, the mechanisms of the two-stage biofilm, such as bacterial and metazoan communities, membrane fouling and dissolved oxygen transfer, should be further investigated to enhance the removal efficiency and stability of this system.

**Point 3:** Introduction section must be written on more quality way, i.e., more up-to-date references addressed. Please target the specific gap such as 2015-2021 etc

**Response 3:** Thanks for the reviewer’s comment. The introduction has been revised carefully, including using up-to-date references. For example, the outdated references have been removed. Meanwhile, the articles published within the last two years were added, such as (the Reference2 Yu et al., 2020) and (the Reference21 Tang et al., 2018).

**Point 4:** Page 1 Line 38. Please cite this reference with existing reference 3….Role of nanotechnology for design and development of cosmeceutical: application in makeup and skin care.

**Response 4:** Thank you very much for your kind reminder. The reference has been cited. Please see line 38 in the revised manuscript.

**Point 5:** The novelty of the work must be clearly addressed and discussed, compare previous research with existing research findings and highlight novelty.

**Response 5:** Thanks very much for this comment. The BAC biofilter can remove the PPCPs, and then the following UF can reject micro-organisms and particles flowing out from the biofilter to ensure the quality of drinking water. The above coupling process makes up for the defects concerning respective operations of the BAC biofilter and UF. Although many pilot-scale setups were used to treat secondary wastewater effluent for water reclamation, this type of raw water quality was different from the river water, causing the different potential of biofilm growing. As far as we investigated, the lack of enough attention to long-term pilot study exists. Therefore, in this study, a BAC-UF system was carried out for several months with pilot scale to access the long-term removal performances and the respective contributions of BAC and UF. The relevant content has been supplemented and revised in the abstract to highlight the innovative points. The comparison of previous research with existing research findings was added in the final part of the results and discussion.

**Point 6:** What is the main challenge? Why author choose this material? Please highlight in the introduction part.

**Response 6:** Thanks very much for this comment. Biological activated carbon (BAC) combined the adsorption and biologic degradation consuming low power energy and chemicals without concern of DBPs production as well as no frequent updates for activated carbon media. The BAC biofilter can remove the PPCPs pollutants, and then the followed UF can reject microorganisms and particles flowing out from the biofilter to ensure the quality of drinking water. Thus, the BAC-UF process makes up for the defects of the respective operations of BAC biofilter and UF. Although many pilot-scale setups were used to treat secondary wastewater effluent for water reclamation, this type of raw water quality was different from the river water, causing the potential of biofilm growing differently. As far as we investigated, the lack of enough attention to long-term pilot study is present. Therefore, in this study, a BAC-UF system was carried out for several months with a pilot scale.

**Point 7:** Page 2 Line 55 need a reference. Please consider these at end of this sentence……The oxidation method exhibited a fast reaction speed and high removal efficiency…(i) Role of nanomaterials in the treatment of wastewater: A review (ii) Advances and challenges in developing efficient graphene oxide-based ZnO photocatalysts for dye photo-oxidation.

**Response 7:** Thank you very much for your kind reminder. The two references have been cited. Please see the lines 58.

**Point 8:** The main objective of the work must be written on the more clear and more concise way at the end of introduction section.

**Response 8:** T Thanks very much for this comment. The introduction has been improved for better reading.

**Point 9:** Please check the abbreviations of words throughout the article. All should be consistent. Please revise your paper accordingly since some issue occurs on several spots in the paper.

**Response 9:** Thank you very much for your kind reminder. Two PPCP mistakes have been revised. The abbreviation of chemical oxygen demand in previous version has been corrected to CODMn. The unnecessary abbreviations have been also corrected in the abstract.

**Point 10:** Please add chemical reagents section and stated all chemical with brand specifications.

**Response 10:** Thank you very much for your kind reminder. The added paragraph is as follows:

Potassium permanganate, H2SO4, potassium sodium tartrate tetrahydrate, Nessler’s reagent and NaOH were purchased from a commercial company and certified as AR purity (Guangzhou Chemical Reagent Factory, Guangzhou, China), while PPCP standards were provided by three companies. Sulfamethoxazole (SMX), Sulfadoxine (SD), Sulfachloropyridazine (SCP), Sulfamethoxypyridazine (SMP), Sulfaquinoxaline (SQX), Sulfathiazole (STZ), Doxycycline (DOX), Erythromycin (EM), Anhydroerythromycin (EA), Roxithromycin (ROX), Penicillin-G (PEN G), Clarithromycin (CAM), Norfloxacin (NOR), Oxociprofloxacin (OFL), Enrofloxacin (EFL), Flumequine (FQ), Acetaminophen (APAP), Diclofenac sodium (DCF), Naproxen (NAP), Indomethacine (IND), Metoprolol (METO), Propranolol (), Atenolol (ATL), Primidone (PRM), Carbamazepine (CMZ), and Sulpiride (SP) were obtained from the Dr. Ehrenstorfer Company in Germany. Furthermore, Sulfadimidine (SM2), Sulfadiazine (SDZ), Sulfapyridine (SP), Sulfamonomethoxine (SMM), Tetracycline (TC), Ofloxacin (OFL), Amoxicillin (AM), Dimetridazole (DMZ), Trimethoprim (TMP) were bought from the National Institute of Metrology in China, whereas Oxytetracycline (OTC), Caffeine (CF), and Diethyltoluamide (DEET) were acquired from the Toronto Research Chemicals Company in Canada.

**Point 11:** Regarding the replications, authors confirmed that replications of experiment were carried out. However, these results are not shown in the manuscript, how many replicated were carried out by experiment? Results seem to be related to a unique experiment. Please, clarify whether the results of this document are from a single experiment or from an average resulting from replications. If replicated were carried out, the use of average data is required as well as the standard deviation in the results and figures shown throughout the manuscript. In case of showing only one replicate explain why only one is shown and include the standard deviations.

**Response 11:** Thanks very much for this comment. These three GAC-UF systems were operated in parallel. Samples of the feedwater and effluent from three systems were taken simultaneously and measured once. The standard deviation was obtained by the detection over the full period of the experiment.

**Point 12:** Please use Fig. or figure? It very confusing. Article should be in one pattern. Please follow the journal guidelines. Please revise your paper accordingly since some issue occurs on several spots in the paper.

**Response 12:** Thanks very much for this comment. The figure is used as the only pattern in the revised manuscript.

**Point 13:** Please provide high quality image of figure 1.

**Response 13:** Thank you very much for your kind reminder. The front sizes in the figure 1 have been increased. The quality of the figure 1 has been also improved.

**Point 14:** Please use one style for units such as m3/h or m3h-1 Please revise your paper accordingly since some issue occurs on several spots in the paper.

**Response 14:** Thank you very much for your kind reminder. The style for units has been revised and unified.

**Point 15:** Please add a comparative profile section to compare your results and prove how it better than previous.

**Response 15:** Thanks very much for this comment. The comparison of previous research with existing research findings was added in the final part of the results and discussion. The added paragraph is as follows:

PPCPs risks have been posing severe challenges to the safety of drinking water supply in rural areas due to the absence of the process with simple operation and maintenance as well as reliable performance. In this study, BAC coupling gravi-ty-driven UF was performed continuously, and the rejection performance of mem-brane filtration and BAC filtration both showed barriers for the conventional pollu-tants and PPCPs. Furthermore, this study indicated the respective contributions of BAC and UF, showing the role of the two-stage biofilm. Previous works involving BAC generally combined the ozonation with the BAC filter for treating the contaminants of emerging concern, eliminating a majority of PPCPs by more than 90% [34]. However, the regulation and maintenance of machines for ozone products are complicated, and the disinfection by-products will be newly generated in the effluent, which is incon-venient to use in rural areas [35]. In general, coagulation, filtration and single BAC units worked inefficiently and removed the detected PPCPs by less than 50%, as they were not hydrophobic [34, 36]. Hybrid membrane processes such as inline dosing of powdered activated carbon (PAC) prior to UF have already shown promising potential for the abatement of PPCPs; however, the inline dosing PAC is infeasible in rural areas [37]. In this study, the BAC prior to UF enhanced the biological activity by forming a two-stage biofilm system. Therefore, the integrated BAC-UF process can be considered as an economically and technically feasible approach to the decentralized and emer-gency drinking water treatment.

**Point 16:** Section 4 should be renamed by Conclusion and Future perspectives. Conclusion section is missing some perspective related to the future research work, quantify main research findings, highlight relevance of the work with respect to the field aspect. In the present form conclusion is very weird.

**Response 16:** Thank you very much for your kind reminder. The conclusion has been modified as suggested by reviewers. Please see lines 325-350 in the revised manuscript.

**Point 17:** To avoid grammar and linguistic mistakes, Major level English language should be thoroughly checked. Please revise your paper accordingly since several language issue occurs on several spots in the paper.

**Response 17:** We are so sorry to make reviewer’s reading uncomfortable. We have used an English Language Editing service to correct the grammatical and spelling errors and to make the expressions conform to correct scientific English (the Language Editing Certification is attached below). Two native English-speaking colleagues help us verify the manuscript. Hope the revised manuscript would be more satisfactory.

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**Point 18:** Reference formatting need carefully revision. All must be consistent in one formate. Please follow the journal guidelines.

**Response 18:** We agree with the reviewer. The reference formatting has been corrected. Thanks very much for the patient review again.