**Manuscript CJCE-24-0503**

Response to Reviewers

Dear Editor-In-Chief

João B. P. Soares

Thank you for the opportunity to submit a revised version of our manuscript, titled “Mathematical Modelling of Essential Oil Supercritical Carbon Dioxide Extraction from Chamomile Flowers,” for consideration in the Canadian Journal of Chemical Engineering. We greatly appreciate the time and effort you and the reviewers have invested in providing thoughtful feedback. The insightful comments and suggestions have significantly improved our work. We have incorporated all of the reviewers' recommendations, and the suggested changes. Below, we provide a point-by-point response to each comment, shown in bold font.

Response to Reviewers 2:

In this manuscript, authors reported the application of a mathematical model for the supercritical extraction process of essential oil from chamomile flowers. Authors pointed out that the applied model describes the governing mass transfer phenomena in a solid-fluid environment under supercritical conditions using carbon dioxide. They mentioned that the flow of carbon dioxide is assumed to be uniform across any cross-section, although the area available for the fluid phase can vary along the extractor. Furthermore, authors remarked that the physical properties of the solvent are estimated based on the Peng-Robinson equation of state. In addition, they also reported that the model parameters, including the partition factor, internal diffusion coefficient and decaying factor, were determined through maximum likelihood estimation based on experimental data assuming normally distributed errors.

In general, the manuscript is very attractive and interesting, as well as beneficial for applied purposes. I strongly recommend it for publishing in the Canadian Journal of Chemical Engineering. Meanwhile, the manuscript needs a major and substantial revision before its acceptance. I also invite authors to completely and fully address the comments raised here. The comments are explained below.

1. Please explain and highlight the novelties of this research work with respect to the previously   
   published papers, in the manuscript.

***Thank you for this comment. We have revised the text to address your concerns and hope that it is now clearer. Please see page 2 of the revised manuscript, lines 144 – 153.***

1. The motivations of conducting this research work are described.  
   ***Thank you for your valuable note! The motivations behind this work have been elaborated as in the text. Please see page 2 of the revised manuscript, lines 144 – 153.***
2. English language of manuscript needs a mild revision throughout the manuscript. For example, in page 9, lines 6 and 12, "the desorptiondissolutiondiffusion", must be corrected.   
   ***Thank you for your valuable note! The typo has been corrected.***
3. The manuscript title is recommended to be improved as "**Mathematical modelling of** **essential oil** supercritical carbon dioxide extraction **from chamomile flowers"**.  
   ***Thank you for your valuable note! The title has been modified according to the comment***
4. The provided highlights must be shortened as well as quantitative reported values.  
   ***Thank you for your valuable note! The revised highlights are presented below:***

* ***A supercritical extraction model with decaying extraction kinetics and Cubic Equation of State is presented***
* ***Model parameters are determined through maximum likelihood estimation based on a set of experimental performed at different operating conditions***
* ***The multiple linear regression is used to find correlations between the estimated parameters and operating conditions***
* ***The close fit between the model predictions and experimental data underscores the model's capability to capture the system dynamics***

1. The last statement of abstract section is ambiguous. In fact, authors did not perform any experiments. Accordingly, please remove it.  
   ***Thank you for your valuable note! The sentence has been removed as suggested***
2. The last paragraph of introduction must be rewritten, focusing the importance and necessity of this work.   
   ***Thank you for your valuable note! The necessity of this work has been elaborated on in the text. Please see page 2 of the revised manuscript, lines 138 – 153.***
3. As you may know, supercritical fluids such as carbon dioxide (SC-CO2) has many applications in various topics. Authors are requested inserting a proper statement to emphasise the importance of the utilising of SC-CO2 technology to be familiar for readers. For this purpose, write and insert this statement in the introduction section of the main manuscript along with fully cited references in below. "supercritical fluids like supercritical carbon dioxide (SC-CO2) has shown a great ability in various fields including extraction of **essential oil [**

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| [**https://doi.org/10.1016/j.supflu.2016.11.014,**](https://doi.org/10.1016/j.supflu.2016.11.014) | [**https://doi.org/10.1016/j.supflu.2014.07.023,**](https://doi.org/10.1016/j.supflu.2014.07.023) |
| [**https://doi.org/10.1016/j.supflu.2016.04.006,**](https://doi.org/10.1016/j.supflu.2016.04.006,) | [**https://doi.org/10.1016/j.supflu.2011.02.002**](https://doi.org/10.1016/j.supflu.2011.02.002)**,** |
| [**https://doi.org/10.1016/j.supflu.2016.05.015,**](https://doi.org/10.1016/j.supflu.2016.05.015) | [**https://doi.org/10.1016/j.supflu.2017.04.007,**](https://doi.org/10.1016/j.supflu.2017.04.007) |

[**https://doi.org/10.1016/j.jtice.2015.11.003**](https://doi.org/10.1016/j.jtice.2015.11.003)**, ], seed oil [**[**https://doi.org/10.1016/j.supflu.2016.08.019,**](https://doi.org/10.1016/j.supflu.2016.08.019)[**https://doi.org/10.1016/j.supflu.2015.12.004,**](https://doi.org/10.1016/j.supflu.2015.12.004)[**https://doi.org/10.1016/j.supflu.2017.12.026]**](https://doi.org/10.1016/j.supflu.2017.12.026)**, solubility [**[**https://doi.org/10.1002/cben.202200020,**](https://doi.org/10.1002/cben.202200020) **https://doi.org/10.1007/s11814-018-0125-6],**

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| **nanoparticle formation** | **[**[**https://doi.org/10.1016/j.supflu.2017.10.015,**](https://doi.org/10.1016/j.supflu.2017.10.015) |
| [**https://doi.org/10.1016/j.heliyon.2020.e04947,**](https://doi.org/10.1016/j.heliyon.2020.e04947) | [**https://doi.org/10.1016/j.supflu.2021.105163,**](https://doi.org/10.1016/j.supflu.2021.105163) |
| [**https://doi.org/10.1016/j.jcou.2021.101799**](https://doi.org/10.1016/j.jcou.2021.101799)**,** | [**https://doi.org/10.1016/j.supflu.2018.11.007**](https://doi.org/10.1016/j.supflu.2018.11.007) |
| [**https://doi.org/10.1016/j.cherd.2018.12.020**](https://doi.org/10.1016/j.cherd.2018.12.020)**,** | [**https://doi.org/10.1016/j.fluid.2018.11.006**](https://doi.org/10.1016/j.fluid.2018.11.006)**,** |
| [**https://doi.org/10.1016/j.supflu.2018.06.009]**](https://doi.org/10.1016/j.supflu.2018.06.009)**,** | **impregnation** |
| **[**[**https://doi.org/10.1016/j.supflu.2020.104892,**](https://doi.org/10.1016/j.supflu.2020.104892) | [**https://doi.org/10.1016/j.supflu.2022.105674]**](https://doi.org/10.1016/j.supflu.2022.105674)**,** |
| **optimisation and mathematical modeling** | [**[https://doi.org/10.1080/14786419.2017.1361954**](https://doi.org/10.1080/14786419.2017.1361954)**,** |
| [**https://doi.org/10.1016/j.supflu.2017.04.007]**](https://doi.org/10.1016/j.supflu.2017.04.007)**,** | **polymer synthesis** |

**[**[**https://doi.org/10.1016/j.supflu.2022.105679]**](https://doi.org/10.1016/j.supflu.2022.105679)**, etc.**

***Thank you for your valuable note! As suggested by the reviewer, the literature review has been expanded. Please see page 1 of the revised manuscript, lines 7 – 42.***

1. Literature review on the available models could be completed in the introduction section.  
   ***Thank you for your valuable note! The literature review on the available models has been expanded. Please see page 1 of the revised manuscript, lines 64 – 68 and page 2, lines 69 – 123.***
2. As the authors did not conduct any experimental work by themselves, please remove section 2.4.  
   ***Thank you for your valuable note!  
   The section 2.4 was removed. The reference to the experimental work are presented is presented in the revised manuscript on page 2, lines 155 – 166***
3. Please explain the superiority of the applied model over the similar models in the literature.  
   ***Thank you for your valuable note!  
   The superiority of the model has been elaborated in the text. Please see page 8 of the revised manuscript, lines 510 – 518 and page 9, lines 519 – 538.***
4. Could you report the error between experimental data and model predictions by statistical metrics including AARD%?  
   ***Thank you for your valuable note! The information on modelling errors can be found in the revised manuscript in Table 2, page***
5. In the material section, please insert the name of material.  
   ***Thank you for your valuable note! The reference to the experimental work can be found on page 2 of the revised manuscript, lines 155 – 166.***
6. The results and discussion section must be strengthening by comparing the similar works and researches such as **[**[**https://doi.org/10.1016/j.supflu.2017.04.007**](https://doi.org/10.1016/j.supflu.2017.04.007)**, https://doi.org/10.1080/14786419.2017.1361954].**   
   ***Thank you for your valuable note! We agree with the reviewer that comparing our model with other works enhances the discussion of how implementing the modifications suggested in this work improved the model performance.***

***Please see page 7, lines 480 – 501 and page 8, lines 502 – 509 for comparison between the findings of this study and those of Povh et al. and Rahimi et al.. Both cited articles utilized the same dataset, allowing for a direct comparison of models.***

***The study described in*** [***https://doi.org/10.1016/j.supflu.2017.04.007***](https://doi.org/10.1016/j.supflu.2017.04.007) ***explores the supercritical extraction of “E. billardieri” under various operating conditions. The manuscript suggested by a review utilises a different dataset and models, such as a first-principle model based on BET theory and an empirical model using second-order polynomial regression. Given these differences in datasets and model structures, a direct comparison with the current work become ambiguous.***

***Similarly, the study in*** [***https://doi.org/10.1080/14786419.2017.1361954***](https://doi.org/10.1080/14786419.2017.1361954) ***examines the supercritical extraction of oil from “Dracocephalum kotschyi Boiss” seeds. The manuscript suggested by the reviewer utilises a different dataset and a different model (an empirical model based on second-order polynomial regression). Due to different nature of the datasets and model structures, direct comparison with the present study become ambiguous.***

1. Explain clearly the limitations of this study?  
   ***Thank you for your valuable note!  
   The discusson on model limitations has been elaborated in the revised manuscript. Please see page 8 of the revised manuscript, lines 510 – 518 and page 9, lines 519 – 538.***
2. Could you correlate the experimental data to two empirical models for solubility determination of components in supercritical carbon dioxide, proposed in these references [[](%5b)[**https://doi.org/10.1016/j.supflu.2019.01.006,**](https://doi.org/10.1016/j.supflu.2019.01.006)**<https://doi.org/10.1016/j.jcou.2024.102687>]**?  
   ***Thank you for your valuable note! We appreciate the reviewer’s insightful suggestion and agree that it would be useful to demonstrate the effect of soubility, however such an analysis is beyond the scope of our paper, which aims only to show a gernalized model with pseudo-one-component solute, which is valid for a range of operating conditions.***

***The empirical correlations proposed by the reviewer has been prepared for pure “Imatinib Mesylate” (***[***https://doi.org/10.1016/j.supflu.2019.01.006***](https://doi.org/10.1016/j.supflu.2019.01.006)***) and “Dapagliflozin Propandiol Monohydrate” (***[***https://doi.org/10.1016/j.jcou.2024.102687***](https://doi.org/10.1016/j.jcou.2024.102687)***) and they are not applicable to this work. The presence of none of the substances mentioned above was confirmed in our work. Moreover, the presented model does not incorporate solubility directly. As discussed in the manuscript, it is assumed that the system has not reached saturation, meaning the maximum mass of solid dissolved in the solvent or the solute concentration in the fluid phase remains unknown. The solubility could theoretically be used to define an upper boundary for the state space but in the absence of empirical data. In such a case, solubility estimates may be arbitrary, which improves model fitness without necessarily having physical interpretation. Additionally, correlating the estimated parameters with solubility models would introduce more parameters into the empirical correlations for DiR and ϒ, which could result in overfitting, given the limited dataset. For accurate solubility estimation, it would be necessary to analyze the oil composition and conduct dedicated experiments focused on solubility..***

1. Please provide abbreviations and symbol lists.  
   ***Thank you for your valuable note!  
   See the last page of the revised manuscript***
2. It is recommended removing the last paragraph of conclusion section.  
   ***Thank you for your valuable note!   
   The paragraph was removed***
3. Please provide consistency between the abstract and conclusion sections.  
   ***Thank you for your valuable note!  
   The paragraph and concusion section has been revised***
4. The number of references could be increased for this type of research work.  
   ***Thank you for your valuable note!  
   The number of references increased from 20 to 30***
5. Please set the reference style as per the Journal guidelines.   
   ***Thank you for your valuable note!  
   The citations style has been revised***

We would like to thank the referee again for taking the time to review our manuscript.