Reviewer #1 – Anonymous

I believe the paper offers a significant contribution to scientific progress within the scope of the journal, and it will be of wide interest to the geoscience community, particularly in the field of 3D mineralogy and petrology. This paper introduces a new workflow for digital image processing, and aims to revolutionize current methodologies of XCT image processing in the field of in-situ time-evolving synchrotron XCT datasets, which are often very large in size and time-consuming to analyse due to the challenges in low contrast, noise, evolving mineral phases. I believe the proposed workflow is quite robust and valid, as the authors cross-check the accuracy of the proposed method against theoretical molar evolution of gypsum to bassanite during the reaction, and their measured values fall within 2% confidence intervals. In addition, they also provide a robust presentation and comparison with other more traditional methods of image segmentation, with and without data augmentation or machine-learning labelled ground truth data, and also provide suggestions to make the method even less time-consuming. Overall, I believe this method would be applicable in many fields and will greatly improve digital image analyses in time-evolving synchrotron XCT datasets or even standard XCT scans where mineral phases may overlap in intensity.

We sincerely thank Reviewer #1 for their thorough review and positive feedback on our manuscript. We are pleased to know our work is considered a valuable contribution to the Geosciences' community. We appreciate the constructive comments, which we have largely accepted and incorporated into the manuscript. Our responses are colour-coded in blue.

I would be interested in seeing how the accuracy of this workflow can be checked when there is no prior-knowledge of a reaction, or when there is no theoretical curve to check it against with. This might be the case for other geological processes, where for example different mechanisms play a role, and where the molar volumes of mineral phases are not so well known. How do the authors propose to check their workflow in those instances? It would be good to include this explanation in the discussion.

As explained in the Discussion section of our paper, a prevalent approach in domains like medical imaging and material science involves external calibration techniques. These techniques utilize phantoms—objects with predefined dimensions and/or compositions—as reference standards. Phantoms function as external benchmarks, enabling the evaluation of segmentation method accuracy through imaging and segmentation. Assessing the segmented volumes or dimensions of these phantoms against their actual known values provides insights into the accuracy and dependability of segmentation techniques in µCT imaging.

In addition to these strategies, other methods that could be considered for benchmarking segmentation outputs include: (a) Comparative Studies: Validation can be achieved by conducting comparative studies with established, well-validated segmentation algorithms. Aligning new segmentation results with those from recognized methods enhances the credibility of the novel approach. (b) Synthetic or Simulated Data: Employing synthetic or simulated datasets, where the accurate segmentation is pre-determined, offers a robust validation tool. By applying the segmentation algorithm to these controlled environments, we can comprehensively evaluate its precision and reliability.

I found that the overall presentation quality could be improved as some concepts mentioned in the paper are too technical for a non – expert audience, and they need explanation.

In response to the detailed suggestions from Reviewer 1, we have incorporated explanations for technical terms and expressions into the manuscript. Further elaboration on these additions is available in our responses to the Detailed Comments section.

For instance, many readers will not be familiar with terms such as "supervised deep learning". What is a supervised deep learning method? How does it differ from an unsupervised one? The authors mentioned both concepts in the paper, yet they fail to explain what they are and how they differ. They also did not explain why they chose one rather than the other. It would be good to at least explain the difference between the 2 methods (since both are mentioned) and why the authors made that choice, so that the readers can better understand what may or may not work in other contexts where this workflow may help in the analysis.

As recommended, we have added a concise yet thorough explanation of Deep Learning in the context of image segmentation to the 'Introduction' section. We have also elaborated on the differences between Supervised and Unsupervised Deep Learning for image segmentation and explained our rationale for opting for Supervised segmentation in our study.

Furthermore, when possible, terminology belonging to machine-learning should be avoided, as this journal covers a great variety of topics, and while some readers may be familiar with terms such as "(hyper)-parameters", these may not always be clear to a non-expert reader. Why are they (hyper) parameters and not just parameters? I would suggest avoiding such technical terminology when possible, or if needed, then it needs some explanation. The authors explain what (hyper)-parameters they used, but it is not clear what (hyper)-parameters are.

While we understand that some readers might find unclear the use of terms such as "(hyper-)parameters", these are well-established and precise terms

in the AI user community which have been used in the scientific literature for more that 10 years. We agree with the reviewers that we need to provide some explanation for the terms used, however we believe that is also important to use the correct terminology particularly if this is has recently introduced in the Geosciences community. A full explanation of the term (hyper-)parameter is provided for the comment in the Detailed Comments section.

Some concepts are introduced without explanation of if there is one, it is presented in different sections. I flagged in the commented text where I could: for instance, Random Forest is not cross-referenced with the section in the Appendix. I think introducing cross-referencing to these sections next to the concept would help non-expert readers (example: random forest, sec. 3.3, Appendix X).

We thank the reviewer for pointing this out. We have now updated the text so that it points to the Appendix.

The paper also contains some minor typos and small inaccuracies, like lack of introduction of acronyms.

We thank the reviewer for flagging these errors, we have now reviewed and corrected them, please see the "Detailed Comments" section here below where we reported the notes left by the reviewer on the PDF version of the manuscript.

Some of the figures could be improved and be bigger (not sure if it is the formatting of the generated pdf). It would be good to have an overall figure (with the grain of celestite) showing all the steps, including the post-processing cleaning up.

We have considered the suggestions made by Reviewer 1 and updated Figures 1 and 2 so that now they show larger image and bigger labels.

Overall, I think the paper is a great scientific contribution to the community. Providing minor revisions are made (specifically targeting the improvement of clarity for non-expert readers), I suggest that the paper is accepted for publication.

Once more we thank Reviewer 1 for the suggestions which have helped clarify some aspect of the manuscript.

Detailed Comments:

Line 28: why only micro and not nano? surely you can make this XCT so that it includes both nano and micro. Also the acronym has not been introduced before and needs explanation.

We prefer using the term µCT as x-ray microtomography is the standard term for the technique. XCT as an abbreviation for X-ray computed tomography is more consistent with medical CT.

Line 28 – 30: this sentence is too long, break it.

Now: "Time-resolved (4D) operando experiments in X-ray Computed Microtomography (µCT) scanners have emerged as a promising way of studying solid-state reactions offering unprecedented insight into mineral phases and volume changes. This method is becoming a technique of choice for many geoscience problems because it provides information about both the spatial and temporal evolution of the microstructure of a sample."

Line 31: spatial resolutions.

Added as suggested

Line 53 – 55: Worth explaining that using any type of filtering may restrict even further the limited greyscale information available.

We have now added the following sentence: "This is most clearly seen in data that contain low contrast phases, for which filtering processes to reduce noise or enhance feature visibility may modify or remove variations in intensity that are critical for accurate phase differentiation and segmentation"

Line 55 – 56: ..also worth explaining to a non-expert audience that this is because the threshold may vary across different time steps.

Following Reviewer's suggestion we have modified the text as: "Moreover, as the grayscale image inputs vary over time in time-series datasets, the effectiveness of histogram thresholding diminishes. This is because the optimal threshold for one time step may not be applicable for others, leading to inconsistent or inaccurate segmentations."

Line 57: do you meant characteristics?

Modified as suggested.

Line 60: what is a supervised deep learning? and how is it different from an unsupervised one? worth explain why you chose this one in particular?

We have now added the following sentence to clarify the meaning of supervised and unsupervised deep learning: "Supervised deep learning is a type of machine learning where the model is trained on a labelled dataset. This means that each output produced by the model is paired with the correct output, enabling the model to learn by comparing its predictions to the actual outcomes. This contrasts with unsupervised deep learning, where the model attempts to identify patterns and relationships directly from the input data without labelled outcomes."

We opted for supervised deep learning primarily due to its accessibility, as it is supported by several commercial and freely available segmentation software (such as Avizo, Dragonfly, and ilastik), and its relative ease of implementation compared to unsupervised learning, which often requires high-performance computing clusters

Line 84: as in creation of more porosity.

That's correct. However, here in the text we are describing the chemical reaction, therefore we think is more accurate to talk about water and not porosity.

Line 90: any specific reason why you choose these 2 in particular?

The two samples were chosen because they represent "endmembers" experiments: with VA17 deformed under low differential pressure (with confining pressure > than differential), and VA19 under high differential pressure (and low confining).

To include the motivation we have modified the text as follows: "For the work presented in this paper, we focus on data from two specific experiments, chosen as 'end-member' scenarios for their distinct evolving mineral fabric during the dehydration process…"

Line 91: not to be picky but CT stands for computed tomography and you used this acronym before to indicate X-Ray microComputed Tomography, now you call synchrotron microtomography with the same acronym even though we lost the computed and we add synchrotron. perhaps use a different acronym? or just call it synchrotron XCT since you introduced XCT before?

We have answered a similar question earlier in the comments. For this work we think the use of µCT is more correct since the technique employed is indeed X-ray microtomography or µCT, with Synchrotron X-ray Microtomography as S-µCT.

Line 92: SLS not defined... well I know it means Swiss Light Source but you introduce acronyms without explaining them first!

Added to text in line 85.

Line 93: why 27KeV?

This is the peak energy value observed at TOMCAT beamline, after optimising the filters for imaging with the Mjolnir rig.

The filter white beam characteristic is defined by the synchrotron source and beamline parameters. It mainly depends on (non-extensive) the storage beams current and energy for the source and the type of insertion device (bending magnet, wiggler etc.) for the beamline. For these experiments, we filtered the lower energy range of the white beam for improving the imaging quality. The appropriate filtering quantity is determined as a function of the rig and sample materials (nature, thickness) to have an optimal number of photons on the detector. Filtering shifts the peak energy of the white beam and was here measured at 27 KeV. This energy is high enough to provide a good flux of photon able be transmitted through t the aluminium, pressure vessel and the sample.

Line 95: are these timings referring to those 2 samples or all experiments? unclear, do they vary between 150 and 314 minutes?

The time frame we are referring to here is indicative of the whole suite of experiments.

Line 98: can we make the figure bigger? especially the individual slices. I can see the changes but if the figures were a tad bigger that would be better

We have now increased the size of the image in the PDF file. We have also modified the figure to include the direction of the slices.

Line 100: why? needs explanation for non-expert readers earlier that this statement

To clarify the sentence we have now added: "… as the optimal threshold varies across different time steps".

Line 117: reference formatting

Fixed

Figure 2 caption: "slice in the tomographic scan" instead of "individual image". "slices" (and which direction) instead of "images".

Modified as suggested

Line 126: what are network "hyper"-parameters. why can they not be called just parameters in this case?

In the context of machine learning and deep learning, 'parameters' and '(hyper-)parameters' refer to two distinct types of variables within a neural network model. Parameters are the internal elements of the model, such as weights and biases, that the model learns through training. These are adjusted automatically during the training process as the model learns from the data.

In contrast, (hyper-)parameters are the external settings of the model that are set prior to training and remain constant during the training process. These include choices such as learning rate, number of hidden layers, batch size, and epochs. Hyperparameters guide the learning process but are not learned from the data themselves. They are crucial for the model's architecture and learning process but require manual setting or optimization, often through experimentation.

Therefore, we refer to these settings as '(hyper-)parameters' to distinguish them from the learned 'parameters' of the network, as they play a different but equally crucial role in the development and performance of the model.

To clarify this we have modified the sentence in Line 126: "Choosing the best training neural network architecture and tuning the network (hyper-)parameters – i.e., those settings of the model that are set prior to training and remain constant during the training process – requires time and some knowledge of neural network architecture."

Line 151: obtained through random forest?

This is a general behaviour of the Neural network architecture, independent from the kind of input data.

Line 153: "training"

Modified as suggested

Line 163 – 165: this sentence is unclear to me: you say for example i) histogram thresholding then ii) ..as provided by histogram thresholding.. what do you mean? it feels like a repetition, so please clarify the sentence. Also, the second part of the sentence is grammatically disconnected.

We have revised the sentence as follows: "To establish the ground truth set for initial image classification, we initially considered methods with differing informational depths. One such method is histogram thresholding, which relies on basic greyscale values and typically results in low-

information-level outcomes. However, this approach alone proved inadequate for our purpose, as it often led to gradients with diffuse phase boundaries, underscoring the need for more sophisticated classification methods."

Line 168: a visual quality check?

The check was done both visually but also using evaluation metrics (See figure in the appendix). We have now added a reference to the figure and the text in the appendix for clarity.

Line 171: perhaps it is me, but there are no values in brackets: better to say that they are indicated by different colours in the histogram curve?

For clarity we modified the text as follows: "The corresponding colour-coded threshold values for the four phases are shown in Figure 3b"

Line 176: I do not understand what this means. What does it mean that you octupled the input data in actual terms? Audience will less expertise on the matter will find hard to understand what this means. From this sentence alone, it is not clear to me what is data augmentation and how that improved the results of the data segmentation.

To clarify, 'octupling the input data' refers to the process of increasing the size of our training dataset by a factor of eight through data augmentation. This technique involves applying a series of basic image manipulations, such as flipping horizontally, flipping vertically, rotating, shearing, and scaling, to each image in our original dataset. These manipulations generate multiple variations of each image, thereby expanding our dataset and enhancing the diversity of training examples. This strategy is critical for improving the neural network's generalization capabilities and reducing the risk of overfitting, especially given our deliberate use of a smaller initial dataset.

To ensure comprehensibility, we propose to slightly rephrase the sentence in the paper to:" The training data were subjected to data augmentation based on the basic image manipulations (i.e., flip horizontally, flip vertically, rotate, shear, and scale). Specifically, we octupled the input data – i.e., generating eight variations of each original image – in order to increase its initial size rendering the neural network more robust, while at the same time compensating for deliberately using a small input dataset."

This revision aims to succinctly explain the concept of data augmentation and its purpose in our study, making it more accessible to readers who may be less familiar with these machine learning practices.

Line 181 and Figure 3: it would help if in the figure you can indicate which grains are celestite.
also why is the red of bassanite in fig 3e more like an orange whereas in d and c it is more like a faint red like in the histogram? the color in the legend does not correspond to the shaded area in the histogram, it is a different shade at least in this figure. please be consistent

The celestite is already labelled in Figure 3a and we prefer not to add extra text risking cramming the figure. We have now fixed the colours scheme in Fig. 3e.

Line 191. how do the various filter affect the output of the random forest? are they applied all toegther or separately and then the one that receive the most votes is chosen?

Thank you for your question. In our study, the Random Forest classifier was employed with a set of predefined features: Morphological, Gaussian Multi-Scale, and Neighbours. Each of these feature sets plays a distinct role in enhancing the classifier's ability to accurately segment phases in the dataset.

Morphological Features: These are used to analyse the shape and structure within the images, enabling the classifier to detect and distinguish different phases based on their morphological characteristics.
Gaussian Multi-Scale Features: These features involve applying Gaussian filters at multiple scales, aiding in smoothing the images and reducing noise. This multi-scale approach helps in capturing features at various levels of detail, contributing to more effective phase differentiation.
Neighbours Features: This set focuses on the local neighbourhood of each pixel, capturing the texture and local contrast, which is essential for identifying subtle boundaries between phases.

All these features are used together in the Random Forest classifier, each contributing to the overall classification task. The classifier does not operate on a voting system between these feature sets; rather, it integrates the information provided by all of them to decide for each voxel in the image. This integrated approach enables a more nuanced and accurate classification compared to using any single feature set on its own, and significantly improves the process over manual thresholding methods.

We have now provided these details in section "Appendix B: Random Forest Segmentation"

Figure 3 caption: "cross-sectional" instead of "Horizontal"

Modified as suggested

Line 215: indicating that is the best result?

Yes, values closer to 1 indicate a better scoring and that the outputs produced by the deep learning model is closer to the labelled input the user has provided.

Line 224 – 226: I would rephrase this sentence with "the model trained using a ground truth from a random forest classifier was the only model producing a DICE score of 0.98 for all phases." to make it clearer.

This is not entirely true. When using the RF model, the DICE score is not 0.98 for all phases (as shown in Table 1). 0.98 is the max value reached by the DICE score, specifically for "Gypsum DICE". The main advantage of RF over Histogram segmentation is that RF is the only model able to produce a score (and therefore correctly identify) for the Celestite phase.

Line 230: why should they not be consecutive?!

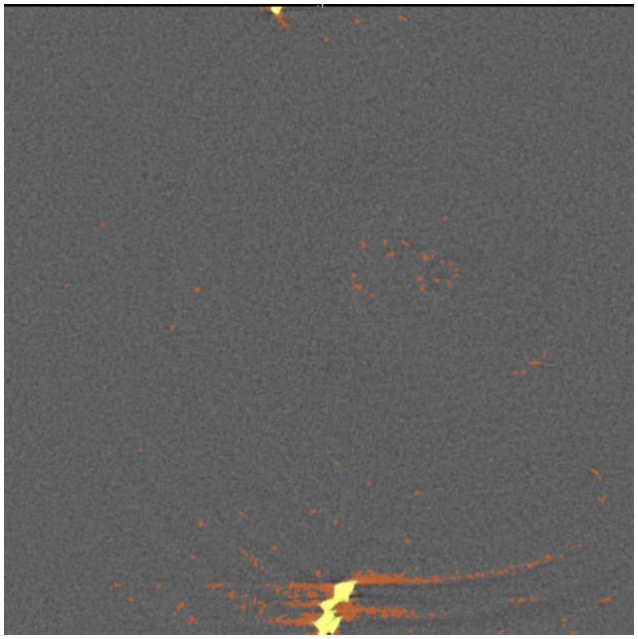
We could have chosen to test the model over a set of not consecutive slices cropped from the µCT volume.

Line 234: can the model be applied as batch-process using some IT cluster?

Yes, the model can indeed be run outside the Dragonfly platform for batch processing on clusters. However, this would involve implementing a series of scripts or code that perform the same steps currently executed within Dragonfly. These steps include data pre-processing, applying the model to the data, and handling the output. While this requires additional effort to set up the necessary computational environment and codebase, it is certainly feasible and allows the model to be utilised in a more flexible and scalable manner.

Figure 5: the results are rather astonishing however I can see for this celestite grain that celestite has been classified where the original image is grey (middle of the grain). Is it possible to see the same grain after the post-processing where errors should have been cleaned up and mislabel pixels fixed? it would be good to see all steps for the same image so including the post-processing. or do you reckon those grey pixels are actually caused by something else? (and what)

The images shown in figure 5 are already post-processed and post-processing does not involve the celestite. As detailed in the paper the main targets of post-processing are mislabelled pixels of Gypsum interpreted as Bassanite during the early stages of dehydration. Mislabelling occurs because at early dehydration stages the S-µCT images are inherently noisy due to the homogeneity of the gypsum phase. I have attached a figure here that exemplifies the issues.



Line 251: I would like to see an example of that. It would be even better if it can be on the previous figure you showed.

See picture above.

Line 257: why is it minor? not clear to me from this sentence

The mislabelling problems are encountered in µCT volumes in the early stages of the dehydration and vanished as soon as the Bassanite phase begins to grow, reducing the homogeneity in the µCT.

Line 298 and Figure 7: are the data points in Fig 7 relative to the same time scan? and if so, how do we know how much reaction % there should be? Perhaps I am reading the figure in the wrong way.. in a), the second data point from the left measured an approx. 27% reaction %, whereas in b) is less than 20% which is quite a big difference. can you clarify why a) is more accurate based on this graph?

I find this figure hard to read, even with the caption.

Figure 7 presents a quantitative analysis of phase volume changes during the gypsum dehydration experiment, specifically focusing on the same time series data (volume VA19) segmented using two distinct methods. The first method (represented by dots in 'a') utilizes the workflow developed in this study, combining a random forest classifier with a deep learning model. The second method (represented by diamonds in 'b') employs a conventional thresholding segmentation strategy.

The discrepancy in reaction percentages between these two approaches, particularly noticeable in the celestite phase, highlights the enhanced accuracy of our developed workflow. Unlike conventional thresholding segmentation, our method can more precisely detect and quantify all phases, including those that are typically challenging to segment like celestite. This is demonstrated by the more consistent and accurate volume measurements obtained with our workflow compared to the underestimation of reaction extent by the conventional method.

Therefore, the differences in reaction percentages as observed in Figure 7 underscore the superior segmentation capabilities of our Random Forest + Deep Learning approach, particularly in accurately capturing phases that are not discernible using standard histogram thresholding methods.

Line 310 – 311: this last sentence is unclear or missing something.

Thanks for picking this up... there was a typo in the manuscript. This is now: "To ascertain the accuracy of the chosen deep learning model we compared the theoretical and measured molar evolution of gypsum to bassanite during dehydration"

Line 333: why is hyperparameters written like that, and earlier on called (hyper)-parameters? we still do not have a definition for what these (hyper) parameters are and why they are just not called parameters.

Thanks for flagging this typo. Now fixed.

Line 340: repetition ot the sentence... : "These external standards aid in the assurance of measurement accuracy [Writers et al., 2021]."

Thanks for pointing this out. This has now been fixed.

Line 349: what if there is no prior knowledge of the chemical reaction involved? what would be an alternative to check segmentation accuracy?

Thank you for your insightful question. Indeed, the a-priori knowledge of chemical reactions plays a crucial role in establishing a framework for assessing the accuracy of data extracted from µCT images, as mentioned in Section 4.1 of our paper.

As highlighted in the Discussion section of our paper, one common strategy in fields like medical imaging and material science is the use of external calibration techniques. These techniques involve employing phantoms—objects with known dimensions and/or compositions—as benchmarks. Phantoms serve as external standards that can be imaged and segmented to evaluate the accuracy of segmentation methods. By comparing the segmented volumes or dimensions of these phantoms with their known

actual values, we can assess the accuracy and reliability of segmentation techniques used in µCT imaging.

Additional alternative strategies that can be consider for benchmarking segmentation outputs are: (a) Comparative Studies: Conducting comparative studies with existing, well-validated segmentation algorithms can also serve as a validation technique. If the new segmentation results are in good agreement with those obtained from established methods, it adds credibility to the new method. (b) Synthetic or Simulated Data: Using synthetic or simulated datasets where the true segmentation is known can be a powerful tool for validation. By applying the segmentation algorithm to these controlled datasets, its accuracy and reliability can be rigorously tested.

Line 377: this sentence is not grammatically correct, please revise.

Thanks for the suggestion. Now: "Future iterations of our method will aim to expand its capabilities and applications."

Line 380: what is transfer learning and reinforcement learning? not clear from the follow up sentences.

Thank you for your question. In the text we have provided few references when mentioning these methods and for the scope of our paper we think that pointing the interested readers to these is sufficient. However, since also Reviewer 3 has raised the issue that this paragraph was unclear, we have now integrated the text as follows: "Future iterations of our method will aim to expand its capabilities and applications. A direction to explore is the integration of deep learning convolutional neural networks with transfer learning and reinforcement learning techniques. Transfer learning can leverage pre-trained models to reduce computational cost and improve generalisation ability (Kim et al., 2022), while reinforcement learning might provide dynamic and adaptive strategies for data acquisition and reconstruction (Le et al., 2022). Specifically, transfer learning could be utilised to adapt models initially trained on datasets derived using imaging techniques which provide higher textural resolutions (such as Scanning Electron Microscope – SEM), thereby enhancing their ability to generalise to complex datasets with minimal retraining. Reinforcement learning could play a crucial role in optimising data acquisition and reconstruction processes. By applying reinforcement learning algorithms, we could develop systems that dynamically adjust acquisition parameters or reconstruction techniques based on real-time feedback, leading to more efficient and accurate image analysis. For instance, in time-evolving systems, reinforcement learning could be used to adaptively select optimal imaging parameters for each time step, based on the changes observed in the previous scans."

Line 385: it would be good to explain how supervised and unsupervised methods differ since you talked about both concept in this paper.

As detailed in a previous response we have now integrated in the text the difference between the two approaches.

Line 387: please connect these two sentences, or if using two separate sentences do not use "while" at the beginning of the sentence

We have now revised the paragraphs as follows: “Unsupervised learning can dramatically reduce the time and effort required for data annotation, thereby accelerating analysis, and enabling the exploration of larger datasets (Mahdaviara et al., 2023). Additionally, leveraging data from before and after a scan in a time series can provide extra information, further enhancing our ability to segment complex datasets more effectively.”