Reviewer #1

We thank Reviewer 1 for a detailed and helpful review. Please see below our response and proposed changes to specific review comments (italic), which we have grouped below. The PDF includes some additional comments that have been pasted into the below response and highlighted with "(pdf)".

Please make clear if the diffusion experiments and especially the model for gas release was conducted in this project or are largely based on Bergelin et al. (2025).

Here and throughout the review and pdf, this reviewer highlights the need to clearly distinguish between work conducted by Bergelin et al., (2025), and that presented here as new work. The other reviewer points this out as well. Throughout the manuscript, where Bergelin et al. (2025) is referenced, we will make the necessary revisions to clearly distinguish between the two.

I understand that you incorporated the results of Method 1 directly in the same paragraph (the same goes for Method 2). Maybe think of changing the subheadings to Method and Results or something similar where you can directly incorporate your results. Otherwise differentiate between the method section and the result section (see Method 3).

Here and throughout the review and pdf, this reviewer highlights partial results to be presented in the result section. As the reviewer recognizes, the organization is more complicated than usual in order to clearly separate the experimental design elements of the paper from the results of the experiment, but it appears the organization needs some improvement. Thus, we will reorganize the sections and subsections in the revised manuscript, such that partial results derived from each of the three methods discussed are presented as a subsection within that method, and not as a final result.

Introduction

Line Section 52-60: Avoid reporting from the we perspective if you refer to your previous paper, as it might be confusing for the reader if you are now writing about the recent project or referring to the published project.

2.1. Diffusion kinetics

Avoid writing from the me/we perspective. In the Method Section (2.1 Diffusion kinetics..) it sounds like you are already discussing data from this new study. Rather write for example: In a recent study, Bergelin et al. (2025) found the diffusion….. This section sounds already like you are showing new results from this manuscript. Differentiate more between what was done compared what is new and the result of this new project.

Here, and throughout the review and pdf, it is noted that the use of "we" when referring to previous work published in Bergelin et al. (2025) is not ideal and can cause confusion about what is presented as previous work and new work in this paper. We will make the necessary changes where needed in this section and throughout the paper.

Line 109 on: Are these results of the fractional releases from the previous work Bergelin et al. 2025 or from this work? If these are new results, they should be placed in the results section.

Please clarify if the modelling is new or done already in Bergelin et al. 2025, or if you model the fractional gas release of plag and pyrx based on results of and presented in Bergelin et al. 2025. Checking Bergelin et al. 2025, it looks like most of this is done/based in Bergelin et al. and serves here as an extra verification for the step-heating separation.

These are new data points presented here. However, as noted above with respect to the overall organization of the paper, they are not results of the virtual mineral separation procedure that is the main focus of the paper, but instead are data that were collected using established and previously reported methods for purposes of the experimental design of this study. Thus, we argue that these do not belong in the result section, as they are data sets used to design the heating schedule which is part of the method. As we outline in line 75-80, this project has multiple components to it, and are divided into three sections in which each describe methods and process validation results for each component, leading to the overall results of measured 3He concentrations in whole rock. To limit confusion, we will reorganize the sections such that the results from each of the three method sections are presented in the subsections.

Line 144 on: Also sounds like results from your modelling and should be placed in the result section.

Line 200: Why did you choose this grain size fraction, compared to the previously introduce pyroxene separates?

A smaller grain size fraction maximizes monocrystalline grains and is advantageous for CT scanning as each grain is more likely to consist of only plagioclase or pyroxene. We will add this to the revised text. We note, however, that in part our decision to use a grain size separate of crushed rock was motivated by the fact that at the beginning of this work we were not sure if the virtual mineral separation method would be possible on uncrushed whole rock. As we have now shown that this is possible, this makes the issue of grain size less relevant for future work.

3.2 CT scan

CT Scan duration 1sample /1hr?

Each batch scan contains 7-8 samples, and acquisition and reconstruction of the 1200 16-bit TIFF images takes approximately 1 hour, with an additional 1- 2 hours to crop and process all samples in a batch individually in Blob3D. The processing time per batch (7-8 samples) is based on an already established workflow for the segmentation. We will specify the duration of the CT scanning process in the revised manuscript.

The frequency distribution was calculated by the Zeiss software or Blob3D? How long does it take for the software to analyse one sample (so 1200 16-bit TIFF images). You might want to add, that Blob3D is a free software (easier access for other scientist if they want to use your virtual approach).

If I understand right, this segmentation strategy was done on each of 1200 16Bit TIFF images per sample, the attenuation groups where then cumulative summed up per sample and % calculated for high attenuation (i.e. other oxides), pyroxene and plagioclase?

This is correct (although it was only done on the cropped subsection of each scan that covered a single sample, and after cropping, the data set for a given sample was fewer than 1200 slices).

Can you incorporate one whole-rock analysis as image (with pyxr, plag and oxides marked with colour) and include this into Fig. 3 for comparison?

Each of the figures 3.d-f shows this, with red highlighting the mineral of interest, and a darker and light shade of grey representing the other two groups of mineral. We will add a description of the shades of grey representing each mineral group in the caption.

Line: 233 (pdf): can you give the CT number to check the supplementary table 2 for the data?

It's unclear which CT number the reviewer is referring to because each phase features a range of CT numbers, for reasons discussed in the text.

Rephrase Fig. 4 caption and text. Fig. 4 shows the mineral composition of your pyroxene separates differentiated by XRF analysis by Bergelin et al. (2024).

We will rephrase the text in the revised manuscript.

4 Method 3

Cosmogenic 3He measurements

Some initial results (for the CRONUS-P standard) are presented, which might be better placed in the results section. Moreover, is there any reason or explanation for the difference in the standard measurements compared to the CRONUS-P value?

Line 318 (pdf): any reason why?

Essentially, we are considering measurement of CRONUS-P as part of the analytical method (and giving a summary of the results in the methods section), instead of a result. By 'difference in the standard measurements', we take the reviewer to mean both the difference between the apparent concentration we observed for CRONUS-P during the analytical sessions described here and the accepted value, as well as the difference between the apparent concentrations for CRONUS-P observed at BGC now and in past work. First, CRONUS-P is not a primary standard and the accepted value is taken from the mean of analyses by multiple laboratories that show significant inter-lab overdispersion, so it is typical for analysis of CRONUS-P by a particular lab to differ from the accepted value. Possible reasons for these interlaboratory variations are discussed at length in Blard et al. (2015; *Quaternary Geochronology* 26, pp. 11-19) and Vermeesch et al. (2015; *Quaternary Geochronology* 26, pp. 20-28). Second, the ~3% difference in apparent CRONUS-P concentrations between this work and past published results from the BGC system, which is of similar magnitude to interlaboratory offsets, cannot be firmly attributed to a specific cause but is most likely because of calibration drift. For example, more than 5 years have elapsed between the measurements reported in Balter-Kennedy et al., and those reported in this paper, during which time the gas standard pipette used as the primary standard for these measurements has been sampled thousands of times. Maintaining the integrity of a primary gas standard for noble gas analysis over long periods is notoriously difficult because of the possibility of leakage and the challenge of making accurate depletion calculations, and this, as noted in the Blard and Vermeesch papers, highlights the value of using a mineral intercomparison standard as a secondary calibration to ensure long-term consistency of results based on a primary gas standard.

5 Results

These are the results from method 3 basically. Either integrate them into the method 3 part (similar to the other methods) and rename the section or include all results in here.

As mentioned above, we will reorganize the sections and integrate the result section into section 4: Method 3.

I would also mention the high difference between the whole rock samples as listed in Table 3. Maybe mark them in the plot of Fig. 5A, B.

Yes, we will highlight the whole rock samples in Fig. 5. Although the two whole rock chips show a slight increase in measured nuclide concentrations, they are not statistical outliers and do not introduce any when all samples are considered together. Therefore, we decide to leave any further mentioning and differentiation between the rock chip and grain samples for the discussion sections.

6 Discussion

6.1 Reproducibility

Can you explain or give an interpretation for the much lower CRONUS-P standard deviation in Fig. 6?

This is most likely just a manifestation of small-number statistics. Three aliquots of CRONUS-P were measured during the several weeks of analysis involved in the present study, whereas the CRONUS-P data set from the Balter-Kennedy study included 19 measurements over a couple of years. Of course, there is a fairly high chance that the standard deviation of a small number of measurements will be much smaller than the standard deviation of a much larger population of equivalent measurements. The true reproducibility of CRONUS-P is most likely better represented by the 2.9% scatter in the Balter-Kennedy study than by the ~1% standard deviation of the three measurements reported here, although it is possible that some of the variation in the Balter-Kennedy data arises from the fact that the data were collected in multiple analytical sessions over a period of years, which was not the case here. Regardless, these are the CRONUS-P data that were collected in the present study and we have used them without further assumptions. It should be noted that this issue does not affect any of the paper conclusions.

6.2 Inclusion of ilmenite as a trace mineral

Are there any other oxide minerals, which should be considered which retain helium? 3He concentrations are slightly lower in the magnetic separates, I would question the distinguishably.

Yes, other iron oxides such as hematite and magnetite have been found to retain helium (Baxter). However, as the reviewer points out, the deficiency in helium in the oxide-rich magnetic fractions is small and approximately consistent with the expected production rate in ilmenite, and in addition ilmenite is generally the dominant oxide mineral in Ferrar samples. Thus, there is no strong evidence that other retentive oxides are important or that accounting for their presence would provide an explanation for the observed scatter.

Can you add the data with the correction for oxides in the supplementary dataset?

The correction for oxides is calculated from the data in Table 3 and the equation is provided in the caption of Figure 7 where the results are presented. We will leave this decision to the editor.

Line 387 (PDF): any reference for this statement?

Limited evidence exists on this, but we use this paragraph to provide evidence using simple experimental design to show that ilmenite displays similar or greater helium retentivity than pyroxene, which is in agreement with similar observation by Margerison et al. (2005).

6.3 Sources of scatter in data set

Line 436-443: Maybe it's worth to conduct XRD (in the future) on a few samples (maybe with highest differences) to get information about the mineralogy of an entire sample?

Yes, one could conduct XRD for a few samples, and it would probably be useful to do if the virtual mineral separation is applied to samples other than the Ferrar dolerite. Both the physical and virtual mineral separation process of Ferrar dolerite will include impurities. The purpose here is to make the sampling process more cost and time efficient, so we focused on simplifying the workflow to the maximum extent possible. It should also be noted that although XRD is effective at identifying the presence of certain minerals, quantitative estimation of mineral fraction by XRD is often quite difficult and relies heavily on appropriate standards. It would also be difficult to conduct XRF and 3He measurements on the same aliquot.

Maybe add to a modified 'fast' virtual separation method a second or prolonged mixing to ensure that any heterogeneity is as good as it gets avoided or mixed? It will not affect the sample preparation significantly.

Essentially the main point of CT-scanning and degassing the same aliquot is so that we don't have to worry about the heterogeneity of a rock sample.

6.4 Advantages and disadvantages

Can you estimate the sample processing time for a pyroxene-separated vs virtual separated sample?

This gets a bit complicated, as it highly depends on the number of samples processed in parallel and the specific arrangement of a particular laboratory. However, general acid washing of multiple samples followed by drying and packaging takes roughly several hours of effort over two to three days. The needed time for the CT scan of a batch of samples and subsequent image processing takes roughly 3-4 hours, given that the workflow for uniquely classifying each mineral phase using blob 3D has already been established. Degassing helium from plagioclase in the vacuum oven for 8 hr at 220ºC prior to analysis in the mass spectrometer takes a day, However, besides loading and unloading the oven this process is hands free. We agree that highlighting the difference in processing time between the pyroxene-separated and virtual-separated method is an important part of this paper and we will include a discussion of the specific details and time benefits that this virtual-separation method presents.

You discuss the whole-rock method, but do not discuss the big differences in 3He between the virtual and the pyroxene separated sample, see Table 2 sample 16-ROB-52-BAS. Is the extra gas cleanup and helium purification the main issue?

The values for the measured 3He concentration for sample DD and EE listed in table 3, were incorrectly entered by mistake and will be updated to the correct values as plotted in fig. 5 and fig. 7. As observed in Fig. 6, the difference between the nuclide concentration measured for aliquote of sample 16-ROB-52-BAS is insignificant and therefore does not require any further discussion.

6.5 Implications of virtual mineral separation

Line 532: Maybe you want to add,"if the specific requirements for whole-rock chip He analysis are met, then sampling of numerous single rock chips…."

In the revised text, we will include a sentence that includes this distinction.

Line 533 (PDF): however, based on your experiments and data, up to know the whole rock analysis needs further developement and can not be used as such

The limited measurements we have from aliquots does provide evidence that we can successfully measure 3He concentration in a whole-rock chip. This is also shown in figure 6 where aliquots show internal replicability. However, as we explain in section 6.4, this comes with some challenges during the helium analysis in the mass spectrometer, where minor inclusion of hydrated minerals increases the pressure during the analysis and suppresses the 3He sensitivity. Although these issues didn't present serious obstacles for the few rock chip measurements reported here, optimizing the procedure for rock chips would most likely require additional gas and helium purification, such as attaching a liquid nitrogen cold trap or incorporating additional getters to absorb CO2 and H2O. We will make this clear in the revised manuscript.

This review included a number of technical corrections and stylistic suggestions, both here and as comments in the pdf. Some of these are a matter of journal style which should be addressed during copy-editing; we will correct or clarify all the others in the revised text or provide a response below.

Line 101: Please add the oven and with which device you captured the fractional release. You cited Bergelin et al. 2025, but please just add this information here.

Fig. 1: Please add a and b to the Figure as mentioned in the figure caption. Maybe it is useful if you also add a cumulative plot as you are referring to them in the text (At 260°C plag…released 97%..).

Table 1: Results should be placed in the result section or rename the subheadings. Add 3He to calculated conc. (atoms/gr). Source = Reference. See also paragraph Line 176 to 182.

Section 6.1 Capitalize aliquots A B C.

To summarize, proposed changes to the text in response to this review include (i) clarification in several parts of the text of the relationship between measurements previously carried out in Bergelin et al. (2025) and new measurements carried out here; (ii) minor changes to the paper organization to clearly identify the relevant results in various phases of the work; (iii) corrections to the text associated with Fig. 4; (iv) minor changes to Fig. 5 to highlight rock chip data; (v) additional discussion of the reduction in sample processing effort compared to pyroxene separation; and (vi) technical corrections and clarifications as noted above.

Review #2

We thank Reviewer 2 for a helpful review. Please see below our response and proposed changes to specific review comments (italic).

The authors take the Ferrar Dolerite as a subject of their work. It constitutes mainly two minerals as specified by the authors; plagioclase and pyroxene. What would be their "virtual separation" approach in the case of other multi-mineral rocks? Especially, in the case of other high He-retention minerals such as quartz, apatite or zircon?

We briefly discuss this in section 6.5. For other multi-mineral rocks, the approach will be similar to the one presented and outlined here in this paper. However, a key requirement is that the rock must be composed of minerals with strongly contrasting helium retentivity, in which each mineral can be uniquely accounted for, and where the degassing of helium from each mineral present can be isolated. We can clarify this further in 6.5.

In many parts of the study, there are repeated citations to Bergelin et al. (2025). It would be helpful to clearly highlight how your study differs from theirs in the introduction.

Throughout the revised text, where we refer to a previous study by Bergelin et al. (2025), we will clearly distinguish and highlight which part of the work comes from Bergelin et al. (2025) and which is introduced here as new work.

This review includes a number of technical corrections, which are grouped below. We will make the necessary corrections in the revised text.

In figure 1, plagioclase is represented as (red), and pyroxene as (blue), but in figure 2, the opposite. It would be good to have same color in both figures.

Line 133-134: There are some acronyms like ROB, LABCO, please specify.

Figure 2: You may write the set point x-y coordinates on the figure.

The resolution of Figure 3 is somewhat low, please increase it.

Figure 5: It would be better if the x-axis of these plots shows the same variables. Thus, switch the Figure-a's axis so that it shows Balter instead of Bergelin.

Figure 9 need a scale.

Table S2 has no title and information.

To summarize, proposed revisions in response to this review are (i) as discussed in the other review response, to more clearly distinguish work already done by Bergelin et al. (2025) from new work done in this study; and (ii) clarifications and technical corrections as noted above.