Response to Referee #1

This manuscript presents new live x-ray tomography 2D and 3D data from long-term injection tests of liquid CO2 into Opalinus Clay shale samples. The overall objective is to study the capacity of shale as caprock for CO2 storage under realistic pressure and temperature conditions. The study sets out to reveal potential chemo-mechanical processes and improve the understanding of localised Thermo-Hydro-Chemo-Mechanical (THCM) interactions in the shale under natural conditions.

The manuscript focuses on microstructural variations and kinematics of Opalinus Clay shale when exposed to CO2 in two different test modes, studied via 3D image analysis of real-time x-ray tomography. For long-term testing, liquid CO2 (8 MPa) was injected into samples and held under confined conditions (10 MPa) for 9 months under constant volume conditions in a PEEKcell. For the second line of testing, samples were exposed to supercritical CO2 (P = 10 MPa, T = 34°C) for up to 56 days. A combination of SEM-EDX mapping and x-ray tomography was used to analyse changes in mineralogical composition and structural evolution of the samples before, during, and after testing. The 3D volumetric response was analysed via digital volume correlation (DVC). Normalised grey level values (GV) x-ray imaging visualised and ultimately quantified CO2 penetration into the material.

Long-term injection of liquid CO2 resulted in re-arrangement of the pre-existing micro-fissures in the clay matrix and significant fissuring of calcite-rich zones that were for the first time visualised and quantified from x-ray images. Tests with exposure to supercritical CO2 showed initial swelling at pre-cracked zones and new micro-fissures in areas of direct contact to CO2. Advanced 3D image analysis showed an increasing CO2 uptake with time and potential CO2 trapping in the material.

The most significant findings include: successfully proving the increasing CO2 uptake and potential CO2 trapping in the Opalinus Clay shale. The method used as well as the results will need further work and will have importance for evaluating and monitoring the integrity and stability of shale caprocks in CO2 storage.

I found that the manuscript contains a variety of useful data that has the potential to add to what is known about CO2 storage regarding shaly rocks.

Thank you.

However, I also identify many issues that need to be addressed before the manuscript can be considered for publication in EGUsphere or any other Journal. My assessment is that, once the raised issues have been adequately addressed (if possible), the manuscript could be acceptable for publication. In my view, the extent of the changes required amounts to a major revision.

Thank you for this overall positive evaluation. We are addressing the different remarks/suggestions here below.

Specific Comments:

Issues range in significance and include: a lack of articulation of the significance of the study in regards of the 'big picture', sub-optimal structure of the manuscript (particularly the results and

discussion), and generally poor quality of writing (See section on Technical Corrections and line-by-line comments).

1 - The study sets out to reveal potential chemo-mechanical processes and improve the understanding of localised Thermo-Hydro-Chemo-Mechanical (THCM) interactions in the shale under natural conditions. **I recommend that the author elaborates more on how the results of their study fit into the bigger context, potentially by adding a sub-section to a possible Discussion section.**

Thank you for the recommendation, we have re-organised the discussion in a separate section:

5. Discussion

5.1 Long-term CO2 exposure

5.2 THMC response

5.3 Implications for geological CO2 storage

We have added a subsection (5.3) where we address possible implications to geological CO2 storage.

'**5.3 Implications for geological CO2 storage**

In this work, a series of coupled phenomena that take place in a shaly caprock material have been addressed and discussed based on qualitative and quantitative measurements of 3D x-ray tomography images. The various THMC mechanisms that have been demonstrated, are related to some extent to the equivalent testing conditions: stress state, CO2 pressure and time exposure, water saturation/dessication, mineral dissolution and precipitation. However, field conditions are different than these testing conditions. For instance, the levels of effective stress in a CO2 storage site are much higher, in the order of 10 to 20 MPa (depending on the storage depth). Consequently, the stress state of the material has an impact on the appearance of fissures. The fissures in the calcite-rich zones of the material have been visualised in an unconfined sample state (scan taken after pressure release). Similarly, dessication fissures due to water evaporation in the invading anhydrous CO2 might not manifest (at the given resolution) under elevated levels of effective stress. However, drying of the caprock due to interaction with undissolved CO2, is a phenomenon that might take place at the interface between the reservoir and the caprock due to the buoyant tendency of CO2. Dessication of the caprock can have implications that can threaten the caprock integrity and sealing capacity, for example, facilitate CO2 breakthrough. The exact CO2 breakthrough pathway in the caprock is not easy to predict because of the high micro-structural heterogeneity of the material. The connected pore space (including fissures) is supposed to drive flow and breakthrough phenomena, however, the different competing mechanisms (e.g. dessication, local effective stress modification) may result in the collapse of initially conductive pathways and the creation of new ones. There has been previous evidence of such phenomena in gas migration tests (Harrington et al. 2012; Cuss et al., 2014). The results of this study and their interpretation, demonstrate the importance of considering the different localised effects for a better understanding of the long-term response of shales in the context of geological CO2 storage. Macroscopic or averaged measurements and observations that do not take into account the micro-structural heterogeneity of shales, are limited for the development representative constitutive and numerical models.

2 - Structural changes to improve readability:

a) Separate observations from interpretations and methods. Description of results is often blended with interpretations of the data, methods applied and comparisons with published literature (which should be done more often). Please could the author revise the results so that all interpretations of the data and comparisons with published literature are moved to a new, separate discussion or new sub-chapters.

b) Long sentences with lots of sub-sections make it harder to understand the precise meaning of sentences. They also divert the attention away from the main message of a sentence and distract the reader. Please could the author consider shortening sentences in order to be more precise on what they want to say.

Thank you, we have revised the text and re-organised the methods, results and discussion more clearly in different sections. We have also tried to reduce the length of the sentences in order to make the manuscript more easily readable.

Technical Corrections:

The manuscript is riddled with major writing style idiosyncrasies and needs a lot of revision. Detailed suggestions on how the text can be improved can be found in a line-by-line comment section.

Thank you for this comment. We have tried our best to apply the suggested modifications as explained below and in the text.

The following issues and others (I recommend also consulting the author guidelines) repeatedly appear throughout the text:

Thank you for taking the time to carefully address author guidelines – we are impressed by the effort of the Referee and somewhat surprised that issues related to guidelines have not been addressed during the initial editing evaluation upon submission.

1. Avoid words/expressions like ‘unveil’, ‘embrace’, thanks to’, as they are very casual.

   Thank you for the suggestion, we have at times modified or deleted such words. Not sure though why the Referee considers expressions like ‘thanks to’ to be casual – if there is a positive impact the term ‘thanks to’ shall be used, if there is a negative impact the term ‘due to’ shall be preferred.

2. Avoid using possessive nouns (the ‘s). For example, instead of ‘the caprock’s response’, use ‘the response of the caprock material’.

   Thank you, we have eliminated apostrophes in the text.

3. Avoid ‘;’ in regular sentences, see lines 75, 187, and throughout. They are preventing the writing to be more fluid.

   Thank you, they have been modified by splitting the sentence in two or (in fewer cases) by replacing them with ‘i.e’, ‘,’ and ‘:’.

4. Commas are frequently missing or in the wrong position, see Lines 89, 91, 93 and throughout the document.

   Thank you for the comment, we have tried to better punctuate the text.

5. Italic font may be used for emphasis, although this should be used sparingly (see journal guidelines). See Lines 384, 444,...

   Removed all italics.

6. If the author's name is part of the sentence structure only the year is put in parentheses ("As we can see in the work of Prakash et al. (2022) the precipitation has increased").

   OK.

7. If the author's name is not part of the sentence, name and year are put in parentheses ("Precipitation increase was observed (Smith, 2009)") – per journal guidelines

   Corrected where needed.

8. Use metaphoric terms instead of clear objective terms ('opening the door' instead of 'possibly', 'on the other hand' instead of 'conversely')

   Thank you for the suggestion, we have tried to modify where judged appropriate or necessary.

9. Avoid incorporation of superfluous words/phrases (e.g., 'indeed', 'will be discussed in section xx', 'as discussed above')

   Thank you for the suggestion, we have tried to eliminate such parts to the maximum.

10. Avoid erroneous usage of temporal terms instead of comparative terms where no time frame is implied (e.g., 'while' instead of 'whereas', 'occasionally' or 'sometimes' instead of 'rarely' or 'less commonly', 'often' instead of 'commonly')

    Thank you for your comment, even though we do not entirely agree with the term 'erroneous' in the usage of the indicated terms, we have tried to be more literal.

11. Use precise or quantifiable language instead of vague and/or relative terms (e.g., 'relatively high/low', 'a very long time', ...)

    Thank you, we have tried to eliminate such terms.

12. As per the journal guidelines on capitalization: only the first word is capitalized in headers (in addition to proper nouns) – per journal guidelines

    Thank you, headers have been corrected.

13. From the journal guidelines: The abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence, e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that...".

Thank you, this has been corrected, as well as the citation style where needed in the text (according to the guidlines).

14. Be consistent throughout the manuscript. For example:

    - Opalinus samples, Opalinus Clay shale, Opalinus Clay samples

      Thank you, corrected 'Opalinus Clay' where missing. Did not modify 'Opalinus Clay shale' in some places in order to stretch out the fact that this material is a shale.

    - 8% and 8% (spaces must be included between number and unit, e.g., 1%, 1 m)

      Corrected.

    - In situ and in-situ (Latin phrases should not be hyphenated (e.g., "in situ", not "in-situ")

      Thank you, corrected.

Figures:

Figure 1: This figure is very self-explanatory. Caption: please use 'grey values' instead of 'grey-values'.

Corrected 'grey values'. Not sure whether the Referee uses the term 'self-explanatory' as a negative aspect of the figure and how to improve it.

Figure 3: Visually separate the columns and/or separate the headlines by a different font or boldness to make it clear at first view that the bottom sections are also from the initial / after 9 months material.

Done.

Figure 6: Please add to the caption explanations: what type of sections, what type of imaging, what the red dotted lines are marking, ... also consider adding (a), (b), (c), (d) to mark the panels.

Thank you, modified accordingly.

Figure 7: Name the columns and describe them in the figure caption. What type of imaging was used? Named panels make it easier to highlight significant features throughout the text. Add arrows to show there is a progression.

OK. Caption modified to: 'Volumetric response of the Opalinus Clay sample in time after exposure to supercritical CO_2 – left: middlde vertical slice of the x-ray CT image, right: corresponding map of volumetric strain from DVC analysis'

Figure 8: Consider naming the panels (a), (b), (c), (d), (e). An arrow or several arrows would indicate to the reader that there is a progression. Add a scale and legend. Mark the significant areas in some way to make it easier for the reader to know what you are discussing specifically.

Thank you, modified accordingly.

Figure 9: Please consider naming the rows (panels), add arrow(s), mark the most significant feature(s), and consider making the images of the second row larger, as it is difficult to see features.

Modified accordingly.

Figure 10: Overall, this figure presents a great summary of the work. I suggest making the subsets larger, as the current size makes it difficult to see what is going on.

Thank you, tried to enlarge the insets as well as possible.

Line-by-line:

Line 11: ... the long-term integrity of the caprock.

Done.

Line 12: Please be more specific, what is a 'very long time'? Compared to what?

Done.

Lines 15-16: Please revise/split this sentence for better understanding.

Done.

Line 19: ...resulted in significant fissuring ...

Done.

Line 21: 'a re-arrangement ... was observed'

Done.

Lines 24-25: The meaning of this sentence is not clear. Please revise.

Done.

Line 30: Please be more specific. Relatively high compared to what?

Done.

Line 50: Replace 'in' with 'on'

Done.

Line 53: Please specify what type of anisotropy. Mechanic, seismic, acoustic, ...? Heterogeneous in terms of what? Mineral composition, fabrics, ...?

Done.

Line 59: Consider replacing 'thanks to' with 'because of' OR: ... due to the fundamental mechanical/physical properties of the material, such as ...

replaced with 'because of' – 'due to' is related to something negative which contradicts the 'favourable properties' that motivated the usage of 'thanks to'.

Line 64: Please explain what is meant by 'representative boundaries'. Critical limitations for testing, perhaps?

As explained in the following words of the sentence, boundaries refer to the applied testing conditions such as applied pressure, temperature but also duration and size of the tested sample. Modified to 'Field-representative testing conditions' and is hopefully more clear now.

Line 66: What are 'large scale experiments?

We believe that the term 'large scale experiments' can stay without explicit precision given the introduction of this paragraph; this refers to a wide range of scales between the lab and the field, i.e. from the higher centimetric scale to a pilot scale (a few meters) scale.

Line 71: Please consider revising this sentence.

Done.

Line 72: . . . on that scale.

Done.

Line 74: . . . on the fundamental material properties . . . ; Consider what properties: i.e., physical, mechanical, . . .

Modified to 'fundamental material properties'. The kind of properties we refer to with 'fundamental' are explained after the colon ':'.

Line 75: Replace ';' with ',' and consider moving the citated authors: ... injection tests in Opalinus samples (references) do not show evidence ...; The references could be moved to the end of the sentence.

Moved citations at the end of the sentence and replaced ';' with ':' for better clarity and splitting of the sentence, *i.e.* avoid s too long sentence.

Lines 77-78: Please clarify what this statement is based on.

When the transport properties of the material are so slow, limited amount of testing duration and/or big sample sizes may be the reason why not much is observed. Waiting longer and/or testing smaller samples may lead to more interesting observations.

Line 82: This sentence could be improved by making clear in what regard the measurements not enough. Also: consider replacing 'not enough' with 'not sufficient' or something similar.

Explained: 'not sufficient in identifying chemical interactions'

Line 88: Consider replacing 'on' with 'about': ... conclusions about their impact on the structural properties ...

We have modified the sentence to be more clear 'It is thus difficult to build solid conclusions on the impact of chemical interactions on the structural properties of the material, and consequently on its transport and mechanical response'.

Line 89: Add a comma: . . . and hence, transport and mechanical response.

Modified sentence.

Line 91: Add a comma: Flow is extremely slow, resulting in . . .

Done.

Line 93: Add commas: . . . since injection pressure, and therefore effective stress, has an important . . .

Done.

Line 95: Please state what has slow transport properties and add ';' before (v) to be consistent. Done. Consider revising (v), as it does not seem to fit into the context of (i-iv). Give more details about the scale? Modified 'scale' to sample size – this last point (v) refers to the testing limitations that are related to the previous points.

Line 99: non-destructive

Done.

Lines 101-104: Commas?

Modified the structure of the sentence to be easier to read.

Line 109: Please change the headline to: . . . Analytical Principles

Modified to ‘Principles of Analysis’.

Line 117: Consider using another term instead of ’exploration’. For example: ’development’.

Thank you for the suggestion, even though we believe that the term ‘exploration’ is more representative of the reality we modified to ‘development’.

Line 126: Please consider: ... in order to improve the temporal resolution ... OR ... in order to increase the temporal resolution ...

Done.

This could be split into two sentences instead of using ()

Line 129: ...subject. The pore size...

Done.

Line 131: Be more specific, for example CO2 injection / exposure / . . .

Thank you, we have corrected this typing mistake: ‘... due to interaction with CO_2 ...’

Line 135: Please specify what type of heterogeneity and anisotropy

Sepcified: ‘mineralogical heterogeneity’ and ‘THM anisotropy’.

Line 136: Microstructures

Not sure what the comment is about – microstructure in singular is more appropriate to our understanding.

Line 141: ...to identify the heterogeneity of the specimen in 3D . . . What type of heterogeneity?

Modified to: ‘...the mineralogical heterogeneity of the specimen in 3D...’

Line 142: To be more specific you could change the beginning of this sentence: X-ray Tomography is a very ...

Thank you, modified.

Line 145: Please consider specifying what type of platform this is?

Added: ‘...platform for X-ray micro-tomography...’

Line 149: Replace ‘of’ with ‘at’

Done.

Line 150: Explain what PEEK is / stands for

Done.

Line 157: ... is placed on both ends (). OR ’Two pressure transducers are in position at the top and the bottom of the cell to monitor pressure levels during the scans.’ Sides are also left, right, front, back.

Done.

Line 158: This sentence could be improved by not using a ’;’. For example: The samples ... mm by cutting rectangular pieces with a saw, followed by ...

Done.

Line 177: ‘a pressure drop’ OR specify how it drops: successive/constant/rapid at the end/beginning/middle, . . .

Done.

Line 187: . . . are reflected . . .

Done.

Line 187: Replace ';' with '.' and start a new sentence.

Done.

Line 197: Make sure all variables are explained.

Modified to $im_{deformed}(\Phi \cdot x) = im_{refrerence}(x)$

Line 202: Please take a look at the citation style / grammar of this sentence and amend accordingly. ...

Modified to: 'Deformation based GV correction as per Stavropoulou *et al.*, (2020) is going to be applied in this work, aiming to investigate phase changes due to chemical reactions between the in-contact CO_2 with the same material'

Line 206: ... monthly ... ALSO consider revising this sentence and/or split it into two.

Removed the content of the parenthesis.

Line 215: Please check the grammar of this sentence. ... based on a single ... for the entire image ...

Corrected.

Line 226: Consider using 'Furthermore' or something similar instead of 'On the other hand'.

Done.

Line 235: Please replace 'is' with 'are'

Done.

Line 237: What is meant by 'demonstrate elements'? Show the presence of elements perhaps?

Thank you, modified.

Line 238: Consider replacing 'by means' with 'by studying'

Modified to: 'and evaluate their response based on the occurring micro-structural modifications that are identified from x-ray tomography (fissuring, swelling, self-sealing *etc.*)'

Line 243: Consider putting the evaluation at the end of the sentence / section. 'quite approximate' is vague, be more specific, for example by citing numbers.

Deleted the first part of the sentence.

Line 247: The meaning of this sentence is unclear due to grammatical and structural issues. Please revise.

Modified to: 'Even after long-term CO_2 exposure, its mineralogical composition does not vary in a significant way compared to the other three untreated samples.'

Line 248: Consider moving this before presenting the Sample A results.

Line 249: analytical

Done.

Line 252: ... Figure 2-a, using sandpaper.

Done.

Line 254: Be consistent: Figure 2-a OR Figure 2 (a).

Done.

Line 256: Consider starting the sentence like this: There are two sets of ...

Done.

Line 257: Consider rewriting this sentence, for example: 'This becomes more significant by taking into account that a second peak representing the lower GB inclusions is visible when a bilateral filter is applied (Figure 2 (b)).'

Thank you, modified accordingly.

Line 261: 'lighter' instead of 'whiter'

Modified to ''brighter'.

Line 263: The meaning of this sentence is not clear. Please revise.

We removed this sentence.

Line 268: Replace 'let' with 'held' or 'subjected to' or similar.

Done.

Line 271: Please be more specific about the significance of this! How significant was the pressure loss? What is the time scale, etc.?

We have explained: 'during the last 5 months the confining pressure has been reduced to half and the CO_2 pressure to 1 MPa'. We discuss possible implications at the end of paragraph 2 in new Section 5.1:

' Possible desaturation of the sample is not likely to explain the creation of these localised fissures, since dessication cracks in shales appear mainly within the clay matrix or at the interface of the clay matrix with other inclusions (calcite, pyrite etc.). Fissuring in the clay matrix is however not observed even after total pressure release (unconfined conditions). On the contrary, the number of pre-existing fissures initially in the sample is reduced. This is additionally ensuring for the potential impact of progressive pressure loss during these 9 months of exposure. Pressure loss may result in desaturation and fissuring of the sample (usually parallel to the bedding orientation) that have not been observed at the given resolution of this study.'

Line 271: Why is there no significant impact? What leads to this statement? If this is based on the results from this study, it should be moved to the discussion.

Please see our previous reply.

Line 282: Please clarify: is the explanation self-sealing followed by precipitation or are both explanations independent from each other?

We have moved this part to the new Discussion section, where hydromechanical self-sealing and self-sealing due to mineral precipitation are more clearly dissociated.

'Fissure closure can be explained by means of hydromechanical self-sealing behaviour of the material under long-term confinement. The self-sealing response of shales is one of the main properties for which this material is studied as a potential sealing material in a broader context of underground storage, such as radioactive waste, CO2 or hydrogen storage (Bossart et al., 2019; Di Donna et al., 2022, Yu et al., 2022). Hou et al., (2022) discussed the self-sealing response of caprock materials in terms of mineral precipitation. They showed that in illite-rich shale precipitation took place in quartz, i.e. Si-rich zones. In the current study, Si-rich zones are unfortunately not distinguishable from the either SEM or x-ray images. Prakash et al., (2022) pointed out a more pronounced precipitation activ-

ity in zones parallel to the direction of the bedding plane. This is in line with the identified orientation of fissures in the sample before and after confinement and CO2 exposure. The pre-existing fissures that were parallel to the bedding orientation disappear, and the fewer fissures after long-term confinement and CO2 exposure are in their majority no longer parallel to the bedding. Other works have shown that incorporation of supercritical CO2 in micro-structural interlayers can induce the beneficial swelling of smectitic clays (Alemu et al., 2011; Busch et al., 2016).'

Line 284: See comment Line 202: a better start of this sentence would be: Prakash et al. (2022) ...

Done.

Line 295: Elaborate – what is the basis for this statement?

Segmentation in a material like shale that is composed by many different phases (not like concrete for example where there are 3 main phases: aggregates, cement/sand and pores), is very sensitive to the user. Accurate image segmentation is an entire research filed by itseld and since more recently is apporached with very sophisticated methods including neural networks and machine learning algorithms. So here we wanted to indirectly acknowledge that the precision of segmentation has not been a topic where we invested our effort to use complex and sophisticated tools for the analysis of our images, but a standard thresholding approach.
We have removed this sentence and rearranged the previous ones to avoid confusion.

Line 297: minimum instead of min.

Done.

Line 309: Please clarify – does this refer to the initial sample?

Yes, modified to 'The initial shape of the 3D histogram' and it is is hopefully more clear.

Line 337: Citation style: These results confirm the findings of Minardi et al. (2021) from carbonate rich Opalinus Clay shale.

OK.

Line 338: Please clarify if these are results from this study or from Minardi et al. (2021).

Modified to: 'These results confirm the findings of Minardi et al. (2021) on carbonate rich Opalinus Clay shale that identified a bimodal pore size distribution, with a second dominant pore size between 50-100 μm corresponding to the interface of carbonate/clay particles.

Line 349: ... are applied. CO2 is introduced into the PEEKcell when the P-T conditions are stable.

Modified to: 'CO_2 is then introduced in the cell in direct contact with the sample. The target pressure and temperature are applied and maintained stable over a period of 56 days.'

Line 351: ... 56 days (scan 03) of CO2 exposure. A final scan is performed after the release of pressure and temperature (scan 04).

Modified, thank you.

Line 359: These ... with an initial maximum aperture of ...

Done.

Line 362: Name the middle slice in the figure to make it easier for the reader to connect what you

describe to what it is you see. In this instance, it is not clear at once what part of the figure is referred to.

Added in Figure 7.

Line 376:. A distinct pattern is absent in the rest of the material, but the calculated volumetric ...

OK.

Line 384: Consider regular, not italic

Done.

Line 387: ... suggests that the sample is not completely saturated.

Done.

Line 390: ... at full saturation, which means a decrease of free water and increase in CO2. Is this correct? Please be more specific here

Line 395: ... has been discussed little in the . . .

Done.

Line 395: Add references

Line 403: Whose calculation?

Modified: '...the volumetric evolution of the fissures in the different scans is calculated and presented in Figure. For this calculation...'

Line 410: Please elaborate what mechanisms may be active and discuss why these mechanisms.

Done.

Line 417: Consider deleting the first sentence of this section. Start with: Analysis of the evolution of GV from x-ray images after correction for volumetric strain (Stavropoulou et al., 2020) is used to visualise and quantify the CO2 penetration into the material.

Thank you for the suggestion, modified accordingly.

Line 420: Consider using 'difficult' instead of 'ambitious'.

Changed to 'challenging'

OR say something like the method is not particularly sensitive for the small scale of density variations due to supercritical CO2 invasion. Also, give details why this statement is made. Are there other studies that show the 'slight density variations'? Are these results from this study? Give more details.

When sc-CO2 invades the open porosity sample, the density of the sample is not going to change in a striking way. That would be the case if we were injecting a dense fluid, e.g. Mercury. This is where the challenge comes from.

Line 423: This whole section describes a method and should be moved. Thank you moved to the end of section 2.2.

Line 430: After Stavropoulou et al. (2020) the attenuation ...

Done.

Line 432: Explain $\Delta\mu$ as well.

Done.

Line 439: Move the explanation of the legend to the figure caption.

OK.

Line 444: Consider using regular instead of italic

Done.

Line 448: Please clarify: ... around and in crack locations or either around or in crack locations.

Done.

Line 451: ... fissures. Therefore, the density will always be decreased in the direct vicinity of the fissures.

Done.

Line 452: Please consider: ... of the material increases homogeneously ... otherwise words like 'eventually' and 'relatively' are vague terms that make the statement highly speculative.

Done.

Line 461: 'the' instead of 'their'

Done.

Line 465: For this work... Please mention briefly why the interaction (...) was studied.

Line 476: ... of a shaly material ... OR ... of Opalinus Clay shale

Done.

Line 477: What are those conditions? Give numbers.

Done.

Line 478: ... that develop fissures after 9 months of exposure.

Done.

Line 486: ... as shown in Stavropoulou and Laloui (2022) but do not re-appear 9 months later, after pressure release.

Done.

Line 488: Capacity; ..., the x-ray scan after 9 months of CO2 exposure shows new micro-fissures in the ...

Done.

Line 498: It is significant that after CO2 release....

Done.

Line 501: Consider replacing 'sums up' with 'highlights'.

Done.

Line 504: Non-destructive

Done.

Line 506: Please consider naming the phenomena here to underline the significance of the contribution.

These phenomena include the volumetric reponse and water evaporation during exposure to supercritical CO_2, the localised chemo-mechanical interactions in calcite-rich zones, the CO_2 uptake and the role of micro-fissures in the material, and the volumetric response upon CO_2 breakthrough.

Added references:

Bossart, P. & Thury, M.: Characteristics of the Opalinus clay at Mont Terri, Reports of the Swiss Geological Survey 3, 2011.

Busch, A., Bertier, P., Gensterblum, Y., Rother, G., Spiers, C. J., Zhang, M., & Wentinck, H. M.: On sorption and swelling of CO2 in clays. Geomechanics and Geophysics for Geo-energy and Geo-resources, 2(2), 111-130, https://doi.org/10.1007/s40948-016-0024-4, 2016.

Cui, G., Zhu, L., Zhou, Q., Ren, S., & Wang, J.: Geochemical reactions and their effect on CO2 storage efficiency during the whole process of CO2 EOR and subsequent storage. International Journal of Greenhouse Gas Control, 108, 103335, https://doi.org/10.1016/j.ijggc.2021.103335, 2021.

Cuss, R., Harrington, J., Giot, R., & Auvray, C. : Experimental observations of mechanical dilation at the onset of gas flow in Callovo-Oxfordian claystone. Geological Society, London, Special Publications, 400(1), 507-519, https://doi.org/10.1144/SP400.26, 2014.

De Jong, S. M., Spiers, C. J., & Busch, A.: Development of swelling strain in smectite clays through exposure to carbon dioxide. International Journal of Greenhouse Gas Control, 24, 149-161, https://doi.org/10.1016/j.ijggc.2014.03.010, 2014.

Di Donna, A., Charrier, P., Dijkstra, J., Andò, E., & Bésuelle, P. (2022). The contribution of swelling to self-sealing of claystone studied through x-ray tomography. Physics and Chemistry of the Earth, Parts A/B/C, 127, 103191.

Espinoza, D. N., & Santamarina, J. C.: Water-CO2-mineral systems: Interfacial tension, contact angle, and diffusion – Implications to CO2 geological storage. Water resources research, 46(7), https://doi.org/10.1029/2009WR008634, 2010.

Harrington, J. F., Milodowski, A. E., Graham, C. C., Rushton, J. C., & Cuss, R. J.: Evidence for gas-induced pathways in clay using a nanoparticle injection technique. Mineralogical Magazine, 76(8), 3327-3336, Harrington, 2012.

Hou, L., Yu, Z., Luo, X., & Wu, S.: Self-sealing of caprocks during CO2 geological sequestration. Energy, 252, 124064, https://doi.org/10.1016/j.energy.2022.124064, 2022.

Michels, L., Fossum, J. O., Rozynek, Z., Hemmen, H., Rustenberg, K., Sobas, P. A., Kalantzopoulos, G. N., Knudsen, K. D., Janek, M., Plivelic, T.S., & da Silva, G. J.: Intercalation and retention of carbon dioxide in a smectite clay promoted by interlayer cations. Scientific reports, 5(1), 1-9, https://doi.org/10.1038/srep08775, 2015.

Miri, R., & Hellevang, H.: Salt precipitation during CO2 storage – A review. International Journal of Greenhouse Gas Control, 51, 136-147, https://doi.org/10.1016/j.ijggc.2016.05.015, 2016.

Schaef, H. T., Ilton, E. S., Qafoku, O., Martin, P. F., Felmy, A. R., & Rosso, K. M.: In situ XRD study of Ca2+ saturated montmorillonite (STX-1) exposed to anhydrous and wet supercritical carbon dioxide. International Journal of Greenhouse Gas Control, 6, 220-229, https://doi.org/10.1016/j.ijggc.2011.11.001 2012.

Yu, H., Zhang, Y., Lebedev, M., Li, X., Wang, Z., Verrall, M., Squelch, A., & Iglauer, S.: Swelling-induced self-sealing mechanism in fractured caprock: implications for carbon geosequestration. AAPG Bulletin, (20,221,001), https://doi.org/10.1306/09232219136, 2022.

Zhang, M., de Jong, S. M., Spiers, C. J., Busch, A., & Wentinck, H. M.: Swelling stress development in confined smectite clays through exposure to CO2. International Journal of Greenhouse Gas Control, 74, 49-61, https://doi.org/10.1016/j.ijggc.2018.04.014, 2018.