Response to reviewers

Reviewer Comments to Author:  
  
Reviewer: 1  
  
Comments to the Author  
Manuscript ID: JPET-Apr-24-0074  
  
Title: Depths in a day - A new era of rapid-response Raman-based barometry using fluid-inclusions  
Authors: Charlotte L. DeVitre, Penny E. Wieser, Alex Bearden, Raela Richie, Berenise Rangel, Matthew Gleeson, John Grimsich, Kendra Lynn, Drew Down, Natalia Deligne and Katie Mullike  
  
General comments:  
This manuscript reports the application of Raman-based fluid inclusion geobarometry to fluid inclusions in olivine erupted from recent Kilauea volcano activity. The authors successfully estimated the magma chamber depth rapidly. The primary conclusion is that this method provides estimations of magma chamber depth in a relatively short time compared to geobarometers based on the major element chemistry of minerals or the volatile composition of melt inclusions. However, since the authors did not develop innovative technology that allows for such rapid measurement, these results may be modest for publication in the Journal of Petrology.

We thank the reviewer for their thorough work. While we did not develop technology in this contribution, we did develop the tools necessary as well as demonstrated the reliability of the technique in three previous contributions (DeVitre et al. 2021, Wieser and DeVitre (2024) in J.Volcanica, and DeVitre and Wieser (2024) in Geochemical Perspective Letters). We emphasize that the purpose of this study was to demonstrate through a rigorous simulation in a low-risk scenario that the method is now viable and fast enough to be used as a monitoring tool. This had not been demonstrated previously. These were also the reasons why we originally chose a Letter format. In this contribution, we developed and optimized the workflow which allow for extremely fast results to be obtained (e.g. use of production lines, recommendations for speeding up various parts of the process).

Specific comments:  
L25-29: This sentence does not specify which concerns are addressed by this study. As a result, readers cannot discern what specific issues were resolved. Please elaborate on the specific concerns that were mitigated by your research.

We agree with the reviewer and have changed the abstract to include more details on what concerns were addressed by this contribution. The abstract now reads as follows: “Rapid-response petrological monitoring is a major advance for volcano observatories, allowing them to build and validate models of the plumbing systems that supply eruptions in near-real-time. The depth of magma storage has recently been identified as high-priority information for volcanic observatories, yet this information is not currently obtainable via petrological monitoring methods on timescales relevant to eruption response. **Fluid inclusion barometry (using micro-thermometry or Raman spectroscopy) is a well-established petrological method to estimate magma storage depths and has been proposed to have potential as a rapid-response monitoring tool, although this has not been formally demonstrated. To address this deficiency, we performed a near-real-time rapid-response simulation for the September 2023 eruption of Kīlauea. We show that Raman-based fluid inclusion barometry can robustly determine reservoir depths within a day of receiving samples** - a transformative timescale that has not previously been achieved by petrological methods. Fluid inclusion barometry using microthermometric techniques has typically been limited to systems with relatively deep magma storage (>~ 0.4 g/cm3 or ~7 km) where measurements of CO2 density are easy and accurate given that the CO2 fluid homogenizes into the liquid phase. Improvements of the accuracy of Raman spectroscopy measurements of fluids with low CO2 density over the past couple of decades has enabled measurements of fluid inclusions from shallower storage systems. **However, one caveat of examining shallower systems is that the fraction of H2O in the fluid may be too high to reliably convert CO2 density to pressure. To test the global applicability of rapid response fluid inclusion barometry, we compiled a global melt inclusion dataset (>4000 samples) for which we calculated fluid composition at the point of vapour saturation (XH2O). We show that fluid inclusions in crystal-hosts from mafic compositions (<57 wt% SiO2) – likely representative of recharge magmas worldwide – may trap fluids with XH2O low enough to make fluid inclusion barometry useful at many of the world’s most active and hazardous mafic volcanic systems (e.g. Iceland, Hawai’i, Galápagos, East African Rift, Réunion, Canary Islands, Azores, Cabo Verde).”**

L51-52: After thoroughly reviewing Re et al. (2021), I could not find any mention of storage depth estimation taking several months to years using melt inclusion geobarometry. Could you specify the source of this information?  
While it is true that Re et al., (2021) do not report specific numbers, they do report MI barometry as a slow method (MI-volatiles; which implies barometric measurements using volatile solubility modelling). These timelines are from our own extensive experience as melt inclusionists. DeVitre was part of a review of melt inclusion best practices (Rose-Koga et al., 2021) and is well connected with the community and knows of no one who has performed results in less than a year. Her own recently published a melt inclusion paper in PNAS took 2 years to finish. Similarly, Wieser has been involved in 5 published melt inclusion studies (Wieser et al. 2021, 2022, Wong et al. 2023, Dayton et al. 2024, Van Gevre et al. 2024), none of which took less than a year. For example, for the two Kilauea papers, Wieser et al. 2021 was submitted 7th Aug, 2020, despite samples being collected in Aug 2018. This was how long it took to perform Raman analyses, book and do SIMS time in 2 sessions (because SIMS time is hard to get), collect EPMA data afterwards, perform PEC corrections, and solubility modelling. Equally, Lerner et al. was submitted 8th Oct, 2020. These two groups were also in competition working on the same samples,

Certainly, if one had unlimited access to a Raman, an SEM/EPMA and a SIMS/FTIR, perhaps it could be done in a few weeks on a set of ~20 melt inclusions. But unlimited access to a SIMS with suitable standards and well-defined workflows is highly unlikely outside of a very small number of institutions. From our extensive experience, the preparation and analytical complexity of MI, and the sheer number of analytical techniques required, is simply incomparable to FI.

In reality no one has done a simulation in near-real time with Melt inclusion work, but we think that any melt inclusionist could agree that it is impossible that one could obtain barometric data from ~20 melt inclusions in 1 day and ~46 melt inclusions in 2 days.

In any case, we’ve added clarification to the text and removed the reference to “years” (see screenshot).

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L51-52: While I agree that estimating storage depths based on volatile content analysis takes longer than Raman-based FI geobarometry, suggesting it takes several months to years seems exaggerated. If a team with access to SIMS and Raman spectroscopy equipment prioritizes sample preparation and analysis, the process could be completed within one week to one month.  
We agree that it may be possible to conduct H2O-CO2 analyses in 1 month at Woods Hole, which has a Raman spectrometer and a SIMS. However, this is extremely unusual; there are very few SIMS facilities worldwide, and they are normally booked up well in advance. Given how many people travel to use these facilities, we think it is highly unlikely the staff scientists would re-arrange other users time, resulting in loss of flight and accommodation money. We enquired in May about use of SIMS for melt inclusion work, we received no response from several facilities, WHOI had next availability in September 2024 and EIMF (UK) in December 2024. Even at WHOI, there is no SEM-EPMA for obtaining glass chemistry required to perform PEC corrections and calculate saturation pressures. One could argue that instead of SIMS, FTIR could be conducted. However, FTIR preparation on single MI is much more time consuming/difficult than SIMS and requires fairly large melt inclusions.

In any case, we agree that perhaps the yearly reference is too long in the context of the simulation. It’s hard to compare what would happen if such a simulation was done with Melt inclusion work, but it is highly doubtful that barometric data from ~16 MI in one day, ~46 in two and~59 in three could be obtained or even in one week. However, if the reviewer feels strongly about this, we encourage them to conduct such a simulation, we are very happy to be proved wrong! We’ve reduced the statement to “often taking months to complete”

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L51-58: CO2 fluid inclusion geobarometry using microthermometry has been applied to peridotite and phenocrysts since at least the 1980s (e.g., Andersen et al., 1995; Belkin et al., 1985; Hansteen et al., 1998, 1991; Klügel et al., 2020, 2005; Roedder, 1983) for estimating magma plumbing system structures. However, this study appears to overlook the findings of these prior works. Microthermometric density measurements are more precise than those obtained by Raman spectroscopy when CO2 densities exceed approximately 0.6 g/cm³ (Bakker, 2021; Kobayashi et al., 2012). Additionally, analyzing several tens of fluid inclusions can be completed within a timeframe similar to that of Raman-based density measurements, typically ranging from one day to one week. Therefore, a review of previous studies on fluid inclusion geobarometry using microthermometry and a clear distinction between this method and Raman spectroscopy-based methods should be provided.  
We have added a bit more details on both techniques, appropriate references as requested by the reviewer and expanded a bit on the difference between the two methods now that we are not limited to the letter word count. However we note that more detail was provided in a previous contribution (Devitre and Wieser, 2024 in GPL) and the purpose of this paper is not to compare the techniques but rather to present a near real time simulation, a stress test of the Raman method.

See lines 109-182 “ Fluid inclusion barometry via micro-thermometry and Raman spectroscopy is a well-established technique that has regularly been applied to determine the structure of certain magma plumbing systems over the past four decades (e.g., Roedder and Bodnar, 1980; Roedder, 1983, 1984; Belkin *et al.*, 1985; Hansteen *et al.*, 1991; Andersen *et al.*, 1995; Hansteen *et al.*, 1998; Yamamoto *et al.*, 2002; Frezzotti *et al.*, 2003; Klügel *et al.*, 2005; Yamamoto *et al.*, 2007; Bali *et al.*, 2008; Hansteen and Klügel, 2008; Ladenberger *et al.*, 2009; Hildner *et al.*, 2011, 2012; Zanon and Frezzotti, 2013; Levresse *et al.*, 2016; Klügel *et al.*, 2020; Boudoire *et al.*, 2023; Dayton *et al.*, 2023; Zanon *et al.*, 2024a). Micro-thermometry – which consists of measuring the temperature at which phase changes occur in a fluid under a microscope – can determine the CO2 density of fluid inclusions with bulk densities above critical (>~0.45 g/cm3) with great accuracy (Hansteen and Klügel, 2008; Bakker, 2021). However, it is difficult to observe the evaporation of a thin liquid film in fluid inclusions that homogenize to the vapour phase (Hansteen and Klügel, 2008) and therefore the technique is limited in its applicability to estimate the shallow structure of magmatic plumbing systems (<5-7 km), except perhaps in the case of large inclusions in clear host phases such as quartz (e.g., Clocchiatti *et al.*, 1994; Zanon *et al.*, 2003). Developments in the past two decades in the accuracy and precision of Raman spectroscopy measurements of CO2 density, which uses spectral features of CO2 fluids to calculate a CO2 density using an instrument-specific calibration (e.g., Rosso and Bodnar, 1995; Kawakami *et al.*, 2003; Yamamoto and Kagi, 2006; Fall *et al.*, 2011; Wang *et al.*, 2011, 2019; Lamadrid *et al.*, 2017; Sublett *et al.*, 2020; DeVitre *et al.*, 2021; Le *et al.*, 2021), have made it possible to accurately measure lower CO2 densities, and thus identify shallow storage regions more reliably. Recent work demonstrated that Raman-based barometry of CO2-rich fluid inclusions returns the same results as melt inclusion barometry and provide much smaller uncertainties than mineral barometry, while requiring far less time and resources than melt inclusion analyses (Dayton *et al.*, 2024; DeVitre and Wieser, 2024; Lerner *et al.*, 2024). Although micro thermometry is generally more accurate than Raman spectroscopy for high-density CO2 fluids (>0.6 g/cm3; Bakker, 2021), Raman spectroscopy has the advantage of being able to easily probe nearly the entire range of geologically relevant CO2 densities (from very low to high) with reasonably consistent precision and accuracy. It is also possible to assess at the same time the composition of the fluids and/or solids in the inclusions using Raman spectroscopy. The CO2 density obtained from either technique, along with an estimate of entrapment temperature, is converted into an entrapment pressure using an equation of State (EOS, Fig. 2, either pure CO2, or CO2-H2O). “

L59-62: The manuscript describes Raman-based fluid inclusion geobarometry as if it were a recently developed technique. Consequently, many prior studies that do not fit this narrative are ignored. In reality, Raman-based fluid inclusion geobarometry has been applied to phenocrysts and peridotite for understanding magma plumbing system structures for quite some time (e.g., Bali et al., 2008; Boudoire et al., 2018; Ladenberger et al., 2009; Levresse et al., 2016; Yamamoto et al., 2014, 2007, 2002). The results of prior studies on Raman-based fluid inclusion geobarometry should also be accurately described.  
We agree with the reviewer and reformulated this to clearly show that the method itself is not recent or novel, but that recent developments have made it possible to consider it a potential monitoring technique. We have added citations as requested by the reviewer. However, we would like to point out to the editor that many of these studies did not calibrate their Raman instrument (e.g. Bali et al. 2018), and likely suffered from unquantifiable laser heating issues (see Wieser et al. 2024, treatise of Geochemistry for discussion).

See lines 109-186 as pasted in previous comment. “ Fluid inclusion barometry via micro-thermometry and Raman spectroscopy is a well-established technique that has regularly been applied to determine the structure of certain magma plumbing systems over the past four decades (e.g., Roedder and Bodnar, 1980; Roedder, 1983, 1984; Belkin *et al.*, 1985; Hansteen *et al.*, 1991; Andersen *et al.*, 1995; Hansteen *et al.*, 1998; Yamamoto *et al.*, 2002; Frezzotti *et al.*, 2003; Klügel *et al.*, 2005; Yamamoto *et al.*, 2007; Bali *et al.*, 2008; Hansteen and Klügel, 2008; Ladenberger *et al.*, 2009; Hildner *et al.*, 2011, 2012; Zanon and Frezzotti, 2013; Levresse *et al.*, 2016; Klügel *et al.*, 2020; Boudoire *et al.*, 2023; Dayton *et al.*, 2023; Zanon *et al.*, 2024a). Micro-thermometry – which consists of measuring the temperature at which phase changes occur in a fluid under a microscope – can determine the CO2 density of fluid inclusions with bulk densities above critical (>~0.45 g/cm3) with great accuracy (Hansteen and Klügel, 2008; Bakker, 2021). However, it is difficult to observe the evaporation of a thin liquid film in fluid inclusions that homogenize to the vapour phase (Hansteen and Klügel, 2008) and therefore the technique is limited in its applicability to estimate the shallow structure of magmatic plumbing systems (<5-7 km), except perhaps in the case of large inclusions in clear host phases such as quartz (e.g., Clocchiatti *et al.*, 1994; Zanon *et al.*, 2003). Developments in the past two decades in the accuracy and precision of Raman spectroscopy measurements of CO2 density, which uses spectral features of CO2 fluids to calculate a CO2 density using an instrument-specific calibration (e.g., Rosso and Bodnar, 1995; Kawakami *et al.*, 2003; Yamamoto and Kagi, 2006; Fall *et al.*, 2011; Wang *et al.*, 2011, 2019; Lamadrid *et al.*, 2017; Sublett *et al.*, 2020; DeVitre *et al.*, 2021; Le *et al.*, 2021), have made it possible to accurately measure lower CO2 densities, and thus identify shallow storage regions more reliably. Recent work demonstrated that Raman-based barometry of CO2-rich fluid inclusions returns the same results as melt inclusion barometry and provide much smaller uncertainties than mineral barometry, while requiring far less time and resources than melt inclusion analyses (Dayton *et al.*, 2024; DeVitre and Wieser, 2024; Lerner *et al.*, 2024). Although micro thermometry is generally more accurate than Raman spectroscopy for high-density CO2 fluids (>0.6 g/cm3; Bakker, 2021), Raman spectroscopy has the advantage of being able to easily probe nearly the entire range of geologically relevant CO2 densities (from very low to high) with reasonably consistent precision and accuracy. It is also possible to assess at the same time the composition of the fluids and/or solids in the inclusions using Raman spectroscopy. The CO2 density obtained from either technique, along with an estimate of entrapment temperature, is converted into an entrapment pressure using an equation of State (EOS, Fig. 2, either pure CO2, or CO2-H2O).

Recent studies have speculated that fluid inclusion barometry, using either micro thermometry or Raman spectroscopy, could be performed quickly enough to be useful for near-real-time volcano monitoring (Dayton *et al.*, 2023; Zanon *et al.*, 2024b)

L59-62: This statement is incorrect, as geobarometry using the density of CO2-rich fluid inclusions measured by Raman spectroscopy has been in use for a long time (Bali et al., 2008; Boudoire et al., 2018; Ladenberger et al., 2009; Levresse et al., 2016; Yamamoto et al., 2002). While analytical and data processing methods have indeed become more sophisticated over the last 20 years (Bakker, 2021; Fall et al., 2011; Hagiwara et al., 2021, 2020; Kawakami et al., 2003; Lamadrid et al., 2017; Le et al., 2020, 2019; Remigi et al., 2021; Rosso and Bodnar, 1995; Song et al., 2009; Sublett et al., 2020a, 2020b; Wang et al., 2019, 2011; Yamamoto and Kagi, 2006; Yuan et al., 2017), the time required for analysis has changed very little.  
We agree with the reviewer that the technique has been in use a long time and that the analytical time has not changed. What has changed is the accuracy of the measurements, owing to improvements in calibrations, hardware and software, and much more recently the capability to process data in a more streamlined, reproduceable way (e.g., Wieser and Devitre 2024 Volcanica). We have reformulated to indicate that while the technique is not novel, all of these developments over the past two decades are what make possible to even consider this a potential monitoring technique.

See lines 183-192:

“Recent studies have speculated that fluid inclusion barometry, using either micro thermometry or Raman spectroscopy, could be performed quickly enough to be useful for near-real-time volcano monitoring (Dayton *et al.*, 2023; Zanon *et al.*, 2024b). Indeed, advances in the accuracy of Raman measurements from two decades of improvements in CO2 densimetry calibrations along with new capabilities to process data in a more streamlined and reproduceable way (e.g., Wieser and DeVitre, 2024) show that Raman-based fluid inclusion barometry could now be fast-enough for this application. However, this has not been formally demonstrated.”

L59-67: The advantages of Raman-based fluid inclusion geobarometry are correctly described, but several drawbacks are not mentioned, which may give readers a misleading impression. Therefore, the following major drawbacks of this method should also be properly described: 1) Geobarometry using the residual pressure of fluid inclusions can only be used in volcanic ejecta where fluid inclusions are present in phenocrysts. Thus, although this method has been used to investigate the structure of magma plumbing systems since at least the 1980s and has been the subject of much research, it has only been applied to specific MORB and OIB volcanoes. 2) The density of CO2 inclusions in olivine is known to significantly decrease due to the plastic deformation of host minerals, fluid diffusion, and fluid-host reaction during transport by magma, compared to that in pyroxene and spinel (e.g., De Vivo et al., 1990; Oglialoro et al., 2017; Scambelluri et al., 2009; Viti and Frezzotti, 2000; Wanamaker and Evans, 1989; Yamamoto et al., 2012). Therefore, the pressure of magma chambers estimated from the density of CO2 fluid may not always reflect the true pressure of the magma chambers for all volcanic eruptions. Even fluid inclusions that have not undergone decrepitation may have unreliable densities. The amount of density reduction due to plastic deformation and fluid diffusion depends on factors such as the host mineral species, the depth of the magma chamber, the ascent rate of the phenocrysts, the temperature of the magma, and the cooling rate at the surface, making simple interpretations impossible. Therefore, it is dangerous to describe fluid inclusion geobarometry as a reliable method without discussing the various uncertainties mentioned above. For example, I cannot agree with using the results of this method to make decisions on whether to evacuate residents. Thus, the authors should describe the applicability and limitations of this method more conservatively.

We agree with the reviewer that a discussion of drawbacks of the method is important and therefore we have added an entire section before the broad applicability discussion where we describe them. See section 3. Advantages and limitations of fluid inclusion barometry. At line 560.

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We also refer the reader to Devitre and Wieser 2024 GPL, where more detail is provided, including a python implementation of the model of Wanamaker and Evans 1989 for plastic deformation. We discuss there that in the case of Kilauea, for example, the effect is minimal and within the uncertainty of the method. Regardless, we agree this effect can be much more important in other settings and thus is briefly discussed here though the reliability of the method is not the topic of this paper.

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Reviewer: 2  
  
Comments to the Author  
See attached.

Review of “Depths in a day – A new era of rapid-response Raman-based barometry using fluid inlusions” by DeVitre et al.

Dear editors and authors

Please accept my apologies for the somewhat delayed review of “Depths in a day”. The review request landed exactly on exam and dissertation marking season, which unfortunately had to take priority. The authors of the manuscript allowed themselves some remarks about the bottlenecks in academic publishing and I am sorry to have caused yet another hold-up in this manuscript’s journey through the review process.

I read this manuscript with great interest. A notable feature of melt and fluid inclusion barometry papers over the past few years has been the hopeful promise of developing these approaches for real-time application during volcanic crises, but until now I’m not aware of any study that has actually attempted this. It was refreshing to read a manuscript that has actually been able to demonstrate a proof-of-concept that near-real-time petrological monitoring using fluid inclusions is a viable route towards providing information on magma storage depths to volcano observatories. I agree with the original reviewers that the approach itself is not novel; however, in my opinion there is great scientific value in having a published account of this first demonstration that magma storage pressures can be measured and communicated in near-real time. The readership of JPet will include many scientists using melt and fluid inclusion barometry approaches in their own research, so a JPet letter demonstrating a case study of rapid-response fluid inclusion barometry will reach an interested audience. The letter is well written, clearly explained, and reaches beyond the immediate case study of the September 2023 Kilauea eruption into a well-articulated discussion of the types of volcano and volcanic setting where rapid-response barometry has genuine potential as a near-real-time monitoring tool. I recommend to publish the letter with minor revisions.

We thank the reviewer for their support, and appreciating why we have performed such a simulation.

In my view the authors have done an excellent job responding to the comments of the original reviewers. Specifically, they have explained and quantified the uncertainties in their temperature estimates; they have addressed concerns regarding EOS for pure CO2 fluid vs a mixed H2O-CO2 fluid, and the effect of variable XH2O on the calculated pressures; justified their choice of Raman densimeter; and justified the types of images used in their workflow. I would have appreciated specific direction to the changed text in the revised manuscript where these concerns were addressed.

I thought the authors did not quite address the concern of Reviewer 1 “How are the data used by decision makers” – and this is also in my mind whenever rapid-response petrological monitoring is discussed. I agree with lines 81-84 (specific recommendations of CONVERSE) that assessing magma storage depth is a key science question in an unfolding eruption, but it’s less clear how observatories would actually use that information to inform the hazard response and management. Perhaps it is as simple as being able to maintain good relationships with local communities by being able to answer questions on where the magma is coming from; how long it may have been stored for; what is its pre-eruptive temperature and viscosity. But are there other ways in which real-time barometry informs ongoing hazard assessments or changes to the crisis response? The case study presented was a low-hazard event so perhaps there are no examples for this particular event, but the discussion or conclusion could usefully include a couple of sentences explaining specifically how the HVO envisages that near-real-time fluid inclusion barometry would feed into their workflows and decisions in a more hazardous scenario.

We agree with the reviewer and have expanded on the uses of the method specifically from the observatories’ perspective.

See lines 945-995.

“Importantly, this simulation showed that rapid-response work in collaboration with universities was not taxing on observatory or academic staff, particularly considering the usefulness of information provided. This means this methodology can be employed during future eruptions to help observatories deduce the geometry of the plumbing system supplying magma, adding a crucial information for interpreting activity (Re *et al.*, 2021), without detracting from other essential duties during eruption responses. For example, during the 2018 LERZ Kīlauea eruption, HVO’s near-real-time chemical monitoring with bulk rock ED-XRF identified the appearance and disappearance of many magma batches (Gansecki *et al.*, 2019). Fluid inclusion barometry could have linked these distinct chemical signatures to different storage regions, addressing the questions of scientists and residents alike. Similarly, the return of eruptive activity at Kīlauea in 2020 was accompanied by many questions about how the magmatic plumbing system had changed following the summit collapse in 2018 (Lynn *et al.*, 2024). Fluid inclusion barometry would have allowed been a critical addition to understanding the eruption and the system.

The case study presented here intentionally targeted a low-hazard eruption as a testbed for implementing near-real-time fluid inclusion barometry as a monitoring tool. This type of work, on low-hazard, short-lived eruptions, has a few advantages. First, it allows observatories to iron-out bottlenecks to implement the tool for future events. Second, it enables researchers to establish baselines that can be used for routine monitoring, which is fundamental to notice changes and trends in systems as volcanoes like Kīlauea continue their eruptive activity and evolve over human perceptible timescales. Although this eruption was short-lived, one can also imagine how such data could be useful in the context of a much longer eruption (e.g., Kīlauea 2018 LERZ). For instance, if FI barometry indicated depths of ≥5km within 48 hours, this data would be used by HVO to inform discussions with partners about the potential for a deeper source than seen in previous events and therefore an expectation for greater volume, increased duration, or potential waxing and waning if magma transport involves shallow sources along the way. In terms of regular near-real-time monitoring of a longer-lived eruption, FI barometric data could aid in understanding pre-eruptive deformation and unrest. Specifically, such data could be used to identify the reservoir being tapped by the eruption and how associated stresses could change. In the case of Kīlauea, if pressurization was observed in both the South Caldera and the Halema‘uma‘u reservoirs, but the eruption primarily taps Halema‘uma‘u, this could mean that the South Caldera reservoir could be primed to support eruption. This can change the expectations for developing activity at the volcano.

”

I’ve attached commented word documents of the manuscript and the supplementary appendix. In the manuscript, please note that “fluid inclusion” and “melt inclusion” almost never need a hyphen – please could this be changed throughout? I picked up a few other typos and grammatical errors. Sorry this is picky, but I think it will aid good communication to sort these out. Almost all the other comments on the mark-up word documents are very minor and should take very little time to fix. Thanks to the authors for an interesting read, and best of luck seeing this manuscript through to publication.

Best regards,

Margaret Hartley

University of Manchester

We thank the reviewer for their thorough work and appreciation of our study. We have addressed the specific comments from the attached pdf documents and screenshot below the majority of unrepeated comments (for example, we do not screenshot all the hyphen corrections, though we have addressed all of them).

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We have changed this throughout the text.

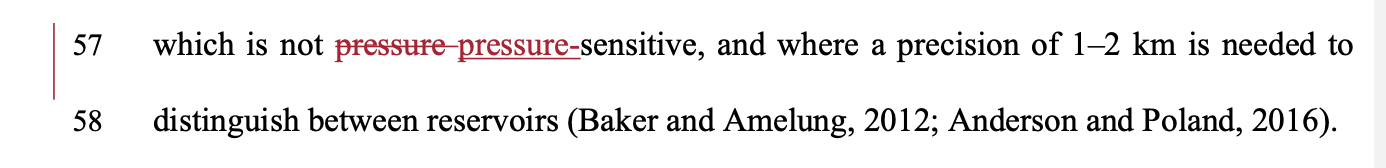


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We have significantly modified this section, which now reads a lines 125-174 “Developments in the past two decades in the accuracy and precision of Raman spectroscopy measurements of CO2 density, which uses spectral features of CO2 fluids to calculate a CO2 density using an instrument-specific calibration (e.g., Rosso and Bodnar, 1995; Kawakami *et al.*, 2003; Yamamoto and Kagi, 2006; Fall *et al.*, 2011; Wang *et al.*, 2011, 2019; Lamadrid *et al.*, 2017; Sublett *et al.*, 2020; DeVitre *et al.*, 2021; Le *et al.*, 2021), have made it possible to accurately measure lower CO2 densities, and thus identify shallow storage regions more reliably. Recent work demonstrated that Raman-based barometry of CO2-rich fluid inclusions returns the same results as melt inclusion barometry and provide much smaller uncertainties than mineral barometry, while requiring far less time and resources than melt inclusion analyses (Dayton *et al.*, 2024; DeVitre and Wieser, 2024; Lerner *et al.*, 2024).”



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We moved this sentence earlier as suggested by the reviewer. It is now: “ Recent studies have speculated that fluid inclusion barometry, using either micro thermometry or Raman spectroscopy, could be performed quickly enough to be useful for near-real-time volcano monitoring (Dayton *et al.*, 2023; Zanon *et al.*, 2024b). Indeed, advances in the accuracy of Raman measurements from two decades of improvements in CO2 densimetry calibrations along with new capabilities to process data in a more streamlined and reproduceable way (e.g., Wieser and DeVitre, 2024) show that Raman-based fluid inclusion barometry could now be fast-enough for this application. However, this has not been formally demonstrated. The CONVERSE Hawai‘i Scientific Advisory Committee (Cooper *et al.*, 2023) specifically recommended that key science questions should be identified, and pre-planning science activities performed, to facilitate more rapid implementation across a broader scientific group during hazardous eruptions. Here, we performed a near-real-time simulation to rigorously assess how quickly fluid inclusion depths can be obtained from erupted material using Raman spectroscopy, and whether these timescales are short enough to have utility as a petrological monitoring tool..[..]”

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We amended this to observatory or academic staff, given that no special circumstances or outside of hours work was needed to complete the work on the academic research side.

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We agree and hence changed these to match the reviewer’s suggestion.

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We added a reference to DiadFit earlier in the text, right before discussing the H2O presence in the fluid; 295-301: “All spectra processing and subsequent calculations were performed using DiadFit (Wieser and DeVitre, 2024), allowing for a conversion of raw spectra to CO2 densities within ~15 minutes. Pressures were calculated using the pure CO2 EOS of Span & Wagner (1996) implemented in DiadFit. At the time of our simulation, it was challenging to perform EOS calculations considering the possible presence of H2O in the exsolved fluid due to a lack of publicly available software running on modern operating systems.”

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Comments were addressed in the text (see tracked document)

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Changes made. Hyphens for fluid-inclusion removed throughout the text.

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Changes made. We replace with percentile.

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Change made as requested by reviewer.

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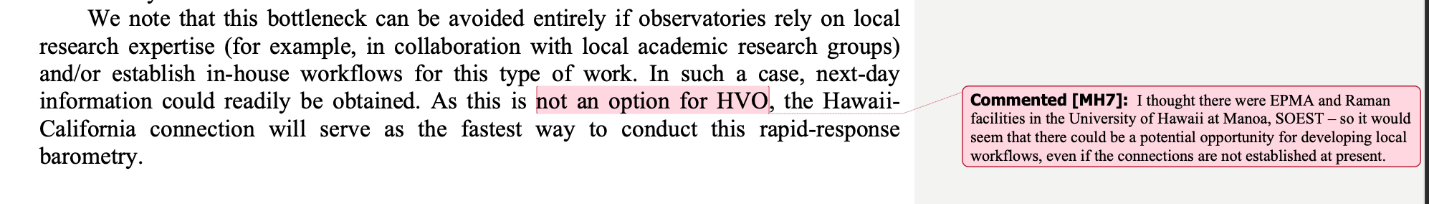
Regarding the first comment, this was unfortunately out of our hands – we had to submit the files as requested by the journal and pdf was not accepted as a figure format at time of submission.

As for the second comment, we thank the reviewer for pointing this out. It was just a mistake in the x axis scale of pressure for panel a and has now been fixed.

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These comments have been addressed in the supplement. There was a mismatch of versions and figures that has been fixed.



We agree that there may be potential to build a local workflow, though using the facilities at Manoa would still require shipping samples in some capacity, thus not significantly different from the current collaboration. Additionally, the Raman facilities at UH Manoa are not currently calibrated for this type of measurement, and this work also requires students and postdocs to be trained in fluid and melt inclusion hunting, which are not expertise currently reflected at Manoa, and may not be enough of a research priority to drop all other work for a few days.

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This was an issue with the scale, it is fixed.

All other supplement comments (hyphens, etc) have also been fixed.

Reviewer: 3  
  
Comments to the Author  
Review for “Depths in a Day”

20-21: depth of magma storage … high-priority information for volcanic observatories that is not currently obtainable on timescales relevant to eruption response: I wonder if this is a stretch. There are several cases with geophysical monitoring and seismic data are fully capable of detecting the depth of magma storage and movement prior to eruption onset. Grindavik for example. What I think would be better is instead highlighting the petrological monitoring aspect.

We agree with the reviewer that geophysical methods can probe for this information when the monitoring networks are good enough. Therefore, we emphasize that the information is not obtainable via petrological monitoring methods (screenshot).

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90: I would have liked to see this method applied to a volcano with much less precise information than Kilauea. Is it possible to either do this, or add on a section where you go through the hypothetical process of navigating a case study with little to no prior information?

Using Kīlauea as a testing ground was intentional because the information is so well constrained, and the eruption was a very low-risk and low-stakes scenario and therefore the responsible choice for a stress-test such as this. This is based on the suggestions of CONVERSE, and different other studies such as Andrews et al., 2019 and Diettrich and Neal 2022 which we mention near lines 79-80. “The CONVERSE Hawai‘i Scientific Advisory Committee (Cooper *et al.*, 2023) specifically recommended that key science questions should be identified, and pre-planning science activities performed, to facilitate more rapid implementation across a broader scientific group during hazardous eruptions. […] Performing these simulations during relatively small, low-hazard eruptions (as here) or as hypothetical simulations (e.g., (Andrews *et al.*, 2019)) is vital to iron-out bottlenecks so that we are as prepared as possible for the next large volcanic crisis (Dietterich and Neal, 2022).”. We appreciate that due to the manuscript’s organization, this was not clear in the previous version and in accordance with Reviewer#2’s suggestion, we’ve now moved this statement earlier in the introduction.

As the reviewer is probably aware, getting samples on recent activity is often very political, and requires having solid existing ties with observatories. We also chose Hawai’i because they were willing to work with us, and had written a letter of collaboration for our NSF grant proposing this work. We have requested samples from Iceland, but did not receive a response. We hope that after publishing this letter demonstrating what our laboratory can do, we can leverage this to build connections with other volcano observatories to apply to future eruptions at lesser known regions around the globe.

However, we have expanded the discussion on how to perform analyses on volcanoes with little information available in various parts. For example see 816-875: “It is also interesting that within hotspot and intraplate settings, regions with tholeiitic compositions (e.g. Iceland, Hawai’i, Galápagos, Réunion, Deccan Traps) generally have lower values than regions with more alkalic magmas (e.g., Canary Islands, Azores, Cabo Verde, Fig. 5). This likely represents the lower melt extents in alkalic settings, and the possibility of more volatile-rich sources (e.g., DeVitre *et al.* 2023). This observation is useful when tackling different volcanoes in specific tectonic settings. For example, even if there is no pre-exiting volatile data for a specific volcano in an OIB setting, a first approximation of the expected can be made from this compilation and some knowledge of the chemistry of the erupted magma. One could fit a general polynomial for an alkalic or tholeiitic OIB type magma with a generous error window encompassing available data from different volcanoes and perform a reasonable correction on fluid inclusion pressures.

And lines 887-916: ” Of course, at many volcanoes worldwide, detailed melt inclusion measurements may not be available to accurately calculate XH2O, particularly given the paucity of studies worldwide measuring both the bubble and glass phase of melt inclusions (Wieser *et al.*, 2024). However, this does not mean rapid-response fluid inclusion barometry cannot be used in these systems. Clearly, unless detailed melt inclusion measurements have been performed demonstrating high CO2, low H2O magmas, fluid inclusion barometry should not be applied to arc volcanoes during an eruptive crisis. However, excluding arcs, we believe the knowledge of the tectonic setting, major element chemistry, and phase abundances (which are often available, or easily constrained, even if there is no volatile data) will allow an informed guess of the likely range of XH2O values. For example, our global compilation shows a clear pattern, where more alkalic systems have higher XH2O values at a given pressure (Fig. 5). Thus, by conducting rapid XRF, EPMA, or EDS analyses, an informed guess would be possible. Using these general trends (Fig. 5), we would feel comfortable applying the rapid-response fluid inclusion method to any OIB or intraplate volcano erupting mafic compositions. Calculations would be conducted with a wide range of XH2O values informed by local studies/our global database, and the associated uncertainties propagated when calculating pressures. As more data become available (e.g. post eruption), these estimates could be revised within uncertainty to better pinpoint magma storage depths. For volcanoes that are not currently erupting but have had historic eruptions, an informed guess on a likely XH2O range can be made using chemical information from such previous eruptive events. For instance, one could easily determine the chemical tendency of a volcano from already existing major element data (e.g., Fig. 4a). In many cases, even when no chemical data is available for a specific volcano, analogy may be possible to draw from neighbouring volcanoes. For example, during the 2022 eruption of Mauna Loa, no detailed melt inclusion measurements accounting for the vapour bubble were available. However, as a first estimate, the P-XH2O relationships from neighbouring Kīlauea could be used. We note that once arc magmas are excluded from the compilation, even if is entirely unconstrained, fluid inclusion barometry is still more accurate than other methods such as mineral-melt thermobarometry in recovering magma storage pressures.

”

108: What about samples without CO2 fluids? As in, samples that are pure melt inclusions?

Is the presence of fluids a pre-requisite for this approach?

Fluid inclusion barometry requires the presence of fluids and specifically of CO2 with little other fluids (the effect of H2O, for example, is discussed in the supplement). In the case of melt inclusions, the appropriate approach would be volatile solubility modelling. We are not sure what the reviewer is getting at in this comment. If the samples do not have fluid inclusions, then this method is inherently not appropriate. We have added a sentence implying that the system must be CO2-saturated for this to worIk.

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241-244: I think this goes against your initial paragraph – indeed, this method is not

applicable to regions with little to no data, its only applicable to highly active regions that

inherently has a lot of available/published data.

We respectfully disagree with the reviewer, the method is applicable to regions with little data – a survey can be conducted in such regions, with some educated guesses about for example temperature of entrapment based either on existing glass/mineral data and or regional information. As we demonstrated, the effect of entrapment temperature is minimal on the results. In the case of XH2O, we suggest that a first order estimation can be made based on the compilation presented in this study, and we have expanded this discussion, including adding two figures in the main text showin XH2O trends. For example, if one were to look at a poorly studied alkaline OIB, the trend in pressure vs XH2O for alkaline OIBs could be used to gain a first order approximation of XH2O for the sample suite in question. In such cases, the appropriate course of action would be to conduct a survey of melt inclusions in existing sample suites from previous events.

However, we appreciate that this may not have been clear enough and have expanded on this at lines 887-916 for example:

” Of course, at many volcanoes worldwide, detailed melt inclusion measurements may not be available to accurately calculate XH2O, particularly given the paucity of studies worldwide measuring both the bubble and glass phase of melt inclusions (Wieser *et al.*, 2024). However, this does not mean rapid-response fluid inclusion barometry cannot be used in these systems. Clearly, unless detailed melt inclusion measurements have been performed demonstrating high CO2, low H2O magmas, fluid inclusion barometry should not be applied to arc volcanoes during an eruptive crisis. However, excluding arcs, we believe the knowledge of the tectonic setting, major element chemistry, and phase abundances (which are often available, or easily constrained, even if there is no volatile data) will allow an informed guess of the likely range of XH2O values. For example, our global compilation shows a clear pattern, where more alkalic systems have higher XH2O values at a given pressure (Fig. 5). Thus, by conducting rapid XRF, EPMA, or EDS analyses, an informed guess would be possible. Using these general trends (Fig. 5), we would feel comfortable applying the rapid-response fluid inclusion method to any OIB or intraplate volcano erupting mafic compositions. Calculations would be conducted with a wide range of XH2O values informed by local studies/our global database, and the associated uncertainties propagated when calculating pressures. As more data become available (e.g. post eruption), these estimates could be revised within uncertainty to better pinpoint magma storage depths. For volcanoes that are not currently erupting but have had historic eruptions, an informed guess on a likely XH2O range can be made using chemical information from such previous eruptive events. For instance, one could easily determine the chemical tendency of a volcano from already existing major element data (e.g., Fig. 4a). In many cases, even when no chemical data is available for a specific volcano, analogy may be possible to draw from neighbouring volcanoes. For example, during the 2022 eruption of Mauna Loa, no detailed melt inclusion measurements accounting for the vapour bubble were available. However, as a first estimate, the P-XH2O relationships from neighbouring Kīlauea could be used. We note that once arc magmas are excluded from the compilation, even if is entirely unconstrained, fluid inclusion barometry is still more accurate than other methods such as mineral-melt thermobarometry in recovering magma storage pressures.

245: I’m pretty sure the Venugopal paper was not a Cascades paper, it was a Garibaldi Arc

paper – these are two different subduction zones based on the tectonics, chemistry, and

age. I think this needs to be updated in your figures too

We disagree with the reviewer here. While there are some debates about exactly how to classify the different segments of the Cascade arc (e.g. Pitcher and Kent, 2019), many of the seminar publications (e.g.Hildreth 2007), classify the Garibaldi Volcanic Belt as part of the Cascades – it results from the subduction of the same plate, and the Canada-US border is insignificant, Glacier Peak and Mt. Baker are often included as GVB, and we think the reviewer would happily classify these as the Cascades.

250: There is no denying that Raman analyses of vapour bubbles is essential to crack the

code on CO2 – but if this is a pre-requisite for samples that qualify for your “Depth in a day”

aim, then I think there is a big discrepancy here. If pre-determined Raman data is needed,

how would you provide magma storage depths? Please correct me if I’m wrong but it seems

like there is an inherent positive bias here –systems with lots of data are further spotlighted,

and systems with little to no data are made even more inaccessible. This further highlights

my point above.

We disagree that this is a discrepancy. The Raman data on melt inclusion vapour bubbles is not a pre-requisite to be able to use the fluid inclusion barometry. It simply provides better constraints on XH2O than those available otherwise.

Therefore, we do not mean that it is a pre-requisite, but rather that XH2O estimated worldwide from our compilation is probably an overestimate (meaning the real uncertainty is smaller than we estimate). What this means is that our XH2O compilation shows “a worst-case scenario”, such that IF all the melt inclusion data in the compilation had originally considered CO2 in the bubbles, the XH2O estimated would likely be much smaller than they currently portrayed. In essence this means that perhaps the technique is more broadly applicable than this shows and has a smaller uncertainty. In any case, it also means that the correction on pressure is likely smaller than what could be estimated using glass-only melt inclusion data. It does not mean that systems with little data are made more inaccessible, it only means that the uncertainty is a bit smaller on systems with more data – but not prohibitively. One can still use a basic estimate of XH2O from the compilation based on the composition of the samples, or from existing MI data (if it exists) along with an estimate of entrapment temperature based on composition and calculate a pressure from fluid inclusion data. One could even assume an XH2O as many fluid inclusion barometric studies have done in the past. As before lines 887-916

256: I appreciate this point as it addresses my concerns! But what are the range of errors

when needing to estimate XH2O? Is it within the capabilities of the depths in a day

approach?

This question is addressed by now Fig. 2 and the discussion on sensitivity of the method to XH2O. It will depend on the context. For example, in the context of Kīlauea (depicted by the stars on panels b and d of Fig 1), an error of +-0.1 XH2O (+-10 mol%; a fairly large range) results in an uncertainty that is within the range of depths of a single reservoir. Even an error of +- 20mol% H2O would not drag fluid inclusion data from one reservoir to another. In other words, whether there is 0 or 40 mol% H2O in the fluid at Kilauea does not matter in terms of interpretation. On Fig 1d we plot the corrected histogram for Kilauea – note that it did not change anything about the results.

This can easily be assessed on an individual basis at different volcanoes of interest using DiadFit but we think that it is beyond the scope of this paper to discuss it any further.

Again lines 816-916 expand on how to approach volcanic systems with little data.

259: I think this line is a little too vague and almost feels like an afterthought. I think this

needs to be a section on its own. How would datasets with and without prior Raman data

subjected to your Depths in a day method compare to solely using mineral-melt

thermobarometry?

Uncertainties on mineral-melt thermobarometry have been discussed in great detail in Wieser et al., 2023 (Barometers behaving badly) and Wieser et al. (2024, termining P-T-X conditions of magma storage) and we think that this comparison is beyond the scope of the paper. Errors on mineral-melt barometers can easily span several km while an error of +-10 mol% H2O would barely amount to ~1 km at Kilauea. We think that Fig 1d and Fig1 show the effect of this clearly. Tests on other volcanoes and system are beyond the scope of the paper. Fluid inclusion barometry is often compared to mineral-melt barometry (i.e., Magma storage at Ocean Islands from Barker et al. 2021, Lo Forte, 2023; and many previous fluid inclusion studies). They overall give the same answers, though the error on mineral-melt barometers is much larger.

Overall, I find this to be a great study with broad applicability. My main concerns stem from

the sensationalizing of the lack of magma storage depth information from monitoring

techniques and the applicability to volcanic systems with little to no prior data (when really

its not entirely possible and/or incurs additional error).

We thank the reviewer for their thorough work. We clarified that other petrological methods are not able to acquire storage depth information on monitoring timescales, as we agree that if geophysical instrumentation networks and velocity models are available, these can retrieve this kind of information. We disagree that it is not possible to apply the method to systems with little data – fluid inclusion barometry has been successfully applied to many such systems over decades albeit it had never been rigorously demonstrated to be useful as a monitoring tool which is what our contribution set out to do. It does incur in more uncertainty if you know less about the system; however, this uncertainty is still smaller than that for example obtained from many mineral-melt barometers and this statement applies to any method. We agree, however, that more discussion on specific scenarios is beneficial and therefore we have expanded this see lines 816-916. We also discuss the advantages and limitations of the method in much more detail in the new section 3 Advantages and limitations of fluid inclusion barometry.

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I also have concerns over sample type – what about systems that predominately erupt via lava flows? Would that qualify for this approach?

There is some debate about whether fluid inclusion barometry can be applied to lava flows vs tephra, where for example some studies have suggested that FI from lava flows can be more reequilibrated than those found in tephra (see for instance Hildner 2011). It once again, depends on the trapping conditions and systems. In Devitre and Wieser 2024 GPL, we discussed reequilibration of FI at Kilauea in great length. Briefly, we compare lava flow and tephra samples, and model re-equilibration, and find no change within measurement uncertainty for typical lava flow times (<1 % change). Of course, if the internal pressure of the FI is higher (like in Atlantic OIBs for example), the difference may be larger and 10-20% reequilibration could be achieved within 10s of hours at high temperature. In those cases, the appropriate course of action is to model the conditions of the system of interest, which is easily doable using our FI reequilibration module in DiadFit. We now mention in the new section 3 (line 560) on the limitations of the method that although ideally tephra should be used given that FI in lava flows can re-equilibrate, it is possible to use lava flows and model the effect of transport time and temperature on re-equilibration of the target FI using a model such as that of Wanamaker and Evans 1989, which is implemented in DiadFit.

Lines 685-694

“From the perspective of choosing suitable samples for fluid inclusion work, it has been suggested that fluid inclusions found in lava flow samples may re-equilibrate more readily than those found in tephra (Hildner *et al.*, 2012) as they cool slower upon eruption, however, this effect is more likely to be significant for fluid inclusions trapped at high-pressures. In shallower systems, no significant difference is observed between naturally quenched lava flow samples and those rapidly quenched in water (DeVitre and Wieser, 2024). For a given fluid inclusion density, size, and ascent path, it is possible to model the predicted re-equilibration scenario using a plastic deformation model, allowing assessment of the possible influence of equilibration on the measured density (Wanamaker and Evans, 1989; Yamamoto *et al.*, 2011; DeVitre and Wieser, 2024).”

Systems like Stromboli – where its very hard/dangerous to access freshly erupted tephra – how would that work? I appreciate the concept, but I feel like the execution is a lot more complicated than this paper makes it seem.

Stromboli is not a system where this method would be applicable as shown on Fig3. The Aeolian arc is in general much too wet, XH2O in the fluid is very elevated. We do not suggest that this method is useful at every volcano in the world – just like any other method, it has limitations. The strength of this technique is at lower XH2O volcanoes, which tend to display more lava-flow or fire fountain dominated eruptions, which are possible to sample.

Can an EOS be generated for fluids hosted in minerals other than olivine? Or is it specific to olivine hosts?

The EOS is not specific to the host, it is specific to the fluid. For example, the Span and Wagner (1996) EOS is for pure CO2. The Duan and Zhang (2006) is for H2O-CO2 mixtures. These EOS are applicable regardless of the host, they are just the fluid’s properties. The assumption is that the volume of the void has stayed constant since trapping (Roedder 1984).

I feel like in its present state – it is two separate papers that could be expanded upon. Your

workflow to calculate depths in a day and the theory behind it is excellent. But I feel like

that should be a paper on its own, with the addition of several sections: application to

systems without prior raman data, applications to systems that emit lava flows, applications

to systems to very little knowledge. Once you’ve established this, then I feel you could go

into detail about global applicability as you have done. I fear that this journal format is not

for this paper – either make it longer as a true JPet paper, or publish companion papers and

the broad applicability section could qualify for a short JPet paper.

This comment is in contradiction with the recommendations from previous reviewers. In the original paper we submitted to JPET, there was no broader applicability section but the reviewers requested enhancement of the paper in this direction. This is why the broader applicability section was added. Therefore, we feel that the paper cannot be separated into two. In fact, the central goal of the paper was simply to demonstrate the feasibility of using FI barometry as rapid response petrological tool – a proof of concept and stress test.

In accordance with reviewer#1 and #3, we extended the paper and now include more specifics on applyuing the method under different information scenarios. We also added a section on advantages and limitations of the method.