**Depths in a day - A new era of rapid-response Raman-based barometry using fluid inclusions.**

Running title: Depths in a day – a new era of rapid response barometry

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**Abstract (250 max)**

Rapid-response petrological monitoring is a major advance for volcano observatories to build and validate models of the plumbing systems that supply eruptions in near-real-time. Our rapid-response analysis of tephra from the September 2023 eruption of Kīlauea shows that Raman analyses of fluid inclusions can robustly determine magma reservoir depths within a day of receiving samples – a transformative timescale for decision making that has not previously been achieved by petrological methods. This method is broadly applicable to many ocean island systems dominated by CO2-rich fluids.

**Keywords:** Fluid Inclusions; Geobarometry; Raman Spectroscopy; Rapid Response; Volcano Monitoring

**Main Text**

Volcano observatories increasingly use data collected from erupted lava and tephra samples in near-real-time to obtain information about the magmatic plumbing system to help inform decision-making during volcanic crises (Gansecki *et al.*, 2019; Re *et al.*, 2021; Pankhurst *et al.*, 2022). Most work so far has focused on the chemistry of erupted lavas and crystal cargoes (Pankhurst *et al.*, 2022) to gain insight into changing melt composition and rheological properties (e.g., Gansecki *et al.*, 2019). However, up until now, petrological monitoring has been unable to address the high-priority question– ‘*Where is the magma coming from?*’ (Re *et al.*, 2021). At well-monitored volcanoes, such information can be used to draw analogies to previous eruptive episodes associated with specific storage reservoirs (e.g., vigour, pathway, or length of eruption), and to help interpret geophysical signals of ongoing activity. At poorly-monitored volcanoes, where there may be no prior constraints on magma storage geometry (Wieser *et al.*, 2023b), depths of storage are a vital parameter to begin interpreting new eruptive activity. Melt inclusion (MI) barometry, a widely popular petrological method to determine storage depths from volatile contents, takes months to years to complete (Re *et al.*, 2021). While mineral barometry can be implemented faster than melt inclusion measurements (only requiring electron probe microanalysis (EPMA) measurements on eruptive minerals), it is imprecise (Wieser *et al.*, 2023a), and therefore would only be able to constrain magma storage to very broad depths (e.g., stored in the crust vs. below the Moho). Mineral thermobarometry also has poor applicability at active volcanoes such as Kīlauea or Mauna Loa where the only major silicate phase in most lavas is olivine, the chemistry of which is not pressure sensitive, and where a precision of 1–2 km is needed to distinguish between storage reservoirs (Baker and Amelung, 2012; Anderson and Poland, 2016).

Recent developments have shown that Raman-based barometry of CO2-rich fluid inclusions (FI) provides an alternative to popular petrological barometers, with much smaller uncertainties than mineral barometry, and requiring far less time and resources than MI analyses (Dayton *et al.*, 2023; DeVitre and Wieser, 2024). This method uses spectral features of CO2 fluids to calculate a CO2 density using an instrument specific calibration (DeVitre *et al.*, 2021). Along with an estimate of entrapment temperature, this density is converted into an entrapment pressure using a CO2 Equation of State (EOS, Fig. 1). We highlight that one major advantage of this method is how relatively insensitive to large temperature uncertainties it is (Fig. 1). Even if we consider the entire range of measured temperatures from olivine-saturated liquids erupted in the history of Kīlauea volcano (~1100–1350 ˚C), the resulting pressure and depth difference is no larger than 20% (Fig. 1). Pressures are converted to depths through an estimate of crustal density. However, there has previously been no rigorous assessment of how quickly FI depths can be obtained from erupted material, and whether these timescales are short enough to have use as a real-time monitoring tool.

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**Figure 1. Temperature sensitivity of the pure CO2 EOS method for CO2-rich fluid inclusion pressure calculations.** (a) General relationship between CO2 density and Pressure for different magmatically relevant entrapment temperatures at Kīlauea. (b) Uncertainty in pressure and depth induced by temperature uncertainty over the range of pressures in panel a. (c) Close-up of panel a, depicting a range of pressures and depths relevant to Kīlauea volcano pre-eruptive magmatic storage. (d) Uncertainties in pressure and depth induced by temperature uncertainty over the range of pressures in panel c.

**Figure 1. Temperature sensitivity of the FI barometer.** (a) Pressure vs CO2 density calculated for various entrapment temperatures relevant at Kīlauea volcano. 1150 ˚C was our initial fixed temperature for days 1 and 2, 1240 ˚C is the rounded mean and median of all measured temperatures in our final dataset. (b) Depth and Pressure differences induced by uncertainty in temperature. Blue curves show the % difference in pressure (or depth) and maroon curves show the absolute difference in depth in km. (c) Closeup of panel a, representing relevant PT conditions for Kīlauea volcano. Grey boxes show the depth range of the magma storage reservoirs – HM for Halema’uma’u and SC for South Caldera – inferred from FI and MI barometry as well as geophysics (DeVitre and Wieser, 2024; Lerner *et al.*, 2024). Stars show hypothetical FI trapped at HM and SC reservoirs with error bars representing 1σ uncertainty from MonteCarlo simulations using a temperature uncertainty of ±150 K. (d) Closeup of panel b, showing depth and pressure differences induced by uncertainty in temperature for PT conditions relevant to Kīlauea volcano. Blue curves show the % difference in pressure (or depth) and maroon curves show the absolute difference in depth in km.

The eruption onset of Kīlauea volcano on September 10, 2023 provided an unprecedented opportunity to test the validity of this method during a response, given that depths of the main magma storage regions at this volcano have been well constrained by various independent geophysical and petrological methods, including prior FI barometry (DeVitre and Wieser, 2024). Tephra samples representing the first ~14 hours of the September 2023 eruption were collected by Hawaiian Volcano Observatory (HVO) geologists on September 12 and mailed to UC Berkeley on September 15th (Fig. 2).

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**Figure 2. Workflow of the study, all times on this figure are Pacific Standard Time (PST).** Stick people show the contribution of individual team members, to indicate the total time associated with each step. We note that AB was trained in these procedures during the simulation and all reports were sent to HVO prior to 5:30 pm Hawaii Standard Time (HST), thus allowing for decision making for the following day.

Our simulation started on September 20 at 9 am PST (Day 1), the morning after sample receipt (Fig. 2). We used a production-line style workflow involving two undergraduates, a 1st year graduate student, a post-doc, and an assistant professor, with stations for crushing and sieving, mineral picking, FI preparation, sample cataloguing, and analysis. The first steps were to crush and sieve tephra, pick olivine crystals (size fractions 0.5-1 and 1-2 mm) crystals, and begin mounting crystals in CrystalBondTM\* to search for FI. By 2 pm PST (5 hrs into the simulation), we had collected our first Raman spectra. By ~7 pm PST, we had processed the spectra from 16 FI to get CO2 densities using a calibration based on the relationship between CO2 density and Fermi diad splitting distance (DeVitre *et al.*, 2021; DeVitre and Wieser, 2024). CO2 densities were converted into pressures using the pure CO2 EOS of (Span and Wagner, 1996). At the time of our simulation, calculations considering the possible presence of H2O in the exsolved fluid were not possible in DiadFit but a more recent release allowed us to compared our original results from mixed CO2-H2O EOS calculations using the EOS of Duan and Zhang, (2006). The mean correction factor for all three days is ~10%, using estimates of XH2O from melt inclusion data (DeVitre and Wieser, 2024), which is smaller than the uncertainty of the method and implies that pressures would generally be ~10% higher than originally reported (Fig. 3 arrows, Fig SX in supplementary materials). Regardless, this slight difference in pressure does not affect the interpretation of our results (it does not change the likely reservoir that contributed the erupted magma). In the future, pressures can easily be corrected for presence of H2O using the same methodology as described in (DeVitre and Wieser, 2024). For the first and second days (prior to acquisition of EDS data), we assumed an entrapment temperature of 1150 ˚C, based on geothermometric estimates of erupted liquids for previous events such as the 2018 LERZ eruption (Gansecki *et al.*, 2019; DeVitre and Wieser, 2024). Compared to our assumption, if the inclusions had instead been trapped at 1350 ˚C, the difference in pressure would have been no more than 15%, which at conditions relevant for Kīlauea magma storage, corresponds to a difference in measured depth smaller than 0.4 km (Fig. 1b and d). We note that entrapment temperatures for FI were refined on Day 4 using EDS data and given that the maximum range of measured temperatures in our final dataset is only ~125 K (1182–1307 ˚C), this means that the pressure difference induced by our first assumption of 1150 ˚C is < 12 % for the whole dataset, and on average < 7 % (median and mean temperatures are ~1238 ˚C). On days 1 and 2, pressures were converted into depths using the crustal density model of Ryan, (1987) parameterized by Lerner *et al.* (2021). We shared the resulting histogram (Fig. 3a) of storage depths with HVO collaborators showing that crystals, and thus magma, were likely coming from the shallower Halemaʻumaʻu reservoir of Kīlauea (HMM on Fig. 3a–b). It worthwhile to note that the number of FI reported on Day 1 is comparable to many melt inclusion studies, which often aim for ~20 MI per sample but frequently report fewer. For example, Lerner *et al.* (2021) reported 14 MI with sufficient data – that is at least MI glass volatile contents and major element compositions - to produce saturation pressures for the LERZ eruption of 2018. We also had an additional ~20 FI fully prepared and catalogued for analysis by the end of Day 1.

On Day 2, these 20 FI were analysed, while additional FI were prepared and catalogued. After analysis of ~15 crystals hosting FI, these crystals were passed from the Raman to a workstation where they were removed from CrystalBondTM and placed on tape to make an epoxy mount. Epoxy was poured at the end of the day. By ~8:30 pm PST on Day 2, we shared an updated histogram of 46 FI pressures and depths, confirming the dominant contribution of the Halemaʻumaʻu reservoir (Fig. 3a and c). On Day 3, while waiting for the epoxy to fully set, we finished analysing prepared FI. Then we polished the mount and began cataloguing the regions of crystals on which to perform energy-dispersive spectroscopy (EDS). On Day 4, olivine forsterite contents (Fo = 100\*Mg/(Mg+Fe) molar) were determined by EDS, providing a framework to further interpret the plumbing system (Fig. 3d). The Fo content of an olivine is a function of MgO and FeO in the liquid and the Ol-Liq partitioning coefficient (KD). Thus, the Fo contents of the host olivine close to each FI can be used to assess the calculated storage depth in its broader petrographic context (e.g., distinguishing high-Fo olivines which crystallize from more primitive melts from low Fo olivines forming in more evolved melts). This olivine forsterite content can also be used to estimate the likely entrapment temperature of each fluid inclusion (see DeVitre and Wieser, 2024) for performing EOS calculations, rather than having to use a uniform temperature as on Day 1-2. We recalculated all FI pressures on Day 4 using fluid inclusion specific entrapment temperatures from our EDS data.

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**Figure 3. Evolution of results over 4 days.** a) Schematic model of Kīlauea’s plumbing system, indicating reservoir depths determined by geophysics and prior petrological work (HMM- Halemaʻumaʻu; SC – South Caldera). b) By the end of Day 1, FI revealed that the crystals were supplied from depths consistent with the Halemaʻumaʻu reservoir. Kolmogorov-Smirnov tests show that September 2023 FI are recording depths significantly shallower than those recorded by FI (critical D = 0.22, stat = 0.24, pval=0.016) and MI (critical D = 0.22, stat = 0.41, pval=3.51e-06) from the 2018 lower East Rift Zone eruption, which required a contribution from the South Caldera reservoir. 1 Melt inclusion data for the 2018 LERZ eruption is from Wieser *et al.*, (2021); 2 Fluid inclusion data for the 2018 LERZ eruption is from DeVitre and Wieser, (2023) c) By the end of Day 2, depths from 46 FI were sent to HVO, confirming a dominant role of the Halemaʻumaʻu reservoir. On Day 2 we applied a conservative degassing filter (SO2 mol% < 2.5). d) By the end of Day 4, after taking a mean of repeated analyses of single FI, applying more stringent data filters, using FI-specific temperatures, and a more appropriate crustal model (density of ~2300 kg/m3 with a normal error distribution of 100 kg/m3), entrapment depths with uncertainties were linked to crystal chemistry. Error bars correspond to uncertainties propagated using Monte Carlo simulations (see Supplementary Information S1 Appendix) Olivine Fo equilibrium field is calculated based on Glass EPMA data collected on September 11, 2023 (see Supplementary Information S1 Appendix). We note here that initial data for Days 1 and 2 did not filter out repeated analyses (1 repeated FI in Day 1 and 6 in Day 2), pressures were calculated using an estimated entrapment temperature of 1150˚C (Wieser *et al.*, 2021; DeVitre and Wieser, 2023), and depth was calculated using the model of (Ryan, 1987) described in (Lerner *et al.*, 2021) for crustal density.

Our results clearly show that the majority of FI were entrapped at ~1–2 km below the surface (Fig. 3d), which aligns well with the depths of the Halemaʻumaʻu reservoir interpreted from geophysics (Baker and Amelung, 2012; Anderson and Poland, 2016; Anderson *et al.*, 2019), MI barometry (Lerner *et al.*, 2021; Wieser *et al.*, 2021), and FI barometry (DeVitre and Wieser, 2023). While the greater number of analyses from data processed on Day 2 and Day 4 certainly enhance the story, it is notable that depths calculated on Day 1 fall within final proposed storage reservoir depths. Rapid EDS analyses of olivine Fo contents close to each FI reveal that olivine crystals grew from a wide range of melt compositions. It is interesting to note that FI in the cores of high-Fo (e.g., >86) olivine crystals return pressures indicative of the shallower Halemaʻumaʻu reservoir since it has been suggested in previous eruptions that these high-Fo olivine crystals predominantly grow in the deeper South Caldera reservoir (SC on Fig. 3a), where high MgO melts are thought to reside (Helz *et al.*, 2014; Pietruszka *et al.*, 2015, 2018; Wieser *et al.*, 2019; Lerner *et al.*, 2024). We suggest three possible scenarios to explain the relatively shallow pressures documented in high-Fo olivine crystals:

1) FI in high-Fo olivine crystals were entrapped within the South Caldera reservoir and then transported into the Halemaʻumaʻu reservoir, where the FI re-equilibrated to lower pressures prior to eruption over shorter timescales than would be required to reset the host Fo content.

2) High-MgO melts were injected into the Halemaʻumaʻu reservoir, where high-Fo olivine crystallized and trapped FI at shallow depths (Lerner *et al.*, 2024).

3) Complex skeletal growth of olivine crystals during extensive undercooling (e.g., (Welsch *et al.*, 2013) could mean that high-Fo olivine cores which initially grew in the SC reservoir texturally evolved and trapped lower pressure FI in the Halemaʻumaʻu reservoir.

We think that scenario 1 is unlikely given the that FI from the 2018 lower East Rift Zone eruption appear not to have re-equilibrated despite stalling in the Halemaʻumaʻu reservoir for up to 2 years (DeVitre and Wieser, 2023; Mourey *et al.*, 2023), and our models of FI re-equilibration indicate <10% change in pressure over this time period. Current data does not allow us to resolve scenario 2 vs 3, but this eruption could provide an opportunity to explore this further (e.g., through detailed Phosphorous mapping in olivine around FI as in Esposito *et al.*, (2023)). Regardless of the exact mechanism, our FI pressures indicate that erupted crystal cargo experienced storage at Halemaʻumaʻu reservoir depths prior to eruption, and thus this was the most probable reservoir supplying magma to the surface in the Sept 2023 eruption.

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Figure 4. Global compilation of XH2O in the exsolved fluid phase from melt inclusion data for Continental Rift, Intraplate, Mid-Ocean Ridge and Subduction Zone volcanoes. (a) World map coloured by Median XH2O of the melt inclusion suites, circles indicate Glass-only MI data and stars those for which CO2 has been constrained either by Raman or Homogenization experiments. (b) Boxplot of XH2O for melt inclusion suites plotted on panel a. (c) Boxplot of XH2O showing only MI suites which constrained Total CO2 either by Raman or Homogenization experiments.

As mentioned previously, assuming a pure CO2 fluid when H2O may be present in the exsolved fluid phase results in an underestimation of the entrapment pressure of fluid inclusions (Fig. 3), of which the magnitude depends on the fraction of H2O present at the time of entrapment, which in turn depends on the pressure (e.g., at high pressure the exsolved fluid phase is dominated by CO2). In the case of Kīlauea, the exsolved fluid phase is overwhelmingly dominated by CO2 (Wieser *et al.*, 2021; DeVitre and Wieser, 2023), and as shown previously, the effect of XH2O is small on calculated pressures. However, this may not be the case in other locations, such as subduction zone volcanoes (Fig 4), where H2O can be predominant in the fluid phase owing to the high-water contents of magmas. We compiled melt inclusion suites from all over the world and different tectonic settings and plot XH2O for magmas with SiO2<57 wt% on Fig 4. Overall, it is evident that subduction zones record much higher XH2O globally than Mid-Ocean Ridge basalts, continental rifts and intraplate volcanoes. However, we note that the vast majority of melt inclusion suites measured CO2 in the glass-only, which implies that XH2O is globally overestimated except in few locations. In general, the implication of this is that the fluid inclusion method is not appropriate for example in Alaska, Kamchatka, Central America but is suitable for volcanoes like those in Hawaii, Azores, Cabo Verde, Canary Islands and Reunion as well as the EAR, Iceland, Galapagos and perhaps even certain volcanoes in the Cascades with low XH2O. Better constraints on total CO2 is necessary around the globe to reassess XH2O and the viability of the method in specific locations.

This simulation shows that Raman-based FI barometry has significant potential for rapid-response petrological monitoring globally. It could be applied to any CO2-rich volcanic system – which includes numerous hazardous and frequently active volcanic regions worldwide (e.g., Galápagos, Réunion, Azores, Canary Islands, Iceland, Cabo Verde). The resources and personnel required are modest. Sample preparation was carried out using transmitted-reflected light microscopes from the University of California teaching collection, only using a research-grade microscope for sample cataloguing. Raman spectrometers are widely available at many universities, given that it is a popular technique in many other fields, such as material sciences, physics, chemistry, and biology, and the W-filament SEM used for EDS analyses to get olivine Fo contents has been around for 15 years (See Supplementary Information S1 Appendix).

This simulation also enabled us to identify several ‘bottlenecks’ in this rapid-response workflow (yellow stars, Fig, 1, see Supplementary Information S1 Appendix for further details) so that we could determine depths even faster during future eruptions:

1. No courier services ship packages out of Hilo, Hawaiʻi over the weekend, and estimated delivery days are not reliable.
2. The epoxy took 18 hours to cure enough for polishing (vs. 8 hours on the datasheet). Faster curing epoxies can be used to eliminate this delay.
3. We spent significant time cataloguing samples on a research-grade microscope to help navigate on the Raman microscope, but later realized that smartphone cameras with teaching microscopes would have worked faster.

Overall, we have demonstrated that a modest-sized research group with prior teaching and class commitments working without overnight shifts can obtain pressures on relevant timelines for understanding volcanic plumbing systems during periods of unrest. This technique adds valuable quantitative storage depth information that expands on HVO’s routine near-real-time chemical monitoring with bulk rock ED-XRF(Gansecki *et al.*, 2019). In a true eruptive crisis, magma storage depths could be obtained even faster by removing bottlenecks 1–3, implementing overnight shift work, and requesting teaching release and class absences for students.

**Author contributions**

Author contributions for lab work are shown on Fig. 2. CD and PW wrote the paper. CD, PW, AR, BR, and AB prepared tephra, picked olivine, found Fis, catalogued them, mounted them, and conducted Raman analyses. CD and PW performed all spectral fitting, data processing, and figure making, with schematic illustrations shown in Fig. 2 from AB. JG developed the Mg/Fe calibration for the EDS detector and MG performed EDS analyses with help from JG. KJL, DTD, NID and KMM collected samples, processed them in Hilo, and provided eruption context. KJL and DD prepared the glass mount and did the EMPA analyses.

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**Data availability**

All data are made available in the Supplementary Information associated with the publication. We include detailed materials and methods (S1 Appendix), complete processed Dataset (S2 Dataset), a compilation of microphotographs of the FI and crystals (S3 FI Image Compilation) and a record of emails and tracking receipts related to the samples, data sharing and manuscript submission (S4 Email and tracking record). All raw data and Jupyter notebooks are also stored on Github (<https://github>.com/cljdevitre/2023\_Kilauea-rapid-response-simulation). The Github repository will be archived on Zenodo upon acceptance.

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