# Depths in a day - A new era of rapid-response Raman-based

2 barometry using fluid-inclusions.

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- 3 Running title: Depths in a day a new era of rapid-response barometry
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- Word count: Abstract 148, Main 3990

**Commented [MH1]:** "fluid inclusion" and "melt inclusion" almost never need hyphenating

## ABSTRACT

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. The depth of magma storage has recently been identified as high-priority
information for volcanic observatories that is not currently obtainable on timescales
relevant to eruption response. To address this deficiency, we performed a 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 for decision makingdecision-making that has not
previously been achieved by petrological methods. Additionally, our global melt-inclusion
compilation for which we calculated fluid composition at the point of vapour saturation
demonstrates the robustness of the fluid-inclusion method 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).

- **KEYWORDS:** Fluid\_inclusions; Geobarometry; Raman Spectroscopy; Rapid-response;
- 31 Volcano Monitoring

 $<sup>^{\</sup>star}$  The use of trade names does not signify endorsement by the U.S. Geological Survey.

#### INTRODUCTION

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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 (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 unrest associated with a new-episode of eruptive activity (Pritchard et al., 2019). For example, 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).

Melt-\_inclusion 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 this, it is imprecise (Wieser *et al.*, 2023a), and therefore can only constrain magma storage to very broad regions (e.g., crust vs. sub-Moho). It 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 \*The use of trade names does not signify endorsement by the U.S. Geological Survey.

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which is not <u>pressure-pressure-sensitive</u>, and where a precision of 1–2 km is needed to distinguish between reservoirs (Baker and Amelung, 2012; Anderson and Poland, 2016).

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Recent developments have shown that Raman-based barometry of CO<sub>2</sub>-rich fluidinclusions provides an alternative, with much smaller uncertainties than mineral barometry, and requiring far less time and resources than melt-inclusion analyses (Dayton et al., 2023; DeVitre and Wieser, 2024). This method uses spectral features of CO<sub>2</sub> fluids to calculate a CO<sub>2</sub> density using an instrument-instrument-specific calibration (DeVitre et al., 2021). Along with an estimate of entrapment temperature, this density is converted into an entrapment pressure using a CO<sub>2</sub> Equation of State (EOS, Fig. 1). One major advantage is that the conversion of CO<sub>2</sub> density to pressure is relatively insensitive to the choice of entrapment temperature, a parameter which may not be known at the onset of a new eruptive episode (Fig. 1a-b). The difference in pressure for EOS calculations considering the lower and upper limit of liquidus temperatures for olivine-saturated melts erupted at Kīlauea volcano throughout its history (~1100 and 1350 °C; (DeVitre and Wieser, 2024)) is at most ~20-%, which corresponds to ~0.2-0.4 km at depths representative of the Halema'uma'u reservoir (1-2 km), and ~0.6-1.0 km at the depths of the South- Caldera reservoir (3-5 km; Fig. 1b and Fig. S3-S6). These errors are of similar magnitude to those associated with the conversion of pressures to depths through an estimate of crustal density (an issue affecting all petrological barometers).

Recent studies have speculated that fluid-inclusion barometry could be performed quickly enough to be useful for near-near-real-time volcano monitoring (Dayton *et al.*, 2023; Zanon *et al.*, 2024). Here, we performed a near-near-real-time simulation to rigorously assess how quickly fluid-inclusion depths can be obtained from erupted \* The use of trade names does not signify endorsement by the U.S. Geological Survey.

**Commented [MH3]:** Given the paragraph break, the reader asks "provides an alternative to what". How about just "... fluid inclusions provide much smaller uncertainties than "."

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**Commented [MH5]:** Does the place name need a hyphen? I would have thought its name was South Caldera (no hyphen), and if so then it should not get hyphenated when followed with a qualifier like "reservoir".

material, and whether these timescales are short enough to have utility as a petrological monitoring tool. The CONVERSE Hawai'i Scientific Advisory Committee (Cooper *et al.*, 2023) specifically recommended that key science questions should be identified, and preplanning science activities performed, to facilitate more rapid implementation across a broader scientific group during hazardous eruptions. Performing these simulations during relatively small, low-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). Importantly, this simulation revealed that rapid-response fluid-inclusion work in collaboration with academic institutions was not taxing on observatory staff and can feasibly be employed during future eruptions.

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### TIMELINE OF RAPID-RESPONSE SIMULATION

The eruption onset of Kīlauea volcano on September 10, 2023 provided an unprecedented opportunity to test the validity and speed of the fluid-\_inclusion method, given that depths of the two main magma storage regions (Halema'uma'u at 1-2 km and South-\_Caldera at 3-5 km) at this volcano have been well constrained by various independent geophysical and petrological methods, including prior fluid-\_inclusion barometry (DeVitre and Wieser, 2024; Lerner *et al.*, 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. S1). A schematic of the workflow and detailed timeline is available in the supplement (Fig. S1).

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Our simulation started on September 20 at 9 am PST (Day 1), the morning after sample receipt (Fig. S1). 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, fluid-inclusion preparation, sample cataloguing, and analysis. We crushed and sieved tephra, picked olivine crystals (size fractions 0.5-1 and 1-2 mm), and mounted them in CrystalBond<sup>TM\*</sup> to search for fluidinclusions. By ~2 pm PST, we collected our first Raman spectra, and by ~7 pm PST, we had calculated CO<sub>2</sub> densities from 16 fluid- inclusions using a previously established instrument-specific calibration of the relationship between CO<sub>2</sub> density and Fermi diad splitting distance (DeVitre et al., 2021; DeVitre and Wieser, 2024). We calculated pressures using the pure CO<sub>2</sub> EOS of Span & Wagner (1996). At the time of our simulation, it was challenging to perform EOS calculations considering the possible presence of H<sub>2</sub>O in the exsolved fluid due to a lack of publicly available software running on modern operating systems. However, recent work by Yoshimura (2023) identified errors in the published equations for the H<sub>2</sub>O-CO<sub>2</sub> EOS of Duan & Zhang (2006) and provided opensource C code meaning that such calculations can be now be performed in DiadFit. Using estimates of X<sub>H<sub>2</sub>O</sub> from previously published melt\_inclusion data at Kīlauea (Wieser et al., 2021; DeVitre and Wieser, 2024), calculated pressures would be ~10% higher than originally reported to HVO if the \_\_CO<sub>2</sub>-H<sub>2</sub>O EOS had been used (Fig. 2d, Fig S1 in supplementary materials). This does not affect the interpretation of our results, as the shift is far smaller than the pressure offset between the Halema'uma'u and South- Caldera reservoir.

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For the first and second days of the simulation, we assumed an entrapment temperature of 1150 °C for all fluid-inclusions, based on geothermometric estimates of previously erupted liquids (Gansecki *et al.*, 2019; DeVitre and Wieser, 2024). On day Day 4, we calculated entrapment temperatures for each fluid-inclusion—using the host forsterite content measured by EDS (DeVitre and Wieser, 2024), yielding temperatures spanning 1182–1307 °C. The average error induced by our initial assumption of 1150 °C is only ~7% (with a maximum offset of only 12 %).—While crystallization temperatures at Kīlauea are relatively well constrained relative to other volcanic systems, using similar regression methods to those employed by—(DeVitre and Wieser; (2024) relating liquid compositions to host olivine contents, it should always be possible to constrain temperatures within ~100 K at different volcanic systems using host mineral chemistry.

On days 1 and 2, pressures were converted into depths using the crustal density model of Ryan (1987) parameterized by Lerner *et al.*<sub>5</sub> (2021). We shared the resulting histogram (Fig. 2a-b) of storage depths with HVO collaborators showing that crystals, and thus magma, were likely coming from the shallower Halema'uma'u reservoir of Kīlauea (Fig. 2a-b). It worthwhile to note that the number of fluid-inclusions reported on Day 1 (N=16) is comparable to many melt-inclusion (MI) studies, which often aim for ~20 per sample but frequently report fewer. For example, Lerner *et al.*<sub>5</sub> (2021) reported only 9 MI from the 2018 eruption with sufficient data to calculate saturation pressures (counting MI with glass major element contents and H<sub>2</sub>O contents, MI with glass CO<sub>2</sub> measurements if there was no bubble, and glass + bubble measurements if a bubble was present). Using the same criteria,—Aster *et al.*<sub>5</sub> (2016) only reported 13 complete measurements from Lassen Peak.

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We also had an additional ~20 fluid- inclusions fully prepared and catalogued for analysis by the end of Day 1. On Day 2, these 20 fluid-inclusions were analysed, while additional fluid-inclusions were prepared and catalogued. After analysis of ~15 samples, the crystals were removed from CrystalBondTM\* and placed on tape for epoxy mountmaking. Epoxy was poured at the end of Day 2. By ~8:30 pm PST on Day 2, we shared an updated histogram of 46 fluid-inclusion pressures and depths from 28 crystals, confirming the dominant contribution of the Halema'uma'u reservoir (Fig. 2a and c). On Day 3, we finished analysing the remaining prepared fluid- inclusions. Then we polished the epoxy mount and catalogued the regions of crystals closest to each fluid- inclusion on which to perform EDS analyses. 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. 2d). The Fo content of an olivine is a function of MgO and FeO in the liquid and the Ol-Liq partitioning coefficient (K<sub>D</sub>). Thus, the Fo contents of the host olivine close to each fluid-inclusion 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 (DeVitre and Wieser, 2024) for performing EOS calculations, rather than having to use a uniform temperature as on Day 1-2. We thus recalculated all fluid-inclusion pressures on Day 4 using fluid-inclusion-inclusion-specific entrapment temperatures.

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Our results on Day 4 clearly show that the majority of fluid-inclusions were entrapped at ~1–2 km below the surface (Fig. 2d), which aligns well with the depths of the Halema'uma'u reservoir interpreted from geophysics (Baker and Amelung, 2012; Anderson and Poland, 2016;

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170 Anderson et al., 2019), melt- inclusion barometry (Lerner et al., 2021; Wieser et al., 2021), and 171 fluid-inclusions (DeVitre and Wieser, 2024; Lerner et al., 2024). While the greater number 172 of analyses from data processed on Day 2 and Day 4 certainly enhance the story, it is 173 notable that depths calculated on Day 1 fall within final proposed storage reservoir depths. 174 Rapid EDS analyses of olivine Fo contents close to each fluid\_inclusion reveal that olivine 175 crystals grew from a wide range of melt compositions. It is interesting to note that fluid-176 inclusions in the cores of high-Fo (e.g., >86) olivine crystals return pressures indicative of the shallower Halema'uma'u reservoir, given that it has been suggested based on previous 177 178 eruptions that these high-Fo olivine crystals predominantly grow in the deeper South-179 Caldera reservoir (Fig. 2a) where high-high-MgO melts are thought to reside (Helz et al., 180 2014; Pietruszka et al., 2015, 2018; Wieser et al., 2019; Lerner et al., 2024). We suggest 181 three possible scenarios to explain the relatively shallow pressures documented in high-Fo 182 olivine crystals: 183 1) Fluid-inclusions in high-Fo olivine crystals were entrapped within the South-Caldera 184 reservoir and then transported into the Halema'uma'u reservoir, where the fluid-inclusions 185 re-equilibrated to lower pressures prior to eruption over shorter timescales than would be 186 required to reset the host Fo content. 187 2) High-MgO melts were injected into the Halema'uma'u reservoir, where high-Fo olivine 188 crystallized and trapped fluid- inclusions at shallow depths (Lerner et al., 2024). 189 3) Complex skeletal growth of olivine crystals during extensive undercooling (Welsch et 190 al., 2013) could mean that high-Fo olivine cores which initially grew in the South-Caldera 191 reservoir texturally evolved and trapped lower pressure fluid-\_inclusions in the

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Halema'uma'u reservoir.

We think that scenario 1 is unlikely given the that fluid-inclusions from the 2018 LERZ eruption appear not to have re-equilibrated despite stalling in the Halema'uma'u reservoir for up to 2 years (Mourey et al., 2023; DeVitre and Wieser, 2024), and our models of fluid-inclusion re-equilibration indicate <10% change in pressure over ~ 2 yrs. Current data does not allow us to resolve scenario 2 vs 3, but this eruption could provide an opportunity to explore this further, such as through detailed Phosphorous-phosphorus mapping in olivine around fluid-inclusions (e.g., melt-inclusions in Esposito et al., 2023). Regardless of the exact mechanism, our fluid-inclusion pressures indicate that the erupted crystal cargo experienced storage at Halema'uma'u reservoir depths prior to eruption, and thus this was the most probable reservoir supplying magma to the surface in the September 2023 eruption.

## BROADER APPLICABILITY OF THE METHOD

The use of a pure  $CO_2$  EOS results in an underestimate of the entrapment pressure of fluid-inclusions if there was  $H_2O$  in the fluid at the time of inclusion entrapment (Fig. 2). At Kīlauea, melt-inclusion data indicates that the exsolved fluid phase is ~90%  $CO_2$  at pressures corresponding to the Halema'uma'u reservoir, and >95%  $CO_2$  at pressures indicative of the South-Caldera reservoir (Wieser *et al.*, 2021; DeVitre and Wieser, 2024). As discussed above, the effect of  $X_{H_2O}$  is small on calculated pressures presented here (Fig. 2d). However, to assess the utility of the fluid-inclusion method for rapid-response petrology globally, it is necessary to evaluate  $X_{H_2O}$  contents, and their effect on fluid-inclusion pressures.

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We compiled published melt- inclusion data from all over the world, spanning many different tectonic settings (Fig. 3). We calculate  $X_{\rm H_2O}$  using the solubility model MagmaSat (Ghiorso and Gualda, 2015), implemented in VESIcal (Iacovino et al., 2021). We show the distribution, median, 25th and 75th quantiles of calculated X<sub>H2O</sub> for 4069 meltinclusions with SiO<sub>2</sub><-57 wt%, MgO <-16 wt% and Saturation Pressure pressure > 20 MPa on Fig 3b-c, coloured by tectonic setting. For each volcano, there is a clear correlation between  $X_{H_2O}$  and pressure, where  $X_{H_2O}$  becomes very high at shallow pressures (Fig. S9a-i) as melt-inclusions are trapped during enhanced degassing of H<sub>2</sub>O upon ascent. Thus, in this compilation, the median and 25th quantiles are likely most representative of X<sub>H<sub>2</sub>O</sub> in the main magma storage region. We stress the importance of considering X<sub>H<sub>2</sub>O</sub> when determining the suitability of this method to a particular system given that the X<sub>H<sub>2</sub>O</sub> pressure correction tends to be more significant at higher entrapment pressures. For example, if we consider an  $X_{H_2O}$  of 0.1 (the median  $X_{H_2O}$  of our fluidinclusion dataset at Kīlauea and a commonly assumed X<sub>H2O</sub> in deep storage systems), the pressure correction goes from <-15% at pressures <-220 MPa (~10 km) to ~20% at 700 MPa ( $\sim$ 30 km). Naturally, the correction is even more significant if  $X_{H_2O}$  is greater than 0.1 (e.g., for  $X_{H_2O}$ =0.2, the correction is 25-30% at P<150 MPa and ~50% at P = 700 MPa; Fig 1).

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NoteworthyNotably, most melt\_inclusion suites in this-our global compilation did not measure  $CO_2$  in the vapour bubble, meaning that the total  $CO_2$  content has been underestimated, and  $X_{H_2O}$  overestimated. This can be demonstrated by comparing  $X_{H_2O}$  values at volcanoes where there are some studies with Raman measurements and some

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without (Fig. S9c EAR; Fig. S9h Kamchatka, and Cascades).—Thus, Fig. 3c shows a compilation only using melt-inclusions where bubble CO<sub>2</sub> was measured by Raman spectroscopy.

Both compilations demonstrate that subduction zones record much higher  $X_{H_2O}$  globally than Midmid-Ocean-ocean Ridge ridge basalts, ocean island basalts, continental rifts and intraplate volcanoes. It is also interesting that within hotspot and intraplate settings, regions with tholeitic compositions (e.g. Iceland, Hawai'i, Galápagos, Réunion, Deccan Traps) generally have lower  $X_{H_2O}$  values than regions with more alkaline magmas (e.g., Canary Islands, Azores, Cabo Verde, Supporting Fig. S9). This likely represents the lower melt extents in alkaline settings, and the possibility of more volatile-rich sources (e.g., DeVitre *et al.* 2023).

Thus, while rapid-response fluid-inclusion barometry is highly applicable to active volcanic regions such as Hawai'i, Iceland, East African Rift, Galápagos, Réunion, Cabo Verde, and the Canary Islands, it is not appropriate in subduction zones such as Alaska, Kamchatka, or Central America. Interestingly, although there are only two studies with Raman data in the Cascades arc (Aster *et al.*, 2016; Venugopal *et al.*, 2020), the inclusions with highest pressures have X<sub>H2O</sub> values <0.2. This may indicate that in dryer-drier subduction zones, fluid-inclusions may have some utility for the most mafic, CO<sub>2</sub>-rich magmas.

To increase the accuracy of rapid-response petrological monitoring during future eruptions, it should be a priority to perform melt-inclusion studies accounting for vapour bubble CO<sub>2</sub> in more volcanic systems worldwide, given the large offsets between studies

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accounting for bubbles and those which do not in for  $X_{H_2O}$  space (Fig. 3), and to determine approximate trends in  $X_{H_2O}$ -pressure space for a given volcanic system or region during times of quiescence. This will allow assessment of the suitability of the fluid-\_inclusion method for a given volcano, and permit appropriate corrections for the complexities of mixed fluids without requiring melt-\_inclusion work during each eruptive episode. In systems with no prior constraints, our observations of correlations between alkalinity and  $X_{H_2O}$  can provide a first-first-order assessment of appropriate  $X_{H_2O}$  values to use. We note that once arc magmas are excluded from the compilation, even if  $X_{H_2O}$  is entirely unconstrained, fluid-\_inclusion barometry is still more accurate than other methods such as mineral-melt thermobarometry in recovering magma storage pressures.

### CONCLUSION

This Our simulation shows that magma storage depths can be determined within a day of receiving samples, with modest resources and personnel requirements (e.g. no overnight shift work, with normal semester teaching and class schedules). For example, 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 measure olivine Fo contents has been around for 15 years (S1 Appendix). Importantly, this simulation shows-showed that rapid-response work in collaboration with universities was not taxing on observatory staff, particularly considering the usefulness of information

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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 retracting detracting from other essential duties during eruption responses. For example, during the 2018 LERZ Kilauea eruption, HVO's near-near-real-real-time chemical monitoring with bulk rock ED-XRF identified the appearance and disappearance of many magma batches (Gansecki *et al.*, 2019)<sub>25</sub> Efluid-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 been a critical addition to understanding the eruption and the system.

Our global compilation of  $X_{H_2O}$  values shows that Raman-based fluid-inclusion barometry has utility as a rapid-response petrological monitoring method at many of the world's most active and hazardous basaltic volcanoes (e.g., Galápagos, Réunion, Azores, Canary Islands, Iceland, Cabo Verde). As our understanding of exsolved fluid compositions improves as more studies account for  $CO_2$  held within vapour bubbles, it is likely that the applicability of this method may expand to even more volcanic systems (e.g. to drier arc magmas such as in the Cascades).

Overall, fluid-\_inclusion barometry is broadly applicable, and adds valuable quantitative storage depth information that provides a key advancement for volcano observatories that utilize near-real-time geochemical monitoring to better understand eruptions as they unfold (See-see\_overview-(Re et al., 2021); Hawai'i-(Gansecki et al., \*The use of trade names does not signify endorsement by the U.S. Geological Survey.

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2019); La Palma-(Pankhurst *et al.*, 2022); Fuego-(Liu *et al.*, 2020); Italy-(Corsaro and Miraglia, 2022)).

#### **AUTHOR CONTRIBUTIONS**

Author contributions for lab work are shown on Fig. S1. CD and PW wrote the paper. CD, PW, AR, BR, and AB prepared tephra, picked olivine, found fluid-inclusions, catalogued them, mounted them, and conducted Raman analyses. CD and PW performed all spectral fitting, data processing, and figure making, with schematic cartoons shown in Fig. S1 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, provided eruption context and edited the manuscript. KJL and DD prepared the glass mount and did the EMPA EPMA glass analyses.

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### ACKNOWLEDGEMENTS

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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 fluidinclusion dataset (S2 Dataset), the global meltinclusions dataset (S3
Dataset), a compilation of microphotographs of the fluid_inclusions and crystals that were
used for navigation only during the simulation (S3 FI Image Compilation), and a record of
emails reporting results to HVO and tracking receipts related to sample shipment (S4 Email
and tracking record). All raw data and Jupyter notebooks are also stored on Github at the
following link: <a href="https://github.com/cljdevitre/2023_Kilauea-rapid-response-simulation">https://github.com/cljdevitre/2023_Kilauea-rapid-response-simulation</a> .
The Github repository will be archived on Zenodo upon acceptance.

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#### FIGURE CAPTIONS

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451 Figure 1. Sensitivity of fluid\_inclusion barometry to temperature and molar 452 proportions of  $H_2O$  in the exsolved fluid phase  $(X_{H_2O})$ . (a)  $CO_2$  density vs Pressure 453 pressure for different magmatically relevant entrapment temperatures at Kīlauea using the 454 EOS of Span and Wagner, (1996). 1100 and 1350 °C are the lower and upper limit of 455 liquidus temperatures for olivine-saturated melts erupted at Kīlauea volcano throughout 456 its history, 1150 °C was the temperature used for days 1 and 2 of the simulation, 1240 °C 457 is the rounded mean and median of all measured temperatures in our final dataset. (b) 458 Panel a close-up. Grey boxes show Kīlauea magma storage inferred from fluid-inclusions, 459 melt-inclusions and geophysics (DeVitre and Wieser, 2024; Lerner et al., 2024). HMM= 460 Halema'uma'u reservoir, SC = South- Caldera reservoir. Stars show hypothetical fluid-461 inclusions trapped at Halema'uma'u and South-Caldera with T=1150 °C and error-bars 462 representing 1 \u03c4 uncertainty from Monte Carlo simulations using a temperature uncertainty of  $\pm 125$  °C ( $\frac{1350$ °C -1100°C). (c) CO<sub>2</sub> density vs <u>Pressure-pressure</u> at 1150 °C for various 463  $X_{H_2O}$  using the mixed  $H_2O$ - $CO_2$  EOS of Duan and Zhang (2006). The small discontinuity 464 465 at 200 MPa is due to parameter values being switched (Yoshimura, 2023). (d) Panel c 466 close-up. Stars show hypothetical fluid\_inclusions trapped at Halema'uma'u and South-467 Caldera with T=-1150 °C,  $X_{H_2O}$  inferred using the  $X_{H_2O}$  -P relationship of DeVitre and 468 Wieser (2024), and error-bars representing 10 uncertainty from Monte Carlo simulations using an  $X_{H_2O}$  uncertainty of  $\pm 0.1$  based on the maximum range of  $X_{H_2O}$  inferred in our 469 dataset  $(\frac{XH_2Omax-XH_2Omin}{2})$  when calculated using the upper limit  $X_{H_2O}$ -P relationship for 470 471 Kīlauea from DeVitre and Wieser (2024).

**Commented [MH15]:** To compare the two different equations of state it would help for panels (a) and (c) to have the same scale on the x and y axes. Is the pressure-depth conversion done differently in (a) and (c) or is this an artefact of how the axes are labelled?

**Commented [MH14]:** As an aside, as a Windows user I wasn't able to open the .eps files, and the pdf reproductions supplied by the journal are often not very high resolution. It's helpful if figures can be supplied in pdf format.

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473 Figure 2. Evolution of results over 4 days. a) Schematic model of Kīlauea's 474 plumbing system, indicating reservoir depths (HMM = Halema'uma'u; SC = South-475 Caldera). b) Day I fluid-inclusion depths, as reported to HVO, are consistent with the 476 estimated depths of the Halema'uma'u reservoir. Kolmogorov-Smirnoff tests indicate that 477 September 2023 fluid\_inclusions record significantly shallower depths than fluid-478 inclusions (critical D = 0.22, stat = 0.24, pval=0.016) and melt-inclusions (critical D = 0.026) 479 0.22, stat = 0.41, pval=3.51e-06) from the 2018 lower\_Lower\_East Rift Zone (LERZ) 480 eruption, which required a contribution from the South-Caldera reservoir (Wieser et al., 481 2021; DeVitre and Wieser, 2024). <sup>1</sup>LERZ 2018 melt- inclusions (Wieser et al., 2021); 482 <sup>2</sup>LERZ 2018 fluid-inclusions (DeVitre and Wieser, 2024). c) Day 2 data, as reported, 483 confirmed a likely dominant role for the Halema'uma'u reservoir. A conservative 484 degassing filter was applied (SO<sub>2</sub>/CO<sub>2</sub> peak ratio <-0.1). d) Day 4 data, as reported. Means 485 were taken for repeated analyses of single fluid-inclusions and additional data filters (e.g., 486  $SO_2/CO_2$  peak ratio <-0.22), fluid-\_inclusion-specific temperatures, and a more 487 appropriate crustal density model (~2300 kg/m<sup>3</sup> with a normal error distribution of 100 488 kg/m<sup>3</sup>) were applied. Error\_bars correspond to uncertainties propagated using Monte 489 Carlo simulations and olivine Fo equilibrium field is calculated based on Glass glass 490 EPMA data collected on September 11, 2023 (see Supplementary Information S1 491 Appendix). The shifted histogram ' $H_2O$  effect' shows the effect of  $H_2O$  corrections on pressures recalculated using  $X_{H_2O}$  inferred from melt-inclusions (Wieser et al., 2021; 492 493 DeVitre and Wieser, 2024).

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Figure 3. Global compilation of  $X_{H_2O}$  in the exsolved fluid phase from meltinclusion data for Econtinental Rift, Econtinental Intraplate, Aalkaline and Tiholeitic Oocean Iisland Bbasalt (OIB, see Fig S7), Mmid-Oocean Rridge and Ssubduction Zzone volcanoes (details and references in the supplement). Data is-are filtered to SiO<sub>2</sub> <-57 wt%, MgO <-16 wt% and Ssaturation Ppressure >20 MPa (supplement for details). (a) World map coloured by Median-median  $X_{H_2O}$  of the melt-inclusion suites. C,-eircles indicate Eglass-only MI data and stars those for which  $CO_2$  has been constrained by Raman. (b) Boxplot of  $X_{H_2O}$  for melt-inclusion suites plotted on panel a. Boxplots show the median, Q1 (25th quartile), Q3 (75th quartile) and whiskers mark the last datapoint before Q3+1.5\* (Q3-Q1) and the first datapoint after Q1-1.5\*(Q3-Q1). Violin plots show the density distribution of all the data and are coloured according to tectonic setting. (c) Boxplot of  $X_{H_2O}$  showing only MI suites which constrained Total-total CO<sub>2</sub> by Raman spectroscopy. EAR – East African Rift, GSC-1-Galápagos Spreading Center, NAR – North Atlantic Ridge, JdFR – Juan de Fuca Ridge, GR – Gakkel Ridge, MAR – Mid-Atlantic Ridge, EPR – East Pacific Rise, IBM – Izu Bonin Mariana.

**Commented [MH16]:** I would suggest using first and third quartile, or 25<sup>th</sup> and 75<sup>th</sup> percentile.

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