

20 **Abstract**

21 Rapid-response petrological monitoring is a major advance for volcano observatories to build and validate
22 models in near-real-time of the plumbing systems that supply eruptions. Our rapid-response analysis of tephra from
23 the September 2023 eruption of Kīlauea shows that Raman analyses of fluid inclusions can robustly determine
24 magma reservoir depths within a day of receiving samples – a transformative timescale for decision making that
25 has never been achieved by petrological methods.

26
27 **Main text**

28 Volcano observatories increasingly use data collected from erupted lava and tephra samples in near-real-
29 time to obtain information about the magmatic plumbing system to help inform decision making during volcanic
30 crises^{1–3}. Most work so far has focused on the chemistry of erupted lavas and crystal cargoes³ to gain insight into
31 changing melt composition and rheological properties (e.g., ref¹). However, up until now, petrological monitoring
32 has been unable to address the high-priority question– ‘*Where is the magma coming from?*’². At well-monitored
33 volcanoes, such information can be used to draw analogies to previous eruptive episodes associated with specific
34 storage reservoirs (e.g., vigour, pathway, or length of eruption), and to help interpret geophysical signals of ongoing
35 activity. At poorly-monitored volcanoes, where there may be no prior constraints on magma storage geometry⁴,
36 depths of storage are a vital parameter to begin interpreting new eruptive activity. Melt inclusion (MI) barometry,
37 a widely popular petrological method to determine storage depths from volatile contents, takes months to years to
38 complete². While mineral barometry could be implemented faster (only requiring electron probe microanalysis
39 (EPMA) measurements on eruptive minerals), it is imprecise^{5,6}, and therefore would only be able to constrain
40 magma storage to very broad depths (e.g., stored in the crust vs. below the Moho). This technique has particularly

41 poor applicability at active volcanoes such as Kīlauea or Mauna Loa, where a precision of 1–2 km is needed to
42 distinguish between storage reservoirs^{7,8}.

43 Recent developments have shown that Raman-based barometry of CO₂-rich fluid inclusions (FI) provides
44 an ~~exciting~~ alternative to popular petrological barometers, with much smaller uncertainties than mineral barometry,
45 and requiring far less time and resources than MI analyses^{9,10}. This method uses spectral features of CO₂ fluids to
46 calculate a CO₂ density using an instrument specific calibration¹¹. Along with an estimate of entrapment
47 temperature, this density is converted into an entrapment pressure using a CO₂ Equation of State (EOS). Pressures
48 are converted to depths through an estimate of crustal density. However, there has previously been no rigorous
49 assessment of ~~just~~ how quickly FI depths can be obtained from erupted material, and whether these timescales are
50 short enough to have use as a real-time monitoring tool.

51 The eruption onset of Kīlauea volcano on September 10, 2023 provided an unprecedented opportunity to
52 test the validity of this method during a response, given that depths of the main magma storage regions at this
53 volcano have been well constrained by various independent geophysical and petrological methods, including prior
54 FI barometry¹⁰. Tephra samples representing the first ~14 hours of the September 2023 eruption were collected by
55 Hawaiian Volcano Observatory (HVO) geologists on September 12. ~~Following discussions with HVO over the~~
56 ~~next few days, we requested samples to run a rapid-response simulation.~~

57 Our simulation started on September 20 at 9 am PST (Day 1) ~~following receipt of samples the day before~~
58 (Fig. 1). We used a production-line style workflow involving two undergraduates, a 1st year graduate student, a
59 post-doc, and an assistant professor, with stations for crushing and sieving, mineral picking, FI preparation, sample
60 cataloguing, and analysis. The first steps were to crush and sieve tephra, pick olivine crystals (size fractions 0.5-1
61 and 1-2 mm) crystals, and begin mounting crystals in CrystalBond™ to search for FI. By 2 pm PST (5 hrs into the
62 simulation), we had collected our first Raman spectra. By ~7 pm PST, we had analysed and processed all data for
63 16 FI and were able to share a histogram of storage depths with HVO collaborators showing that crystals, and thus

64 magma, were coming from the shallower Halema'uma'u reservoir of Kīlauea (HMM on Fig. 2a–b). We also had a
65 further ~20 FI fully prepared and catalogued for analysis. On Day 2, these 20 FI were analysed, while further FI
66 were prepared and catalogued. After analysis of ~15 crystals hosting FI, these crystals were passed from the Raman
67 to a workstation where they were removed from CrystalBond™ and placed on tape to make an epoxy mount. Epoxy
68 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
69 and depths, confirming the dominant influence of the Halema'uma'u reservoir (Fig. 2a and c). On Day 3, while
70 waiting for the epoxy to fully set, we finished analysing prepared FI. Then we polished the mount and began
71 cataloguing the regions of crystals on which to perform energy-dispersive spectroscopy (EDS). On Day 4, olivine
72 forsterite contents ($Fo = 100 * Mg / (Mg + Fe)$ molar) were determined by EDS, providing a framework to further
73 interpret the plumbing system (Fig. 2d).

74 Our results clearly show that the majority of FI were entrapped at ~1–2 km below the surface (Fig. 2d),
75 which aligns well with the depths of the Halema'uma'u reservoir interpreted from geophysics^{7,8,12}, MI barometry
76 ^{13,14}, and FI barometry¹⁰. While the greater number of analyses from data processed on Day 2 and Day 4 certainly
77 enhance the story, it is notable that depths calculated on Day 1 fall within final proposed storage reservoir depths
78 as well. Rapid EDS analyses of olivine Fo contents close to each FI reveal that olivine crystals grew from a wide
79 range of melt compositions. It is interesting to note that FI in the cores of high-Fo (e.g., >86) olivine crystals return
80 pressures indicative of the shallower Halema'uma'u reservoir – it has been suggested in previous eruptions that
81 these high-Fo olivine crystals predominantly grow in the deeper South Caldera reservoir (SC on Fig. 2a), where
82 high MgO melts are thought to reside. We suggest three possible scenarios to explain the relatively shallow
83 pressures documented in high-Fo olivine crystals:

84 1) FI in high-Fo olivine crystals were entrapped within the South Caldera reservoir and re-equilibrated at
85 Halema'uma'u pressures prior to eruption.

86 2) High-MgO melts were injected into the Halema'uma'u reservoir, where high-Fo olivine crystallized and trapped
87 FI.
88 3) Complex skeletal growth of olivine crystals during extensive undercooling (e.g., ref ¹⁵) meant that high-Fo
89 olivine cores which initially grew in the South Caldera reservoir texturally evolved and trapped lower pressure FI
90 in the Halema'uma'u reservoir.

91 We think that scenario 1 is unlikely given the that FI from the 2018 lower East Rift Zone eruption appear
92 not to have re-equilibrated despite stalling in the Halema'uma'u reservoir for up to 2 years^{10,16}, and our models of
93 FI re-equilibration indicate <10% change in pressure after such a period. Current data does not allow us to resolve
94 scenario 2 vs 3, but this eruption could provide an opportunity to explore this further (e.g., through detailed
95 Phosphorous mapping in olivine around FI). Regardless of the exact mechanism, our FI pressures indicate that
96 erupted crystal cargo experienced storage in the Halema'uma'u reservoir prior to eruption, and thus this is the most
97 probable reservoir supplying magma to the surface.

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

106 This simulation also enabled us to identify several 'bottlenecks' in this rapid-response workflow which
107 could be smoothed out during future eruptions (yellow stars, Fig 1):

- 1) No courier services ship packages out of Hilo, Hawai‘i 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)
- 3) We spent significant time cataloguing samples on a research-grade microscope to help navigate on the Raman, 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 ~~game-changing~~ 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-XRF¹. 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.

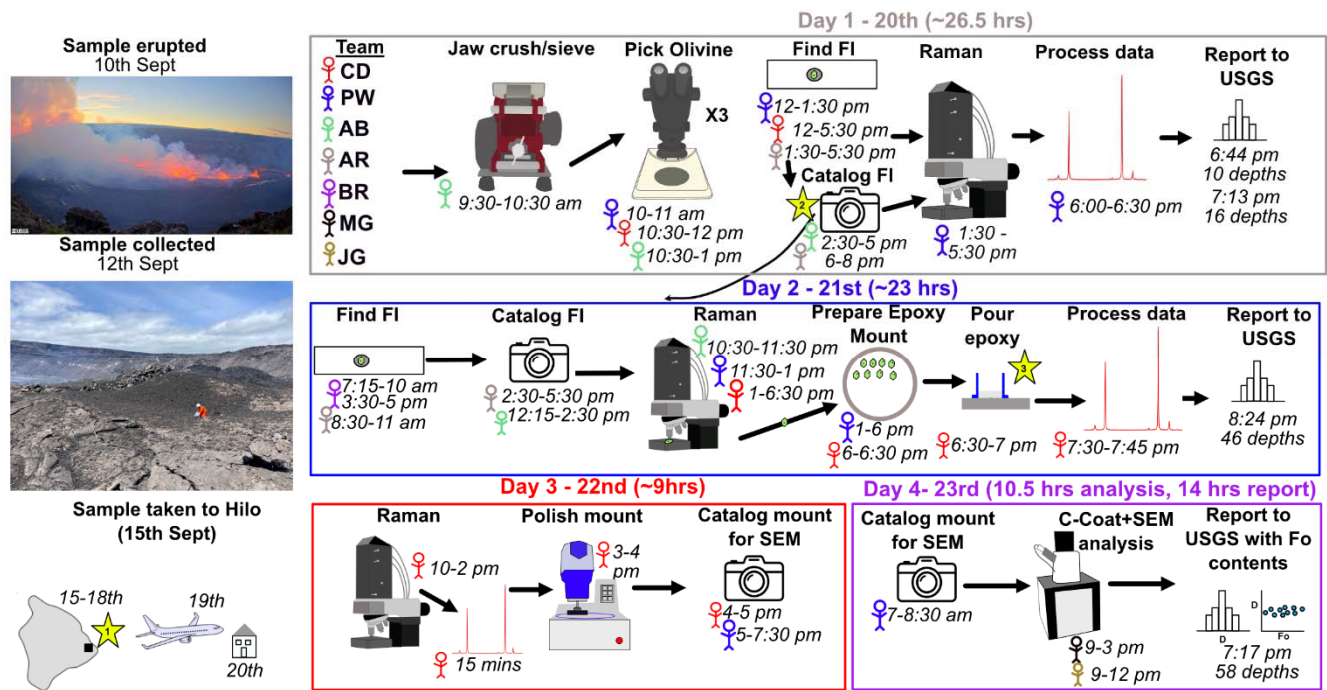
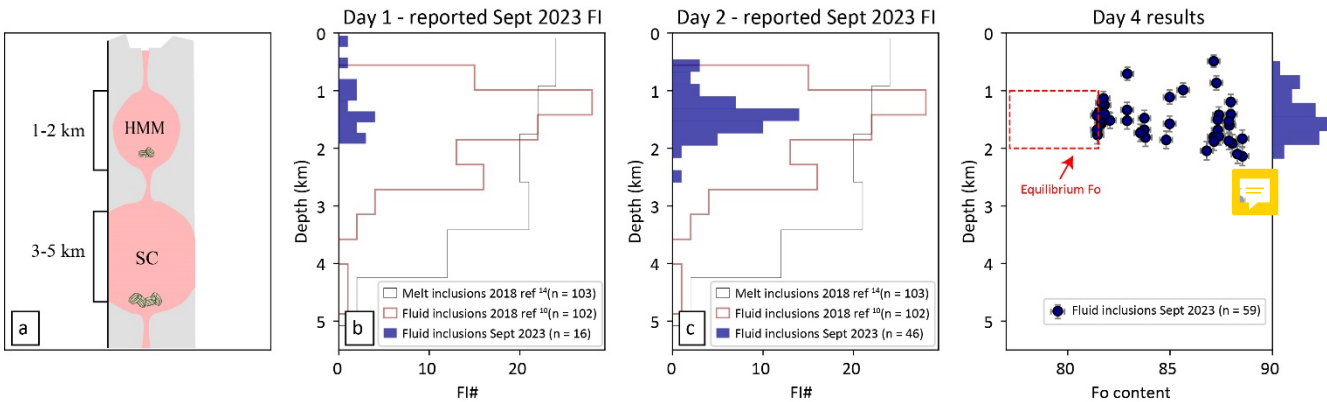


Figure 1: Workflow of the study, all times on this figure are Pacific Standard Time (PST). Stick people show the contribution of individual team members, to indicate the total time associated with each step. We note that AB was trained in these procedures during the simulation! We note that all reports were sent to HVO prior to 5:30 pm Hawaii Standard Time (HST), thus allowing for decision making for the following day.



127 **Figure 2: Evolution of results over 4 days.** a) Schematic model of Kīlauea's plumbing system, indicating reservoir
128 depths determined by geophysics and prior petrological work (HMM- Halema'uma'u; SC – South Caldera). b) By
129 the end of Day 1, FI revealed that the crystals were supplied from the Halema'uma'u reservoir. KS tests show that
130 within 5% confidence this is significantly shallower than depths indicated by FI (critical $D = 0.22$, $stat = 0.24$,
131 $pval=0.016$) and MI (critical $D = 0.22$, $stat = 0.41$, $pval=3.51e-06$) from the 2018 lower East Rift Zone eruption,
132 which required a contribution from the South Caldera reservoir. c) By the end of Day 2, depths from 46 FI were
133 sent to HVO, confirming a very dominant role of the Halema'uma'u reservoir. On Day 2 we applied a conservative
134 degassing filter (SO_2 mol% < 2.5). d) By the end of Day 4, after taking a mean of repeated analyses of single FI,
135 applying more stringent data filters, using FI-specific temperatures, and a more appropriate crustal model (density
136 of ~ 2300 kg/m³ with a normal error distribution of 100 kg/m³), entrapment depths with uncertainties were linked
137 to crystal chemistry. Olivine Fo equilibrium field is calculated based on Glass EPMA data (see Methods). We note
138 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),
139 pressures were calculated using an estimated entrapment temperature of 1150°C, and depth was calculated using
140 the model of ref¹⁷ described in ref¹³ for crustal density.

141

142 **Methods (Online)**

143 **Sample collection and preparation**

144 Tephra samples (USGS code KS23-588) representing the first ~14 hours of the September 10, 2023,
145 eruption of Kīlauea volcano were collected by Hawaiian Volcano Observatory (HVO) geologists on September 12
146 and shipped on Friday September 15 at ~5 pm HST. This tephra was erupted from a fissure which opened at 15:36
147 local time on September 10 (~22 minutes after the eruption started, between 15:13 and 15:14 local time) and ceased
148 erupting between 06:16 and 06:18 am local time on September 11. Following receipt of the samples at the
149 University of California, Berkeley on Tuesday, September 19, material was processed in a jaw crusher, and then

150 sieved into >2 mm, 1–2 mm, and 0.5–1 mm size fraction. Crystals were picked from the 1–2 mm and 0.5–1 mm
151 size fractions using three different binocular microscopes (one of which had the ability to cross polars). Then,
152 crystals were individually mounted in CrystalBond™ on glass slides and progressively polished with 1200-2500-
153 10000 grade wet and dry paper to find FI. FI were then passed onto a team member on a research grade scope to
154 take reflected and transmitted light images to aid with Raman navigation. These images were pasted into a Google
155 slides document so all lab personnel at UC could access them immediately.

156 Glass compositions were measured on USGS sample KS23-587, a lava flow sample collected in a molten
157 state and quenched with water at 06:30 HST on 11 September 2023. The sample was entirely glassy, and fragments
158 were mounted in epoxy in a 1” round and polished for microprobe analysis.

159

160

161 **Raman analyses**

162 Raman spectra were acquired using a WiTec Alpha 300R Raman spectrometer at the Department of Earth
163 and Planetary Science at the University of California, Berkeley. The relationship between CO₂ density and spectral
164 features was determined from a gas calibration cell following the methods of ref¹⁰. All spectra were acquired at 37
165 °C. Spectra were processed and corrected for drift using the Python tool DiadFit v0.0.73¹⁸. We report ratios of SO₂
166 and CO₃ to CO₂ peak areas and calculate approximate mol % SO₂ using the equations of ref¹⁹, implemented in
167 DiadFit. All measured FI had SO₂ mol % <10, we filtered the final dataset for SO₂ mol % <5 (Fig. 2d), to ensure
168 use of the pure CO₂ EOS was valid. We calculated densities from the Raman-measured separation of the Fermi
169 diad using the appropriate calibrated density equations for our instrument in DiadFit¹⁸. We then calculated pressures
170 using the EOS of ref²⁰ and an entrapment temperature estimated from the Fo content¹⁰. We propagated uncertainties
171 in FI depths using MonteCarlo simulations implemented in DiadFit. Entrapment depths in Fig. 2d were calculated
172 using a constant crustal density of 2300 kg/m³ and a normally distributed 1σ error of 100 kg/m³. Error in the CO₂

173 density for each FI was determined from the error in each peak fit, the Ne line drift correction model, and the
174 densitometer¹⁸. We used a 40°C error for temperature. In total we analyzed 62 FI hosted in 31 olivine crystals. Our
175 workflow is detailed in Fig. 1. Pictures of each FI and host crystal are available in the supplement (Image
176 Compilation S2).

177

178 **Epoxy mount making and polishing**

179 After Raman analysis, crystals were removed from CrystalBond™ using a hotplate and placed in Acetone.
180 They were then mounted on double-sided sticky tape with their polished side down. EpoFix™ resin was used to
181 impregnate the samples in a Cast-N-Vac vacuum pourer. After curing, the epoxy mount was polished using an
182 EcoMet30 automatic polisher, with 9, 3, and 1 µm diamond pastes. A reflected light map and image of each crystal
183 was taken using the Raman microscope to aid SEM sample navigation. The location within each FI within the
184 reflected light image was cataloged so the Scanning Electron Microscope (SEM) operator knew where to analyze
185 to obtain an approximate Fo content for each FI.

186

187 **EDS analysis**

188 Samples were carbon coated to an approximate thickness of 25–30 µm for EDS analysis. Chemical data
189 for each host crystal in the proximity of each FI was determined using a Zeiss EVO MA-10 SEM and a single
190 AMETEK EDAX 10 mm² detector at the University of California, Berkeley. The beam was rastered over a 30-by-
191 30 µm area for ~75–80s (a live time of 60 seconds with ~30% dead time). For all analyses we used an accelerating
192 voltage of 20 kV and a 30 µm aperture, giving an approximate beam current of 5 nA. EDS data reduction was
193 performed using an in-built standardless quantification routine (including a ZAF matrix correction), alongside pre-
194 determined “Standardless Element Coefficients” (SECs). The SECs act as correction factors for each element in
195 the standardless quantification routine and have been determined through several years of repeat analyses of

multiple different silicate standard materials and glasses. This method returns an estimate for the relative abundance of each element in the analyzed material and, if a normalization to 100% is assumed, can be used to return semi-quantitative chemical analysis of elemental or oxide weight percent values. However, for the purposes of this study we simply focused on the relative abundance of Mg and Fe in the EDS analyses to calculate the Fo content of the olivine host crystals. Furthermore, by calculating the molar Si/(Mg+Fe) ratio of each analysis we were able to provide a stoichiometric check of data quality: we obtained an average Si/(Mg+Fe) ratio of 0.497 ± 0.006 on Kīlauea olivine crystals, close to the ideal value of ~ 0.5 . Precision and accuracy were determined through repeat measurements of the San Carlos and Springwater olivines, which were not used as part of the standard quantification routine. We estimate the precision and accuracy of the method using repeat analyses of secondary standards (5 at start, 5 at end of day), which have Fo contents similar to our samples (see supplementary dataset S1). The Smithsonian-preferred Fo content²¹ for the San Carlos secondary standard is 90.2 Fo, and we obtained a mean of 89.84 ± 0.07 Fo units. For Springwater, the preferred value is 82.4 Fo, and we obtained a mean of 82.1 ± 0.2 Fo. We also analyzed a Kīlauea olivine crystal previously measured on the USGS Menlo Park EPMA. The average Fo content obtained at Menlo Park was 87.8 ± 0.1 Fo units, and at the University of California, Berkeley SEM, 88.5 ± 0.1 . It should be noted that such offsets also exist between different EPMA labs⁵. Considering these probable differences, we compared data acquired at Stanford University to that obtained at Cambridge University on the olivine crystals of ref¹⁴. The difference observed amounts to ~ 0.62 units at ~ 82 Fo and 0.78 units at 90 Fo¹⁰.

213

214 **EPMA analysis of glasses**

Major and minor element analysis of glass from a 11 September 2023 lava (sampled molten, water quenched) was done using the U.S. Geological Survey's JEOL 8530F microprobe at the California Volcano Observatory. Analyses used 15 kV accelerating voltage and a 10 μm beam with a 10 nA current. Peak counting times were 45 s for S, Cl, and F, 40 s for Ti, P, and Mn, 20 s for Si, Ca, Fe, Al, and Mg, and 10 s Na and K

219 (backgrounds were measured on both sides of the peak for half the peak counting times). Standards for glass
220 analyses were VG2 basaltic glass (USNM 111240/52;ref²¹) for Si, Mg, and Al, Kakanui Pyrope Garnet (USNM
221 143968) for Fe, and Al, Wollastonite for Ca, Tiburon Albite for Na, MnO₃ for Mn, TiO₂ for Ti, Orthoclase OR-1A
222 for K, Wilburforce Apatite (USGS-M105731) for P, Barite for S, Sodalite for Cl, and MgF₂ for F. Two-sigma
223 relative precision, based on two analyses of VG-2 glass (before and after lava sample was run), are 0.19 wt% for
224 SiO₂, 0.15 wt% for Al₂O₃, 0.003 wt% for TiO₂, 0.27 wt% for FeO, 0.009 wt% for MnO, 0.006 wt% for MgO, 0.04
225 wt% for CaO, 0.11 wt% for Na₂O, 0.02 for K₂O, 0.04 for P₂O₅, 0.07 for SO₃, 0.0001 for Cl, and 0.002 for F. X-ray
226 intensities were converted to concentrations using standard ZAF corrections²². Analyses with totals <99.0 wt% or
227 >100.5 wt% were rejected. Reported analyses are an average of four replicate points on individual glass fragments.

228

229 **Identifying and Resolving Bottlenecks**

230 The yellow stars on Fig. 1 identify current bottlenecks in the process that could be easily improved.

231

232 ***Star 1 - Shipping and receiving samples***

233 Distributing samples to the University of California, Berkeley was not a top priority for HVO because this
234 simulation was being attempted for the first time, and as a result, there was no guarantee of obtaining magma
235 storage depths in a timely manner. Samples were shipped from Hilo on a Friday at ~5 pm HST. HVO was asked to
236 ship samples to a private residence under the assumption that they might arrive over the weekend. However, no
237 packages leave Hilo after 4pm on Friday over the weekend, so the samples started their transit to California on
238 Monday. Had the package been taken to the courier's office on Friday morning, it would likely have arrived on
239 Sunday. The tracking information indicated arrival on Wednesday, which is when we planned to start the
240 simulation. However, the samples arrived at the private residence on Tuesday morning during working hours,
241 without notification that the package had been delivered (and no one was home).

242 We have demonstrated that this technique adds valuable quantitative depth information that expands on
243 HVO's routine near-real-time chemical monitoring with bulk rock ED-XRF¹. Under ideal circumstances, HVO
244 geologists would sample tephra or molten lava from the eruption on Day 1 (morning) and dry the samples in the
245 lab on Day 1 (afternoon), dropping the samples for shipment on the evening of Day 1, which would go out early
246 on Day 2 (as long as the drop off did not occur Friday afternoon or over the weekend). Same-day shipping from
247 Hawaii to California is not realistic, but samples shipping Monday through Thursday mornings would allow for
248 arrival on Day 3. Additionally, it would be possible to get samples to the University of California, Berkeley within
249 24 hours if someone in Hawai'i were to take a flight to San Francisco or Oakland airport with the samples, or within
250 ~30 hours if someone based in the University of California, Berkeley flew to Hawai'i, picked up the samples, and
251 returned home immediately.

252 We note that this bottleneck can be avoided entirely if observatories rely on local research expertise (for
253 example, in collaboration with local academic research groups) and/or establish in-house workflows for this type
254 of work. In such a case, next-day information could readily be obtained. As this is not an option for HVO, the
255 Hawaii-California connection will serve as the fastest way to conduct this rapid-response barometry.

256

257 *Star 2 – Sample cataloging*

258 The WITEC Raman microscope used in this study does not have a condenser in its optical path, which can
259 make it very hard to navigate and find FIs, particularly in volcanic crystals that are commonly coated in glass. The
260 first 7–10 crystals were analyzed immediately after preparation, so navigation took some time. After AB had
261 finished crushing, sieving, and picking, he began taking photos on a research-grade scope to help the Raman
262 operator find the FIs they were supposed to be analyzing. Late on Day 2, when students were not available, Wieser
263 began photographing crystals holding her phone to the eyepiece of the teaching-collection reflected light and
264 transmitted light microscopes. This provided enough textural context to easily find FIs on the Raman (Fig. S1 and

265 S2). The main advantage of using smartphones is that the person who found each FI could identify it, rather than
266 passing it off to another person who then must work out where the FI is before photographing it. This would greatly
267 reduce the number of people involved, as we almost always had one person taking photos.

268

269 ***Star 3 – Epoxy impregnation***

270 We used EpoFix™ epoxy, which is what was available in our lab (chosen because it gives low backgrounds
271 during SIMS analysis). After pouring the epoxy at ~7 pm, it was removed from its mount at ~9 am the next morning.
272 The epoxy was still noticeably soft (to the extent it cracked coming out of the mold). This meant that we could not
273 start polishing immediately. Instead, we had to wait a further ~5 hours for it to cure sufficiently to polish. If fast-
274 curing epoxies were available, it is very possible that a team member could have stayed, and polished and cataloged
275 the sample overnight, allowing SEM analysis on Friday (Day 3) rather than Saturday.

276

277 **Author contributions**

278 Author contributions for lab work are shown on Fig. 1. CD and PW wrote the paper. CD, PW, AR, BR,
279 and AB prepared tephra, picked olivine, found FIs, catalogued them, mounted them, and Raman-ed them. CD and
280 PW performed all spectral fitting, data processing, and figure making, with schematic illustrations shown in Fig. 1
281 from AB. JG developed the Mg/Fe calibration for the EDS detector and MG performed EDS analyses with help
282 from JG. KJL, DTD, NID and KMM collected samples, processed them in Hilo, and provided eruption context.
283 KJL and DD prepared the glass mount and did the EMPA analyses.

284

285 **Acknowledgements**

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Data availability

All data are made available with the publication. All 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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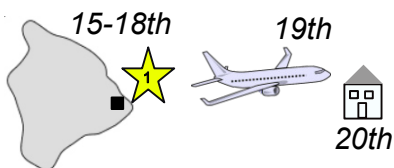
Sample erupted
10th Sept



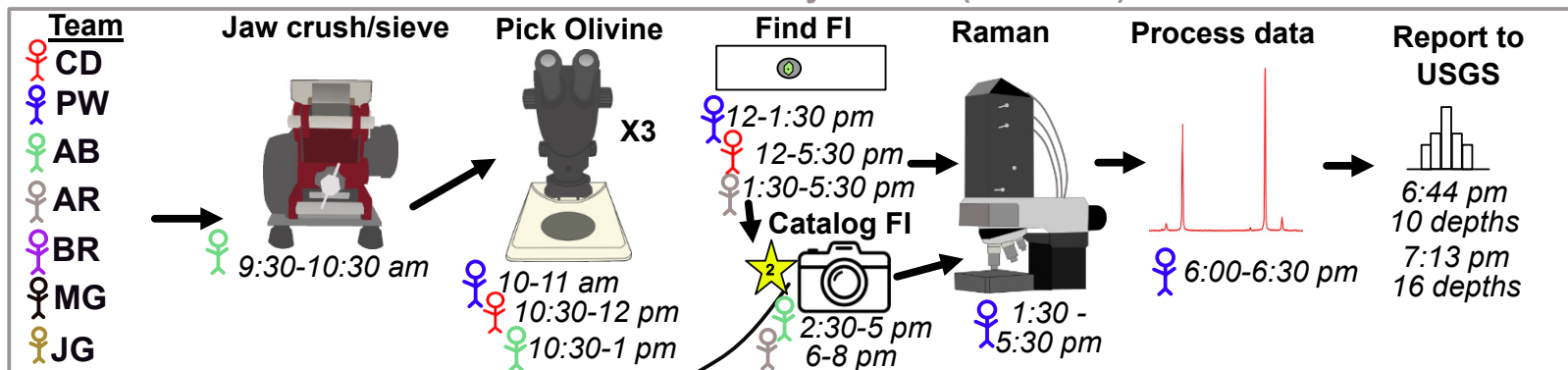
Sample collected
12th Sept



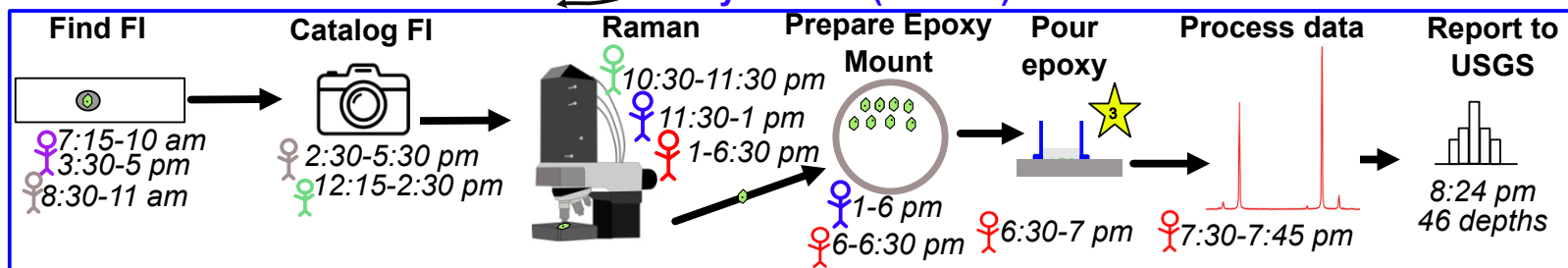
Sample taken to Hilo
(15th Sept)



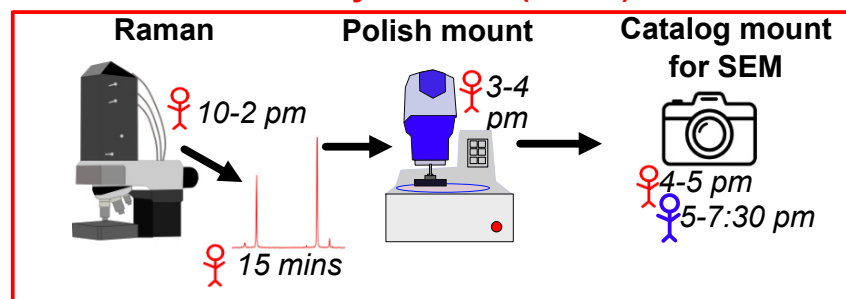
Day 1 - 20th (~26.5 hrs)



Day 2 - 21st (~23 hrs)



Day 3 - 22nd (~9hrs)



Day 4- 23rd (10.5 hrs analysis, 14 hrs report)

