Hydrothermal fluids and where to find them: Using seismic attenuation and anisotropy to map fluids beneath Uturuncu volcano, Bolivia

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Abstract

Mapping fluid accumulation in the crust is pertinent for numerous applications including volcanic hazard assessment, geothermal energy generation and mineral exploration. Here, we use seismic attenuation tomography to map the distribution of fluids in the crust below Uturuncu volcano, Bolivia. Seismic P-wave and S-wave attenuation, as well as their ratio (Q_P/Q_S) , constrain where the crust is partially and fully fluid-saturated. Seismic anisotropy observations further constrain the mechanism by which the fluids accumulate, predominantly along aligned faults and fractures in this case. Furthermore, subsurface pressure-temperature profiles and conductivity data allow us to identify the most likely fluid composition. We identify shallow regions of both dry and H_2O /brine-saturated crust, as well as a deeper supercritical H_2O /brine column directly beneath Uturuncu. Our observations provide a greater understanding of Uturuncu's transcrustal hydrothermal system, and act as an example of how such methods could be applied to map crustal fluid pathways and hydrothermal/geothermal systems elsewhere.

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15 Key Points:

- Seismic attenuation tomography can map crustal fluids and elucidate whether fluids are compressible, incompressible or supercritical
- S-wave velocity anisotropy suggests that crustal fluids at Uturuncu migrate and/or accumulate along fractures
 - Seismic attenuation combined with temperature, pressure and conductivity measurements can provide constraint on fluid composition

2223 Abstract

Mapping fluid accumulation in the crust is pertinent for numerous applications including
 volcanic hazard assessment, geothermal energy generation and mineral exploration. Here, we

- 26 use seismic attenuation tomography to map the distribution of fluids in the crust below
- 27 Uturuncu volcano, Bolivia. Seismic P-wave and S-wave attenuation, as well as their ratio 28 (Q_P/Q_S) , constrain where the crust is partially and fully fluid-saturated. Seismic anisotropy
- 28 (QP/QS), constrain where the crust is partially and fully huid-saturated. Seisine anisotropy 29 observations further constrain the mechanism by which the fluids accumulate, predominantly
- 30 along aligned faults and fractures in this case. Furthermore, subsurface pressure-temperature
- 31 profiles and conductivity data allow us to identify the most likely fluid composition. We
- 32 identify shallow regions of both dry and $H_2O/brine-saturated crust, as well as a deeper$
- supercritical H₂O/brine column directly beneath Uturuncu. Our observations provide a
- 34 greater understanding of Uturuncu's transcrustal hydrothermal system, and act as an example
- 35 of how such methods could be applied to map crustal fluid pathways and
- 36 hydrothermal/geothermal systems elsewhere.
- 37

38 Plain Language Summary

- 39 Locating where water/brines, gas and molten rock are in the crust is important various
- 40 applications, including assessing volcanic hazard, generating geothermal energy and
- 41 exploring for critical metals. Here, we map how seismic energy is absorbed (or attenuated) in
- 42 Earth's crust, in order to look for fluids in the subsurface. We do this at Uturuncu volcano,
- 43 Bolivia. This allows us to image whether the crust is partly or fully saturated with fluids. We
- 44 also use seismic anisotropy to help us understand how the seismic energy is absorbed. We
- 45 then use other data, including pressure, temperature and electrical conductivity data to
- 46 identify what fluids can be found where. We find that we can map where water/brines are and
- 47 whether they contain carbon dioxide (i.e. are "sparkling") or not (i.e. "still").
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50 1 Introduction

51 52 Fluids are present in much of Earth's crust. Mapping where and why these fluids 53 accumulate, as well as identifying their composition are critical questions in the earth sciences. For example, understanding where magma resides is important for volcanic hazard 54 55 assessment, mapping the extent of geothermal systems is pertinent for maximising production 56 efficiency, and identifying the location and properties of metal-rich brines is relevant for mineral exploration. The question that this study addresses is: can seismic attenuation 57 58 combined with seismic anisotropy be used to map the location of fluids and, in combination 59 with auxiliary data, identify fluid composition? Here, we test this hypothesis at Uturuncu 60 volcano, Bolivia.

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62 Uturuncu volcano sits within the Bolivian Andes. The volcano last erupted 250,000 63 vrs ago (Muir et al., 2015), yet has exhibited significant uplift at rates of up to 1 cm/yr (Gottsmann et al., 2018; Pritchard et al., 2018). Uturuncu lies ~20 km above the Altiplano-64 65 Puna Magma (or Mush) Body (APMB), Earth's largest body of silicic partial melt (Pritchard et al., 2018). This melt heats the crust above and potentially provides a source of ascending 66 metal-rich volatiles (Blundy et al., 2021). Uturuncu provides an ideal location for attempting 67 68 to image and identify fluids, since the host crust isolated from the volcanic system is likely 69 predominantly unsaturated except near surface rivers/lakes, while a shallow, partially-70 saturated hydrothermal system likely exists under the volcano that is sustained via heat and 71 volatiles from the APMB (Gottsmann et al., 2022).

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73 Here, we use microseismicity at Uturuncu (Hudson et al., 2022) to perform seismic 74 attenuation tomography and anisotropy analysis. Seismic attenuation is sensitive to the 75 presence of fluids, with tomography enabling any fluids to be mapped (e.g. Hauksson & Shearer, 2006). Attenuation studies of volcanoes have previously been undertaken (Bohm et 76 77 al., 2013; Caudron et al., 2019; Gudmundsson et al., 2004; Lanza et al., 2020; Lees, 2007; 78 O'Brien & Bean, 2009; Sanders et al., 1995; De Siena et al., 2014; Zhao, 2001). Seismic 79 anisotropy in volcanic or tectonic regions is predominantly sensitive to fault structures (e.g. 80 Baird et al., 2015), enabling one to identify whether fluids migrate and accumulate along volcanic and/or fault structures (Bacon et al., 2021; Baird et al., 2015; Gerst & Savage, 2004; 81 Johnson et al., 2011; Maher & Kendall, 2018; Nowacki et al., 2018). The novelty of this 82 83 work lies in using seismic attenuation and anisotropy to identify the dominant attenuation 84 mechanism and fluid saturation level. We then combine these results with auxiliary pressure, temperature and electrical conductivity profiles (Comeau et al., 2016; Pritchard et al., 2018), 85 86 to infer fluid composition and ascertain how fluids migrate and accumulate in the crust. 87

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89 2 Methods

2.1 Earthquake catalogue

92 The Uturuncu earthquake catalogue used in this study is from Hudson et al.
93 (2022). Seismicity is detected using the PLUTONS network (Kukarina et al., 2017),
94 which was deployed from the 13th April 2010 to the 27th October 2012. The
95 PLUTONS network comprised of thirty-three Guralp CMG-3T *120 s* seismometers
96 recording at 100 Hz. Earthquakes are detected using QuakeMigrate (Hudson et al.,
97 2019; Smith et al., 2020) and relocated using NonLinLoc (Lomax & Virieux, 2000).

The velocity model is the same as in Hudson et al. (2022). This produces a catalogue of 1356 earthquakes, which we use in this study (see Supplementary Figure 1).

101 2.2 Seismic attenuation

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Seismic attenuation is the loss of energy of a seismic wave into the medium during propagation. The amplitude of a seismic wave, A(x, f), at a position x with a frequency f, is defined by (Aki & Richards, 2002),

$$A(\mathbf{x}, f) = A_0(\theta, \phi)G(\mathbf{x})e^{-\alpha(f)r(\mathbf{x})}$$

where $A_0(\theta, \phi)$ is the source amplitude in the direction θ from vertical and ϕ from North, $G(\mathbf{x})$ is a geometrical spreading factor, $r(\mathbf{x})$ is the ray path length, and $\alpha(f)$ is the attenuation factor for a particular frequency. Therefore, the greater the value of α , the greater the attenuation.

In seismology, the convention is to quantify attenuation using the seismic quality factor, Q, the inverse of α , which is given by (Aki & Richards, 2002),

112 $O_i = \frac{\pi f}{1}$

$$Q_i = \frac{\gamma}{v_i \, \alpha(f)} \quad ,$$

where v_i is the velocity of phase *i* (P or S). Here, we approximate α , and therefore Q, to be frequency-independent over the bandwidth of the earthquakes studied (0.5 to 20 Hz).

2.2.1 Attenuation tomography

Attenuation tomography uses path-averaged attenuation observations, Q_{path}, between earthquake sources and receivers, to map the attenuation structure. The 3D attenuation tomography performed in this study uses the earthquakes and receivers shown in Supplementary Figure 1, with the ray paths shown in Supplementary Figure S2 and S3. Our method is based upon that described in Wei & Wiens (2018), with some alterations due to the local nature of our study area. A full description of the attenuation tomography methodology can be found in the Supplementary Information.

2.2.2 Q_P/Q_S ratios

P-wave attenuation is sensitive to scattering from faults and other velocity contrasts, as well as the intrinsic attenuation of the host rock and other compressible media, such as supercritical fluids and gases. S-wave attenuation is also sensitive to the scattering from fault structures and the presence of fluids, but in contrast to Pwaves is insensitive to the compressibility of fluids (Chapman, 2003; Chapman et al., 2021; Klimentos, 1995). Attenuation tomography therefore not only allows for mapping the presence of any fluids, but Q_P/Q_S ratios are also diagnostic of fluid saturation, with $Q_P/Q_S < 1$ indicating that rock is only partially-saturated and $Q_P/Q_S >$ 1 suggesting fully-saturated rock (Amalokwu et al., 2014; Hauksson & Shearer, 2006; K. Winkler & Nur, 1979). Here, we assume the concept of partially-saturated rock in its broadest sense, in that it simply has to have some fraction of compressible fluid or gas present (see Figure 4).

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2.3 Seismic anisotropy

Seismic anisotropy in this study refers to S-wave velocity anisotropy,
measured from shear-wave splitting. Such anisotropy can be broadly attributed to two
factors: crystallographic preferred orientation, where individual crystals of the

medium preferentially align; and shape-preferred orientation anisotropy, typically 147 caused by the preferential alignment of fractures (Kendall, 2000). As an S-wave 148 propagates through an anisotropic region, the energy will be partitioned into two 149 orthogonal components, one oriented in the plane of the fast-direction, ϕ , and the 150 other in the plane of the slow-direction. This phenomenon is called shear-wave 151 splitting (Crampin, 1981; Silver & Chan, 1991), measured here using SWSPy (152 Hudson, 2022). ϕ is controlled by the orientation of the fabric and/or fractures. The 153 arrival-time difference between the fast and slow S waves, δt , is controlled by the 154 155 strength and/or spatial extent of the anisotropy.

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159 3 Results160

The overall attenuation tomography results for Q_P , Q_S and Q_P/Q_S are shown in Figure 161 1. Tomographic resolution test results, based on the point-spread-function method 162 163 (Rawlinson & Spakman, 2016), are shown in Supplementary Figure S2 and S3. Regional QP values are $\sim 200-300$, while regional O_S values range from 200-400. These values fall within 164 the range observed at similar crustal depths elsewhere (Hauksson & Shearer, 2006; Lanza et 165 al., 2020; Del Pezzo et al., 1995). Regional Q_P and Q_S both generally increase with depth, as 166 expected for denser crust. A region of lower QP (high attenuation) extends in the SE-NW 167 direction, intersecting Uturuncu. This high attenuation region extends to depths of 8 km bsl. 168 Q_S follows the SE-NW trend observed in Q_P, except with more isolated pockets of lower Q_S 169 (high attenuation) rather than a continuous band. These isolated pockets occur on lateral 170 length scales of ~5-10 km and extend to depths of up to 3 km bsl. One exception is a highly 171 attenuating region ~ 15 km North of Uturuncu, at a depth of ~12 km bsl. However, it should 172 173 be noted that this feature is at the limit of the resolvable region (see Supplementary Figure S2 174 and S3).





Figure 1. Overall attenuation tomography results for an 80x80 km region surrounding Uturuncu volcano, Bolivia. (a) to (c): Map, WE and SN profiles, respectively, for Q_P . (d) to (f): Map, WE and SN profiles, respectively, for Q_S . (g) to (i): Map, WE and SN profiles, respectively, for Q_P/Q_S . Uturuncu is located at 0 km N, 0 km E. Map profiles are for 0 km bsl, and depth

180 profiles are for 0 km N and 0 km E of Uturuncu, respectively. Masked areas correspond to regions where features of at least

181 4 km size cannot be resolved (see Supplementary Figures 1 and 2 for resolution tests). Additionally, Q_P/Q_S results masked

182 where Q_P and/or $Q_S > 500$ (see Discussion for justification on masks).

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The majority of the region has $Q_P/Q_S < 1$ (see Figure 1g-i). A significant feature is a 184 column of $Q_P/Q_S \ll 1$ directly beneath Uturuncu's summit, extending from 1-2 km bsl to at 185 186 least 8 km bsl. This feature is shown in greater detail in Figure 2a-c (labelled A), where Q_P/Q_S is compared to resistivity tomography results from Comeau et al. (2016). Feature A 187 corresponds to a region of low resistivity (< 10 Ω m), indicative of high-salinity brines 188 189 (Afanasyev et al., 2018). Isolated features with $Q_P/Q_S > 1$ also exist. One such feature, beneath Cerro Loromayo (Fracchia, 2009; Soler et al., 2007), is shown in detail in Figure 2d-f 190 191 (labelled C). This feature also corresponds to a region of low resistivity, as do other $Q_P/Q_S >$ 192 1 regions.





Figure 2. Sections from the Q_P/Q_S tomography results, focussing on a $Q_P/Q_S < 1$ feature directly beneath Uturuncu and a $O_P/O_S > 1$ feature laterally offset to the South-East, beneath Cerro Loromayo. Results are overlaid with resistivity 198 tomography results from Comeau et al. (2016). (a)-(c) Map and vertical cross-sections centred about the summit of 199 Uturuncu. (d)-(f) Same as (a)-(c) except centred around a shallow, high QP/QS anomaly at Cerro Loromayo. Hashed 200 regions are where resistivity $> 10 \Omega$ m, for which any brines would be low salinity or not present (Afanasyev et al., 2018). 201 Labels A, to C are as referred to in Figure 4 and the Discussion. Results are masked as in Figure 1g-i.





Figure 3. Shear-wave velocity anisotropy results at Uturuncu volcano. (a) Rose diagram showing binned shear-wave velocity fast-directions and source polarisations from this study compared to fast-direction measurements from Maher and Kendall (2018) and fault strikes from Hudson et al. (2022). (b) Orientation of fast-direction for individual ray paths, plotted at ray mid-points of seismicity used in the shear-wave velocity anisotropy study. Lengths of the bars correspond to the delay times, \deltat. Volcano symbol indicates location of Uturuncu (0 km N, 0 km E) and green triangles are locations of receivers.

210 Seismic anisotropy observations at Uturuncu are shown in Figure 3. The histograms 211 in Figure 3a compare the orientations of the fast S-wave polarisation (yellow) to the source 212 polarisations derived during the S-wave splitting analysis (blue), as well as fault strikes from Hudson et al. (2022) (grey) and previously published seismic anisotropy results (red) (Maher 213 & Kendall, 2018). The fault strikes and source polarisations, which theoretically should be 214 oriented fault-parallel, are primarily oriented SW-NE and NNW-SSE. However, the fast-215 directions are predominantly oriented NNW-SSE, with a minority of fast-directions oriented 216 SW-NE lying to the north of Uturuncu (see Figure 3b). Data showing the orientation and 217 strength of anisotropy with depth are plotted in Supplementary Figure 4, which suggest that 218 219 the strength of anisotropy decreases as depth increases.

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222 4 Discussion

4.1 Attenuation mechanisms

225 In order to make physical interpretations of the attenuation tomography, one first has to identify the dominant attenuation mechanism. Seismic attenuation is 226 comprised of two contributions: intrinsic and extrinsic attenuation. Intrinsic 227 attenuation is controlled by bulk medium properties, while extrinsic attenuation is 228 typically controlled by features such as fractures. Methods for separating these two 229 contributions do exist, underpinned by the comparison of direct P- and S- waves to 230 highly-scattered coda using radiative transfer theory (Fehler et al., 1992; Hoshiba, 231 1991; Wang & Shearer, 2017; Wu, 1985). However, the application of such methods 232 are limited for the Uturuncu dataset since: isolating the P-wave coda from the direct 233 S-wave energy is challenging, thus making the separation of the dominant Q_P 234 mechanism impossible; the method is highly dependent on the coda window length 235 (Hoshiba, 1991); and results depend whether or not multiple scatterers and mode 236 conversions are modelled, especially in volcanic environments (Yamamoto & Sato, 237 2010). Therefore, we instead attempt to identify the likely dominant attenuation 238 mechanism, and what causes it, based on qualitative arguments. 239 240

Firstly, at depths shallower than 8 km bsl, high S-wave attenuation is confined to small pockets and high P- and S-wave attenuation are generally oriented in a narrow band with a strike NW-SE (see Figure 1). This band is parallel to the dominant fast-direction of the seismic anisotropy, as well as earthquake S-wave source polarizations and fault strikes. The high level of spatial variation in attenuation suggests that the dominant attenuation mechanism is due to interactions of the seismic waves with fault structures (i.e. extrinsic attenuation) rather than intrinsic attenuation, since the regional geology is unlikely to vary sufficiently to cause such high perturbations in intrinsic attenuation over such short length scales (Sparks et al., 2008). The attenuation structure deeper than 8 km bsl transitions to become approximately homogeneous, with anisotropy also decreasing with depth. This is likely partially due to resolution limits (see Supplementary Figure 2), but possibly also because at these depths the crust is too ductile to sustain sufficient fault structures required to promote significant scattering.

255 Assuming that the mechanism for the high attenuation regions is governed by 256 257 the presence of fault structures, then a further question is what specifically causes the loss of seismic energy as the seismic waves interact with the faults. Fractures can 258 scatter seismic energy due to reflection off the fracture interfaces (Zhu et al., 2007). 259 260 Fluids in the pore space and along fractures can also cause attenuation due to mechanisms such as squirt flow (Chapman, 2003; Chapman et al., 2002). We suggest 261 that fluids play an important role in facilitating the observed attenuation structure for 262 263 the following reasons. Firstly, there are likely fluids ascending from the APMB, which sits directly beneath Uturuncu. The seismicity above the APMB has seismic b-264 values > 1, consistent with fluids reducing fault normal stresses (Hudson et al., 265 266 2022). Further evidence is rooted in the observation that the high attenuation structures are oriented parallel to one set of fault orientations but not the other. There 267 is evidence from both S-wave source polarisations and fault strikes that two sets of 268 269 fault orientations exist, yet there is only one dominant direction exhibited in the seismic anisotropy and Q_P attenuation. We suggest that the crustal stress regime 270 facilitates preferential opening of faults in one direction and closing of faults in the 271 other, presumably governed locally (< 10 km from Uturuncu) by deformation at the 272 volcano (Gottsmann et al., 2018; Pritchard et al., 2018) and regionally by tectonic 273 stresses. This stress field would facilitate the ascent of fluids preferentially along one 274 set of faults, causing the observed trend in anisotropy and attenuation. Additionally, a 275 276 column of high P-wave attenuation (~5 km diameter, 10 km high) lies directly 277 beneath Uturuncu, extending towards the APMB, with a column potentially observed in the S-wave attenuation too. The location of this column is in close agreement with 278 279 a hypothesised route for fluid ascent (Gottsmann et al., 2017; Del Potro et al., 2013; 280 Pritchard et al., 2018). Based on the above evidence, we suggest that the observed attenuation structures are predominantly caused by faults with fluids ascending and/or 281 282 accumulating along them.

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4.2 Identifying and mapping fluids and their composition

Assuming that the observed attenuation is primarily caused by fluids within faults, then relevant questions are what the fluid composition is and does it vary spatially. For simplicity we consider only three representative fluids: CO₂; H₂O (or analogous brines); and molten rock. Diagnostic data for addressing these questions are Q_P/Q_s ratios, which indicate whether the crust contains a proportion of fluids or gases

that are compressible $(Q_P/Q_S < 1)$ or the crust is fully-saturated with incompressible fluids $(Q_P/Q_S > 1)$ (see Section 2.4 and Amalokwu et al. (2014); Chapman et al. (2021); Hauksson & Shearer (2006)). Additionally, pressure and temperature profiles, as well as resistivity data, can be used in combination with Q_P/Q_S to constrain fluid composition spatially. Figure 4 shows a schematic diagram summarising an inferred map of different fluids at Uturuncu based on the following discussion. Figure 4 also includes crustal pressure and temperature profiles (Pritchard et al., 2018 and references therein) and schematic compressibility and conductivity profiles that are used to further inform this discussion.



Figure 4. Summary of our interpretations based on the seismic data presented in this study, as well as auxiliary data that informs our interpretation. a) Approximate pressure (black line) and temperature (red dashed lines) profiles with depth, based on data from Figure 2 of Pritchard et al. (2018) and references therein. Lithostatic pressure is calculated from density. Conditions where single-phase CO_2 and H_2O become supercritical are shown in light and dark blue, respectively. Conditions where partially molten dacite can be sustained are shown in orange, based on the dacite solidus calculated from the average of the temperature profiles (Holtz et al., 2005; Pritchard et al., 2018). b) Schematic depth profile of how fluid compressibility varies with depth. Compressibility profiles based partially on behaviour of CO_2 (Zhang et al., 2020). c) Same as (b) but for conductivity, based partially on two-phase brine properties with increasing pressure (Watanabe et al., 2021). Conductivity of magma from (Laumonier et al., 2017). d) Schematic diagram summarising the key interpretations for different gas/fluid-filled fractures and the presence of brines beneath Uturuncu volcano.

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| 314 | Below sea-level, features are less well resolved by the attenuation tomography |
| 315 | than at shallower depths. However, one feature we are confident of is the $Q_P/Q_S \ll 1$ |
| 316 | column directly beneath Uturuncu, labelled A in Figure 4 (also see Figure 2a-c), |
| 317 | which extends from ~2 to 8 km bsl. This region of crust is highly faulted (Hudson et |
| 318 | al., 2022), with O_P/O_S implying that these faults contain at least some compressible |
| 319 | fluid component. Given the lithostatic pressures, it is unlikely that any gaseous phase |
| 320 | is present at these depths. Intriguingly, H ₂ O becomes supercritical at these depths (see |
| 321 | Figure 4a), driving the compressibility up and therefore O_P/O_S down (see Figure 4b). |
| 322 | This feature has resistivities $< 10.0 m$ (Comean et al. 2016) consistent with high |
| 322 | salinity brines (Afanasyev et al. 2018) (see Figure 4c) and not molten rock. These |
| 323 | conductivities also rule out this feature being solely comprised of supercritical CO ₂ |
| 324 | (see Figure 4c). Furthermore, the sudden transition from a weak $\Omega_{\rm p}/\Omega_{\rm s} < 1$ signal to a |
| 323 | (see Figure 4c). Furthermore, the sudden transition from a weak $QP/QS < 1$ signal to a strong $\Omega_2/\Omega_2 < 1$ signal at 2 km hall where HeO/brings become supercritical |
| 520 227 | strong $Qp/Qs << 1$ signal at ~2 km osi, where $\Pi_2O/Ormes become supercritical,$ |
| 5Z7 | However, this region of emet could also contain CO or other valetiles in |
| 328 | However, this region of crust could also contain CO_2 , or other volatiles, in |
| 329 | combination with brines. |
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| 331 | At depths shallower than sea-level, Q_P/Q_S ratios vary <1 (red regions, Figure |
| 332 | 2, labelled B in Figure 4) and >1 (blue regions, Figure 2, labelled C in Figure 4), |
| 333 | indicating that some regions of the shallow crust are partially saturated and others are |
| 334 | fully saturated. This observation is expected <1 km from the surface, since this region |
| 335 | of Bolivia is arid, yet surface snow and H_2O are present in places, which likely |
| 336 | percolate into the shallow crust. However, saturated regions of the crust extend to |
| 337 | depths at sea-level (>4 km below surface), too deep for groundwater aquifers. We |
| 338 | interpret the regions in the vicinity of Uturuncu's summit with $Q_P/Q_S < 1$ and |
| 339 | corresponding low resistivities (see label B, Figure 4b,d) as a 2-phase brine-CO ₂ |
| 340 | mixture, which could be referred to as "sparkling CO ₂ ". These brines and CO ₂ likely |
| 341 | ascend from the APMB, via feature A, along faults in the crust. |
| 342 | |
| 343 | Regions of $Q_P/Q_S > 1$, implying crust saturated with incompressible fluids, |
| 344 | typically lie away from Uturuncu's summit. One particularly obvious example is that |
| 345 | focussed on in Figure 2d-f, labelled C in Figure 4d. Again, the feature corresponds |
| 346 | with low resistivities, indicative of high-salinity brines. This feature is therefore |
| 347 | interpreted to be brine-saturated crust. Furthermore, this feature is particularly |
| 348 | interesting as it lies beneath Cerro Loromayo (Fracchia, 2009; Soler et al., 2007), a |
| 349 | volcano that could have exhibited activity as late as 0.9 Ma (Fracchia, 2009). Based |
| 350 | on our Q_P/Q_S results, this feature appears isolated, with no vertical column-like |
| 351 | feature below it. Furthermore, it corresponds to a low-density gravity anomaly |
| 352 | (MacOueen et al., 2021), indicative of fluid accumulation. We therefore suggest that it |
| 353 | is a brine-lens (Afanasyev et al., 2018) that was formed when Cerro Loromavo had an |
| 354 | active hydrothermal system, but has subsequently cooled and is no longer fed by CO ₂ |
| 355 | or other volatiles from the APMB. Features such as this are exciting because they |
| 356 | could potentially be a well-endowed accumulation of metal-rich brines (Blundy et al |
| 357 | 2021) |
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| 360 | 4.3 The bigger nicture |
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361 Mapping what fluids accumulate where in the subsurface is important for 362 numerous applications, including: volcanic hazard assessment; efficiently exploiting

| 363 | geothermal systems; and in searching for brines rich in metals critical for a green |
|-----|---|
| 364 | energy transition (Blundy et al., 2021). For volcanic hazard assessment, it is important |
| 365 | to discriminate between hydrothermal and partial melt storage regions in order to |
| 366 | accurately discriminate the volume of melt at a given volcano. Q _P /Q _S combined with |
| 367 | temperature and pressure profiles makes this possible. Theoretically, our approach |
| 368 | may also allow one to measure the proportion of melt, if densities are adequately |
| 369 | constrained. For geothermal system characterisation, Q_P/Q_S can elucidate and map |
| 370 | fluid-saturated crust vs. potentially dry crust, providing improved spatial constraint |
| 371 | when targeting geothermal prospects. This constraint will reduce geothermal |
| 372 | exploration risk. Similarly, Q_P/Q_S can also benefit the endeavour to find new, |
| 373 | sustainable mineral resources in the form of metal-rich brines. Q _P /Q _S , in combination |
| 374 | with temperature, pressure, density and conductivity profiles (Comeau et al., 2016; |
| 375 | Del Potro et al., 2013; Ward et al., 2014), not only allows for the spatial constraint of |
| 376 | metal-rich brine accumulation but also potentially the volume fraction of these brines |
| 377 | within the crust. This information is critical for maximising the success of extracting |
| 378 | metal-rich brines from geothermal systems going forward. |
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| 381 | 4.4 Limitations |
| 382 | A fundamental limitation of any tomographic method is the spatial resolution. |
| 383 | This spatial resolution is governed by the density of ray path coverage, which is |
| 384 | shown in Supplementary Figure S1. The P-wave coverage is significantly better than |
| 385 | the S-wave coverage, owing to the greater number of picked P-wave arrivals. The Q _S |
| 386 | and Q _P /Q _S tomography results are therefore limited, both in overall spatial extent and |
| 387 | the minimum size of feature that can be resolved. The Q_P/Q_S results in Figure 3 are |
| 388 | masked for regions where we are not confident that we can resolve features greater |
| 389 | than 2 km in size. Significantly, we cannot observe the APMB with any confidence, |
| 390 | primarily because only a small number of earthquakes from below the APMB are of |
| 391 | sufficient quality to use in the attenuation tomography inversions. The resolution of |
| 392 | the tomography could be improved by detecting more S-waves and/or increasing the |
| 393 | number of stations in the network. |
| 394 | |
| 395 | Another potential limitation is that we invert for Q_{P}^{-1} and Q_{S}^{-1} separately, |
| 396 | producing our Q_P/Q_S tomographic map by division of each unit of the tomographic |
| 397 | model individually, as in other studies (e.g. Hauksson & Shearer, 2006). However, |
| 398 | this division method has two potential issues. Firstly, both Q_{P}^{-1} and Q_{S}^{-1} can take |
| 399 | near-zero values for low-attenuation regions, which could lead to large variations in |
| 400 | Q_P/Q_s . Secondly, even if the regularisation and smoothing parameters happen to be |

401 equal for both the Q_P and Q_S inversions, the resolution and associated uncertainty in 402 the tomography results may differ. These issues can be mitigated by performing a direct inversion for Q_P/Q_S . We do not perform a direct inversion for Q_P/Q_S , as the 403 complex velocity structure means that one cannot assume the same ray-path for a 404 given P-wave and S-wave from the same source to the same receiver, a required 405 condition if inverting directly for Q_P/Q_S using our method. Others have avoided this 406 limitation by inverting for Q_P/Q_S by fixing Q_P using the Q_P tomography solution and 407 then only varying Qs in the inversion (Wei & Wiens, 2020). However, for our dataset, 408 absolute values of Q_P/Q_S are highly dependent on the starting model. Instead, to 409 maximise confidence in our direct division-derived Q_P/Q_S results, we mask out 410 regions that have small Q_P^{-1} and/or Q_S^{-1} (Q>500) to minimise any near-zero division 411 issues. We also minimise the effect of differing Q_{P}^{-1} and Q_{S}^{-1} resolution and 412

413 uncertainties by masking out regions where either 4 km size Q_P or Q_S features cannot 414 be resolved (see Supplementary Figures 1 and 2). We therefore have confidence in the 415 remaining Q_P/Q_S results presented in Figure 2 and Figure 4.

A final limitation of note is the ambiguity of Q_P/Q_S ratios for identifying fluid 417 type and the exact ratio of gas to liquid. Here, we require temperature and pressure 418 419 profiles to identify the most likely fluid/s associated with a given Q_P/Q_S ratio. However, pressure and temperature profiles may well be inaccurate, especially as they 420 only describe variations in one-dimension, depth. V_P, V_S and V_P/V_S results could 421 422 provide additional constraint, although velocity is less sensitive to fluid saturation than attenuation (Winkler & Nur, 1982). Other geophysical measurements such as 423 density (Del Potro et al., 2013; Ward et al., 2014) would provide additional constraint, 424 which in combination with other parameters might constrain porosity. However, one 425 particularly useful observation would be resistivity tomography (Comeau et al., 426 2016), which would aid constraint of the presence of conductive brines vs. resistive 427 gases. Ideally, one would perform a joint inversion, including all these parameters to 428 simultaneously constrain additional parameters, such as porosity, conductivity and 429 density, in order to identify fluid composition and prevalence better. 430

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416

432433 5. Conclusions

We present seismic attenuation and anisotropy results at Uturuncu volcano, Bolivia. 434 435 3D attenuation tomography shows higher-than-background P-wave attenuating structures that 436 align parallel to the orientation of fractures (NNW-SSE). The presence and orientation of these fractures is evidenced by seismic anisotropy. Higher-than-background S-wave 437 438 attenuation is isolated to smaller localities, sporadically located along the strike of the high Pwave attenuation regions. Q_P/Q_S ratios, indicative of crustal fluid saturation, show that most 439 of the crust is partially saturated, with only a few pockets of fully-saturated crust. We 440 interpret saturated crust above sea-level to be brine saturated crust in its normal state, likely a 441 brine lens that developed during active volcanism that has now ceased. A column of 442 particularly low Q_P/Q_S directly beneath Uturuncu is interpreted to comprise of supercritical 443 444 fluids, most likely H₂O or metal-rich brines, potentially containing volatiles. This likely feeds CO₂ and other volatiles into a shallow hydrothermal system. 445

446

447 We show that high seismic attenuation features can be attributed to fluid accumulation 448 along faults, if constrained by other observations such as seismic anisotropy. Furthermore, if 449 one has constraint over pressure and temperature profiles through the crust, as well as data 450 constraining crustal resistivity, then it is possible to identify the most likely fluid 451 compositions responsible for each seismic attenuation signature. We therefore conclude that 452 fluid accumulation and composition can be mapped seismically. Such observations at other locations could be used for a range of applications, from volcanic hazard assessment to the 453 exploration of metal-rich brine deposits that could potentially be exploited to facilitate the 454 green-energy transition. 455 456

456 457

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- 463 path-average attenuation for each earthquake is SeisSrcMoment (<u>Hudson, 2020</u>), available
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| 2 | AGU PUBLICATIONS |
| 2 | Geophysical Research Letters |
| 4 | Supporting Information for |
| 5 6 | Hydrothermal fluids and where to find them: Using seismic attenuation an anisotropy to map fluids beneath Uturuncu volcano, Bolivia |
| 7 8 | T.S. Hudson ¹ , JM. Kendall ¹ , J.D. Blundy ¹ , M.E. Pritchard ² , P. MacQueen ² , S.S. Wei ³ , J.H. Gottsmann ⁴ , S. Lapins ⁴ |
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| 15 16 17 18 | Contents of this file Text S1 to S2 Figures S1 to S5 |
| 19 | |
| 20 | Introduction |
| 21 22 23 | Here, we provide exact details of how the attenuation tomography and S-wave velocity anisotropy analysis was performed. We also include supplementary figures showing: the earthquake catalogue, tomography resolution tests, the magnetotelluric data from the literature, and |

additional seismic anisotropy results.

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Supplementary Text 1: Attenuation tomography

36 37 Attenuation tomography comprises of using path-averaged attenuation observations, 38 Q_{path}, between earthquake sources and receivers, to map the attenuation structure. The 3D 39 attenuation tomography performed in this study is undertaken using the earthquakes and 40 receivers shown in Figure 1, with the ray paths shown in Supplementary Figure S1. Our method is 41 based upon that described in (Wei & Wiens, 2018) with some alterations, required due to the local 42 nature of the study area. The critical observation we require to perform this inversion is the pathaveraged attenuation, \bar{Q}_{path} , which is related to t*, given by, 43

 $t_i^* = \frac{t_i}{\bar{Q}_{nath\,i}} \quad ,$ 44

45 where t_i is the travel time of the seismic phase from the source to the receiver for earthquake *i*. We 46 obtain t*, and hence \bar{Q}_{path} , for each earthquake by calculating the displacement spectrum of the earthquake. We then assume that the source can be described by the Brune model (Brune, 1970), 47

48 which is given by, Ω

49

$$\Omega(f) = \frac{\Omega_0 e^{-\pi f t^*}}{\left(1 + \left(\frac{f}{f_c}\right)^2\right)}$$

,

where **f** is the frequency, f_c is the corner frequency and Ω_0 is the long-period spectral amplitude. 50 51 To find t*, we vary the parameters f_c , t*, and Ω_0 to find the Brune model that best matches the

52 observed displacement spectrum for each earthquake source-receiver pair. 53

54 The observed t* values for all source-receiver pairs can then be used to perform a 55 tomographic inversion to image the attenuation structure. The equation describing this 56 tomography inversion is given by (Wei & Wiens, 2018),

- $t_i^* = \sum_{i=1}^{j=n_{nodes}} \frac{l_{ij}}{v_j} \frac{1}{Q_j} = G_{ij}m_j$, 57
- 58 where n_{nodes} is the number of nodes in the 3D grid, I_{ij} is the path length for a ray associated with 59 source-receiver pair i through node j, v_i is the seismic velocity of node j, Q_i is the quality factor 60 measure of attenuation in node j, G_{ii} is the tomography tensor component associated with the 61 source-receiver pair i and node j, and m_j is the tomography model attenuation for node j, where $m_j = \frac{1}{Q_i}$. The above equation can be rewritten in tensor notation as, 62

63 $t^* = G.m$, 64 where t^* is a vector of length n_{pairs}, the number of source-receiver pairs, G is a second order tensor, 65 and m is a vector of length n_{nodes}. We can solve this equation to find the model attenuation vector, 66 $m_{\rm r}$ by performing a regularised linear least-squares inversion. This attenuation tomography

67 method can be applied to P or S seismic phases to obtain Q_P or Q_S, respectively. 68

69 The above theory is implemented practically through the specific attenuation tomography 70 steps as follows:

71 1. First we calculate the t_i^* measurements for each source-receiver pair to find t^* . To do this 72 we take a window around the P or S phase and compute the observed displacement 73 spectrum using the multi-taper spectrum method of Krischer (2016) and Prieto et al. 74 (2009). We then find the best-fitting Brune model to find the best estimates of $f_{c,i}$, t_i^* , and 75 $\Omega_{0,i}$ for that source-receiver path. We repeat this for every source-receiver path to find t^* . 76 The Brune model does not always adequately fit the observed displacement spectrum. We 77 filter out poor fits by removing source-receiver pairs associated with events that have a standard deviation in \bar{Q}_{path} , $\sigma_{\bar{Q}_{path}}$, greater than 400. 78

- 79 2. We then specify the 3D grid nodes for the tomographic inversion. We use a 120 km by 120 80 *km* by *60 km* grid with a uniform grid spacing of 1 km. We include the receivers inside the 81 grid, with the adjacent nodes implicitly accommodating any local site effects in the 82 tomographic inversion.
- 83 3. We then perform ray tracing to calculate the path lengths, l_{ii}, through each cell. We use the 84 ray tracing code (Nasr et al., 2020), which uses the fast marching method (Lelièvre et al., 85 2011; Nicholas Rawlinson & Sambridge, 2005).
- 4. The final component we require to perform the tomographic inversion is the tomography 86 tensor, G. This second order tensor is given by, 87

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$$\boldsymbol{G} = \begin{pmatrix} \frac{l_{11}}{v_1} & \cdots & \frac{l_{1j}}{v_j} \\ \vdots & \ddots & \vdots \\ \frac{l_{i1}}{v_1} & \cdots & \frac{l_{ij}}{v_j} \end{pmatrix}$$

- 5. We can then perform the regularised linear least squares inversion, which involves minimising the function,

 - $f(m) = \|Gm t^*\|_2^2 + \lambda \|m\|$, where λ is the regularisation coefficient. To find the optimal regularisation so as to avoid under- or over-fitting, we perform the inversion for multiple values of λ and select the value of λ at the corner of the L-curve. This provides us with the best fitting attenuation model **m**.
- 6. The best fitting attenuation model may have physically unrealistic jumps in attenuation between individual cells. We account for this by applying smoothing to the model to produce a final, realistic attenuation model. There are various ways of applying smoothing to a tomography model. We apply 2D Gaussian smoothing over a wavelength of 1 km to each xy-plane layer in the 3D model. We apply lateral rather than vertical 2D smoothing since we are primarily interested in the attenuation variation with depth, and so do not want to introduce any unnecessary bias to the tomography result in this axis.

104 The steps to obtain t* measurements are implemented in the open source python code 105 SeisSrcMoment (Hudson, 2020).

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108 Supplementary Text 2: Seismic anisotropy

109 110 Seismic anisotropy in this study refers to S-wave velocity anisotropy. Such anisotropy can 111 be broadly attributed to two factors: crystallographic orientation, where individual crystals of the 112 medium preferentially align; and bulk-fabric anisotropy, typically caused by the preferential 113 alignment of fractures that may be fluid-filled. Seismic anisotropy manifests itself as follows. An S-114 wave radiated from an earthquake source will have an initial polarisation. For a double-couple 115 source, this polarisation is parallel to the slip-direction of the fault generating the earthquake. If a 116 region of the crust is seismically anisotropic, then as the S-wave propagates through this region, 117 the energy will be partitioned into two orthogonal components, one oriented in the plane of the 118 fast-direction, ϕ , and the other in the plane of the slow-direction. Stronger anisotropy or a longer ray path through the anisotropic region will result in greater delay-times between these fast and 119 120 slow S-waves . This phenomenon is called shear-wave splitting (Crampin, 1981; Silver & Chan, 121 1991). Shear-wave splitting is measured using SWSPy (Hudson, 2022), which is based on the 122 eigenvalue method described in Teanby et al. (2004) and Walsh et al. (2013). The strength of 123 anisotropy is quantified by the delay-time, δt . However, it is also useful to measure the strength of 124 anisotropy, a, as the magnitude of splitting normalised by the distance travelled (Thomas & 125 Kendall, 2002). If the delay time is defined by,

 $\delta t = t_{slow} - t_{fast} = \frac{d}{\bar{v}\left(1 - \frac{a}{2}\right)} - \frac{d}{\bar{v}\left(1 + \frac{a}{2}\right)} \quad ,$ where d is the ray path length and \bar{v} is the mean velocity of the medium, then the magnitude of anisotropy is given by (Thomas & Kendall, 2002),

$$a = \delta v_S = -\frac{2d}{\bar{v}\delta t} \pm \sqrt{\left(\frac{2d}{\bar{v}\delta t}\right)^2 + 4}$$

Supplementary figures





Supplementary Figure 1. Seismicity used in this study. Blue inverted triangles show seismometer locations. Red scatter points show earthquake hypocentres, coloured by uncertainty in depth.



139 140 Supplementary Figure 2. QP tomography sensitivity analysis. Sensitivity analysis performed based on the theory in Rawlinson & Spakman (2016). 141



Supplementary Figure 3. Qs tomography sensitivity analysis. Sensitivity analysis performed based
 on the theory in Rawlinson & Spakman (2016).



148 149 **Supplementary Figure 4**. Magnetotelluric tomography results from Comeau et al. (2016).



151 **Supplementary Figure 5.** Shear-wave velocity anisotropy with depth. (a), (b), (c) Fast directions,

- delay-times and fractional-change in S-wave velocity with depth, respectively. Relative sizes of
- scatter points are determined by their associated anisotropic quality factor, Q_w. Red dashed lines in
- 154 (b), (c) indicate the approximate decreasing trend in delay-time and velocity-change with depth.