

# Geodynamic, geodetic, and seismic constraints favour deflated and dense-cored LLVPs

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## Key Points:

- Observed geoid-to-topography ratio sets effective  $\sim 900$  km upper limit on vertical extent of LLVP buoyancy.
- Dynamic topography, geoid, CMB ellipticity, body tides, and Stoneley modes are consistent with dense material in basal 100—200 km of LLVPs.
- This layer is most likely composed of iron- and silicon-enriched crustal material that formed early in Earth's history.

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**Abstract**

Two continent-sized features in the deep mantle, the *large low-velocity provinces* (LLVPs), influence Earth’s supercontinent cycles, mantle plume generation, and its geochemical budget. Seismological advances have steadily improved LLVP imaging, but several fundamental questions remain unanswered, including: What is the true vertical extent of the buoyancy anomalies within these regions? And, are they purely thermal anomalies, or are they also compositionally distinct? Here, we address these questions using a comprehensive range of geophysical observations. The relationship between measured geoid anomalies and long-wavelength dynamic surface topography places an important upper limit on the vertical extent of long-wavelength LLVP-related density anomalies of  $\sim 900$  km above the core-mantle boundary (CMB). Instantaneous mantle flow modelling suggests that anomalously dense material must exist at their base to simultaneously reproduce geoid, dynamic topography, and CMB ellipticity observations. We demonstrate that models incorporating this dense basal layer are consistent with independent measurements of semi-diurnal Earth tides and Stoneley modes. Our thermodynamic calculations indicate that the presence of early-formed, chondrite-enriched basalt in the deepest 100–200 km of the LLVPs is most compatible with these geodynamic, geodetic, and seismological constraints. By reconciling these disparate datasets for the first time, our results demonstrate that, although LLVPs are dominantly thermal structures, their basal sections likely represent a primitive chemical reservoir that is periodically tapped by upwelling mantle plumes.

**Plain Language Summary**

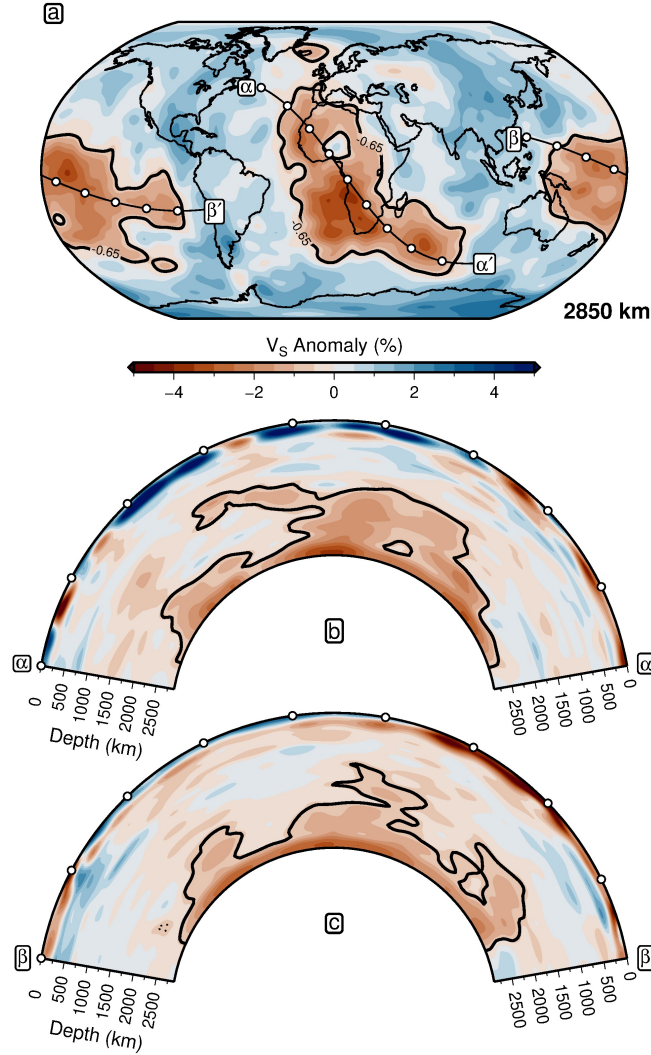
Images of Earth’s deep mantle—constructed by analysing earthquake-triggered waves—reveal two large dome-like regions on top of the core-mantle boundary that slow down such waves. Known as ‘large low-velocity provinces’ (LLVPs), these enigmatic features are thought to influence fundamental aspects of Earth’s evolution, including supercontinent cycles, the distribution of ‘hotspot’ volcanism, and the stability of the magnetic field. Despite decades of research, key questions around their vertical extent and composition remain unresolved, with studies coming to divergent conclusions depending on the data they use. In this study, we integrate a wide suite of recent geodynamic, geodetic, and seismological observations with numerical modelling to gain a new, unified understanding of LLVPs. We find that they extend no more than 900 km above the core-mantle boundary, that the deepest 100–200 km of these regions contain anomalously dense material, and that this material most likely represents fragments of thick crust that formed early in Earth’s history (4 billion years ago) and may have been capped by a meteorite-derived debris layer. These results suggest that, while LLVPs are mainly thermal features, their deepest portions represent a stable, early-formed chemical reservoir that is occasionally sampled by plumes of hot rock ascending from the core-mantle boundary.

**1 Introduction**

Seismic tomographic models consistently image two large regions of slow seismic velocity in the deep mantle that are widely interpreted to be hotter than ambient material and are spatially correlated with positive, long-wavelength geoid height anomalies (Figures 1 and 2a; Garnero et al., 2016). Early mantle flow studies treated these features as buoyant upwellings and found that an increase of mantle viscosity with depth is required to obtain satisfactory model fits to observed non-hydrostatic geoid height anomalies (Hager et al., 1985; Ricard et al., 1993). Nevertheless, these instantaneous flow calculations are non-unique and suffer from trade-offs between the magnitude and distribution of excess buoyancy. While there is emerging consensus on the lateral extent of LLVPs (e.g., Cottaar & Lekic, 2016), numerous controversies remain concerning their structure and composition.

First, body wave coverage in the mid-to-lower mantle ( $\sim 1000$ – $2500$  km depth) is limited, with most ray paths traversing this region near-vertically, making global tomographic

71 models susceptible to smearing artefacts in this depth range (Ritsema et al., 2007; Koele-  
 72 meijer et al., 2018; Maguire et al., 2018). The true vertical extent of thermochemical het-  
 73 erogeneity associated with LLVPs is therefore uncertain, with recent studies suggesting that  
 74 laterally extensive low-velocity structures imaged at depths  $\leq 2000$  km may actually repre-  
 75 sent tomographic aliasing of clusters of distinct plumes (Davaille & Romanowicz, 2020).



**Figure 1. Spatial extent of seismically imaged LLVPs.** (a) Shear-wave velocity ( $V_S$ ) anomalies at 2850 km depth in the TX2011 seismic tomographic model (Grand, 2002), which is used throughout this study. Thick black contour =  $-0.65\%$   $V_S$  anomaly threshold used to delineate LLVP boundary (Burke et al., 2008);  $\alpha-\alpha'$  and  $\beta-\beta'$  = cross-section locations with white circles spaced at 1000 km intervals. (b) Cross-section  $\alpha-\alpha'$  beneath Africa through blended tomographic model (SLNAAFSA above 300 km, TX2011 below 400 km, linearly interpolated between 300–400 km; Schaeffer & Lebedev, 2013, 2014; Celli, Lebedev, Schaeffer, Ravenna, & Gaina, 2020; Celli, Lebedev, Schaeffer, & Gaina, 2020; Hoggard et al., 2020; Grand, 2002). (c) Cross-section  $\beta-\beta'$  beneath Pacific Ocean.

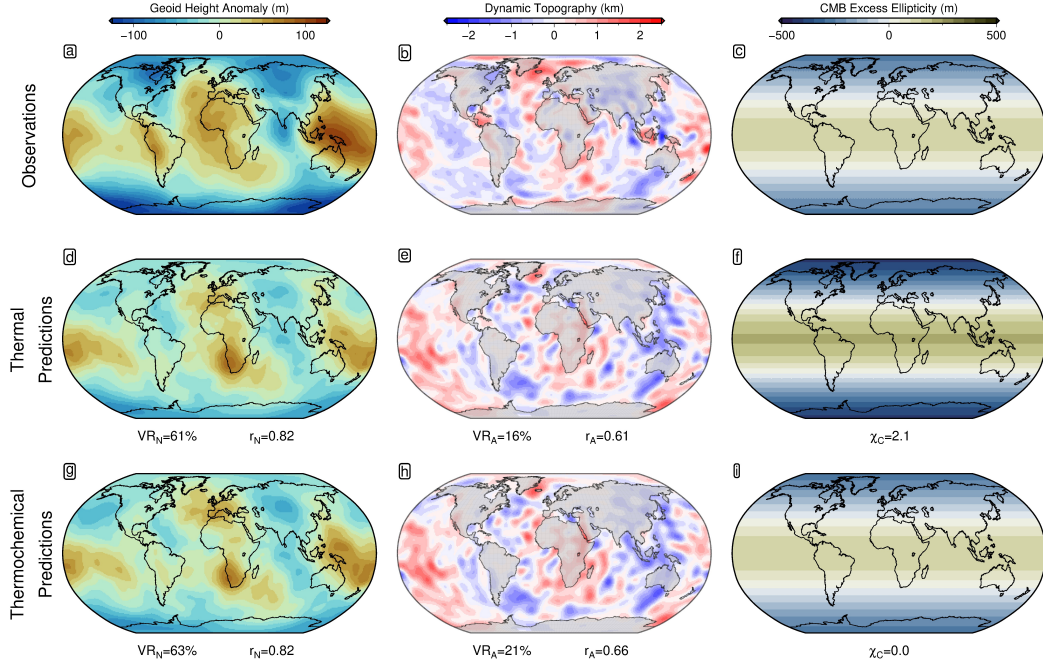
76 Second, considerable debate remains over whether LLVPs are purely thermal or also  
 77 compositionally distinct features. Isotopic variations in intraplate volcanic rocks (Dupré  
 78 & Allègre, 1983; Hart, 1984; Arevalo Jr et al., 2013), joint seismic-geodynamic inversions  
 79 (Lu et al., 2020), body tides (Lau et al., 2017), and their apparent stability with respect to  
 80 the reconstructed locations of Phanerozoic kimberlites and large igneous provinces (Burke  
 81 et al., 2008), all suggest that LLVPs are enriched in chemically distinct and anomalously  
 82 dense material. Numerical models suggest that this material must have a  $\sim 2\text{--}4\%$  intrinsic  
 83 chemical density excess to generate and preserve such compositional heterogeneity over  
 84 billion-year timescales (Kellogg et al., 1999; Zhong & Hager, 2003; Tan & Gurnis, 2005;  
 85 Deschamps & Tackley, 2009; Tan et al., 2011; Tackley, 2012; Y. Li et al., 2014; Mulyukova  
 86 et al., 2015; Jones et al., 2020). Seismic evidence in favour of chemically distinct LLVPs has,  
 87 however, proven less conclusive. For example, the decorrelation between shear-wave velocity  
 88 ( $V_S$ ) and bulk sound velocity ( $V_\phi$ ) below 2000 km depth has been inferred to support both  
 89 thermal and thermochemical interpretations (Trampert et al., 2004; Della Mora et al., 2011;  
 90 Moulik & Ekström, 2016; Koelemeijer et al., 2018). Similarly, strong lateral  $V_S$  gradients  
 91 at LLVP boundaries may point to chemical heterogeneity (Ni et al., 2002; Sun & Miller,  
 92 2013), but several studies suggest that similar features may occur with purely thermal  
 93 variations (Schuberth et al., 2009; Davies et al., 2012; Ward et al., 2020). While normal  
 94 mode studies generally prefer anomalously dense LLVPs (Ishii & Tromp, 2001; Trampert et  
 95 al., 2004; Moulik & Ekström, 2016), recent Stoneley mode observations (i.e., normal modes  
 96 trapped along the CMB) indicate that LLVPs are, on average, positively buoyant, although  
 97 a  $\sim 100$  km-thick, anomalously dense basal layer cannot be ruled out (Koelemeijer et al.,  
 98 2017). This result apparently contradicts inferences from body tide observations, which  
 99 yield a mean excess density of  $\sim 1\%$  within the bottom  $\sim 350$  km of the LLVPs (Lau et al.,  
 100 2017).

101 While LLVP buoyancy structure remains uncertain, their morphology and the po-  
 102 tential presence of chemically distinct basal material is expected to significantly influence  
 103 spatiotemporal patterns of mantle circulation (Gurnis et al., 2000; Forte & Mitrovica, 2001;  
 104 McNamara & Zhong, 2004; Ghelichkhan & Bunge, 2018; M. Li & McNamara, 2018; Lu et  
 105 al., 2020). Since the earliest models of whole-mantle flow (Hager et al., 1985; Ricard et al.,  
 106 1993), there have been several important advances in geodynamic observables, notably im-  
 107 proved present-day constraints on excess ellipticity of the CMB (Dehant et al., 2017) and the  
 108 planform of surface dynamic topography (Hoggard et al., 2016). Moreover, recent geodetic  
 109 and seismological measurements of Earth’s long-period motions—in particular, body tides  
 110 and Stoneley modes—now provide additional bounds on deep mantle density structure.  
 111 These developments allow us to investigate the trade-off between the magnitude and distri-  
 112 bution of LLVP buoyancy, and to re-examine these controversies using new simulations of  
 113 whole-mantle flow, tidal deformation and Stoneley mode oscillations.

114 Using a suite of existing tomographic models, we perform geodynamic inversions to  
 115 determine whether thermal or thermochemical density structures are more compatible with  
 116 observations of the geoid, CMB ellipticity, and dynamic topography. The best-fitting density  
 117 configurations are then tested against independent Stoneley mode splitting and body tide  
 118 measurements, and we demonstrate that the existing discrepancies between these datasets  
 119 can be resolved. Finally, we explore geochemical implications of these inversion-derived  
 120 buoyancy structures using thermodynamic calculations of density and elastic properties of  
 121 possible compositional endmembers. By analysing the fits of the resulting model predictions  
 122 with a wide range of observations, we constrain the nature and distribution of chemical  
 123 heterogeneity within the deep Earth.

## 124 2 Reconciling geodynamic observations and predictions

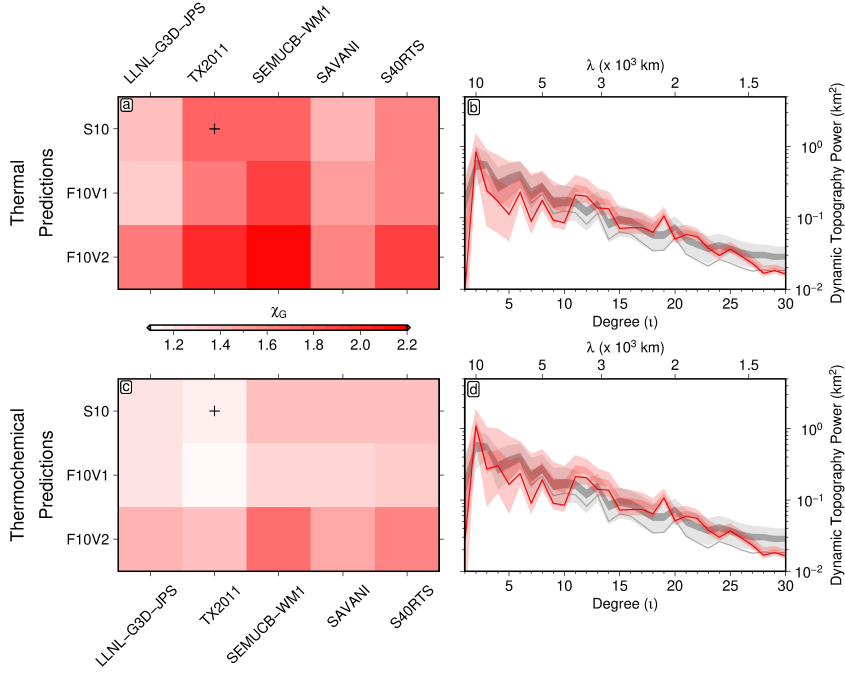
125 Recent re-evaluation of dynamic surface topography using global inventories of residual  
 126 depth measurements confirms that the long-wavelength component of this field is spatially  
 127 correlated with geoid height anomalies (Figure 2a–b; Hoggard et al., 2016, 2017). While



**Figure 2. Observations versus optimal instantaneous flow modelling predictions for TX2011 tomographic model and S10 viscosity profile.** (a) Observed non-hydrostatic geoid height anomalies (Förste et al., 2008; Chambat et al., 2010). (b) Observed dynamic surface topography (Hoggard et al., 2017). (c) Observed excess CMB ellipticity (Dehant et al., 2017). (d) Predicted geoid for optimal mantle density model assuming LLVPs are purely thermal features. VR = variance reduction;  $r$  = Pearson’s correlation coefficient (Appendix C). (e) Predicted dynamic topography for this model. (f) Predicted excess CMB ellipticity for this model.  $\chi_C$  = misfit to observed CMB excess ellipticity (Appendix C). (g–i) Same for optimal density model that includes compositionally distinct LLVPs.

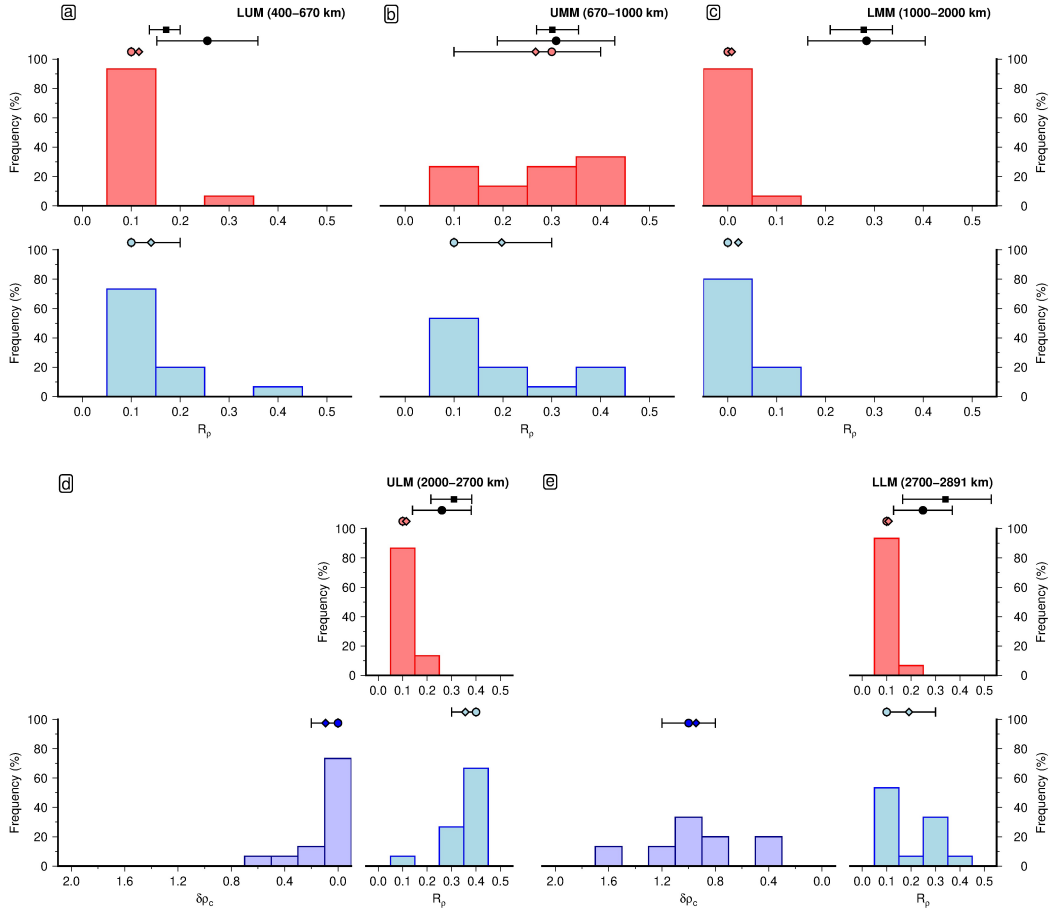
128 there is some disagreement on the appropriate methodology for spectrally analysing these  
 129 data, studies have converged on water-loaded amplitudes of  $\pm 700$  m at spherical harmonic  
 130 degrees  $l = 1-3$  (Hoggard et al., 2016; Yang & Gurnis, 2016; Watkins & Conrad, 2018;  
 131 Davies et al., 2019; Steinberger et al., 2019; Valentine & Davies, 2020). Meanwhile, for its  
 132 core-mantle boundary counterpart, geodetic observations of Earth’s free core nutation place  
 133 a narrow bound of  $\sim 400 \pm 100$  m on the amplitude of the degree-two ( $l = 2$ ), order-zero  
 134 ( $m = 0$ ) component of non-hydrostatic CMB topography (i.e., excess ellipticity; Figure 2c;  
 135 Dehant et al., 2017). Unfortunately, efforts to map global CMB topography at shorter  
 136 wavelengths using seismic data are presently hampered by trade-offs between velocity and  
 137 density structure in the  $D''$  region (Koelemeijer, 2021).

138 In light of these improved and revised constraints, we ask: Can a model of  $V_S$ -derived  
 139 mantle density be constructed that simultaneously satisfies the geoid, dynamic topography,  
 140 and excess CMB ellipticity? To investigate this issue, we construct a suite of  $\sim 10^6$  density  
 141 models, simulate the resulting instantaneous mantle flow, and compute misfits to the ob-  
 142 servational data sets (Appendices A–C). For the upper mantle above 400 km, we adopt a  
 143 modified version of the RHGW20 density model (F. D. Richards et al., 2020), which accounts  
 144 for anelasticity at seismic frequencies and yields demonstrably better fit to short-wavelength  
 145 dynamic topography than previous models. We divide the deeper mantle into five layers,  
 146 and within each layer, we vary the  $V_S$ -to-density scaling factor ( $R_\rho = \frac{d \ln \rho}{d \ln V_S}$ ) between 0.1–



**Figure 3. Geodynamic misfit as a function of input density and viscosity model.** (a) Total geodynamic misfit,  $\chi_G$  (Appendix C), of best-fit thermal models for each combination of viscosity and seismic tomographic input. Black cross = model shown in (b) and Figure 2d–f. (b) Observed and predicted dynamic topography power spectra of best-fit thermal model for TX2011 and S10 viscosity profile. Dark and light gray envelope = 99% and 50% confidence intervals for power spectrum of optimal spherical harmonic coefficients for oceanic residual depth measurements (intervals derived from 100,000 random samples of inverted spherical harmonic coefficient probability distributions; Valentine & Davies, 2020); solid gray line = power spectrum of mean spherical harmonic coefficients determined for oceanic residual depth measurements; dark and light red envelope = 99% and 50% confidence intervals for power spectrum of thermal model constructed by sampling predicted dynamic topography at locations of shiptrack and point-wise oceanic residual depth measurements and determining optimal spherical harmonic coefficients using Gaussian process-based methodology of Valentine & Davies (2020); solid red line = power spectrum of mean spherical harmonic coefficients determined for thermal model. (c) Total geodynamic misfit,  $\chi_G$ , of best-fit thermochemical models for each combination of viscosity and seismic tomographic input. Black cross = model shown in (d) and Figure 2g–i. (d) Observed and predicted dynamic topography power spectra of best-fit thermochemical model for TX2011 and S10 viscosity profile, as in (b).

147 0.4. This range is in line with expectations from mineral physics constraints on pyrolytic  
 148 and mixed pyrolytic-basaltic compositions, which are both hypothetical compositions for an  
 149 isochemical mantle (Deschamps et al., 2012; Stixrude & Lithgow-Bertelloni, 2012; Lu et al.,  
 150 2020). To allow for limited seismic resolution and potential imaging artefacts in the lower  
 151 mid-mantle (1000–2000 km), we also test  $R_\rho = 0$  in this region. In addition to using models  
 152 with vertically varying  $V_S$ -to-density scaling factors, we construct a suite of thermochemi-  
 153 cal models where chemical heterogeneity is represented as a density jump, ranging between  
 154 0.0–2.0%, between the LLVP interior and exterior. We generate density models using five  
 155 seismic tomographic models and perform instantaneous flow calculations using three mantle  
 156 viscosity profiles (Appendices A and B).



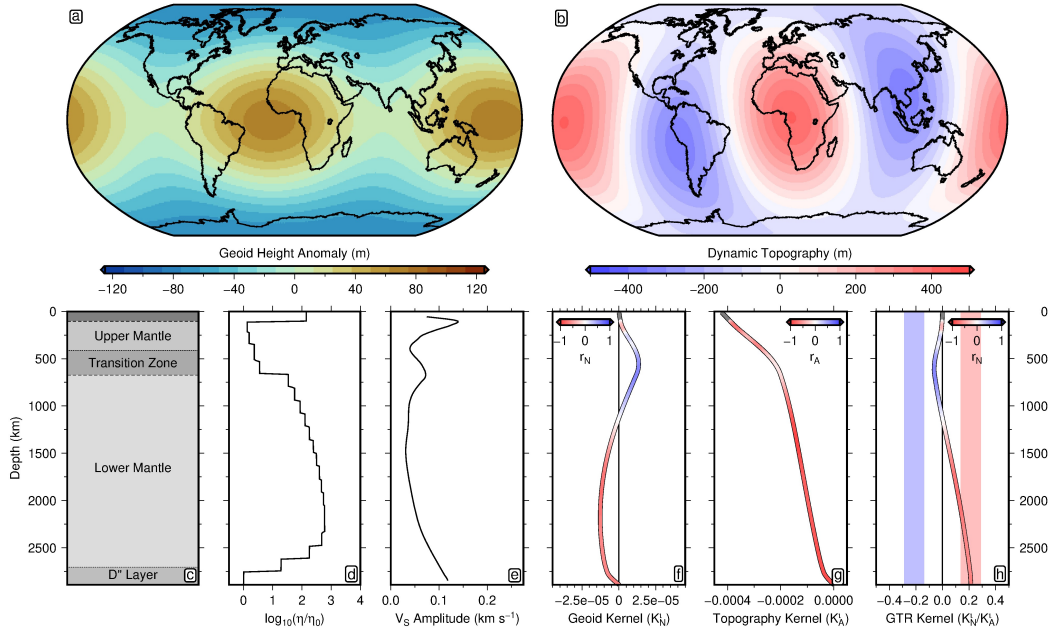
**Figure 4. Best-fitting  $R_\rho$  values for thermal and thermochemical models.** (a) Lower upper mantle layer (400–670 km). Red bars = best-fitting  $R_\rho$  for thermal models; blue bars = best-fitting  $R_\rho$  for thermochemical models; coloured circles and error bars = medians and interquartile ranges; diamonds = mean weighted by total misfit,  $\chi_G$ ; black circle and error bars = predicted values and  $\pm 2\sigma$  errors for a pyrolitic mantle composition, excluding post-perovskite (taken from Lu et al., 2020); black square and error bars = predicted values and  $\pm 2\sigma$  errors for mantle compositions ranging from a mechanical mixture of 50% MORB and 50% pyrolite to 100% pyrolite, determined using *Perple\_X* (Connolly, 2005; Stixrude & Lithgow-Bertelloni, 2011). In both cases, thermodynamic  $R_\rho$  predictions are corrected for anelasticity following methodology outlined in Lu et al. (2020) and Text S2.2. (b) Same for upper mid-mantle layer (670–1000 km). (c) Same for lower mid-mantle layer (1000–2000 km). (d) Same for upper lower mantle layer (2000–2700 km). Dark blue bars = best-fitting compositional density contrast,  $\delta\rho_c$ , for thermochemical models. (e) Same for lower lower mantle layer (2000–2700 km).

157 Three key results emerge from this analysis. First, we find that acceptable fits to both  
 158 the geoid and dynamic surface topography can be obtained for thermal and thermochemical  
 159 density models (Figures 2, 3, and S1–S6; Table S1). Second, we obtain lower misfits, higher  
 160 correlation coefficients, and greater variance reductions for models that include compositionally  
 161 distinct material in the LLVPs relative to purely thermal models (Appendix C). This  
 162 difference is particularly clear for the excess CMB ellipticity (Figure 2f versus 2i). Thermochemical  
 163 models generally prefer strong excess density within the LLVP portion of the D''

164 layer ( $\delta\rho_c \geq +0.8\%$  for 13 of 15 tomographic and viscosity model combinations), but find  
 165 little to no excess density in the shallower 2000–2700 km depth range ( $\delta\rho_c \leq +0.2\%$  for 13 of  
 166 15 models; Figure 4d and 4e; Table S3). The thermochemical models also generally return  
 167  $R_\rho$  values throughout the middle (400–1000 km) and lower (2000–2900 km) mantle that are  
 168 in better agreement with experimental expectations for a pyrolitic composition (Figure 4a,  
 169 4b, 4d and 4e; Deschamps et al., 2012; Lu et al., 2020). Third, all best-fitting models require  
 170  $R_\rho \sim 0$  for the 1000–2000 km mid-mantle layer, irrespective of whether or not LLVP regions  
 171 are modelled as compositionally distinct (Figure 4c; Tables S2–S3; for more discussion see  
 172 Text S1.1–1.2).

### 173 3 Vertical extent of LLVPs

174 The geodynamic inversions exhibit a preference for  $R_\rho \sim 0$  throughout the mid-mantle,  
 175 which is too low for any plausible mantle composition and indicates that geodynamic ob-  
 176 servables are incompatible with strong thermal buoyancy contributions from this depth.  
 177 Given that seismic tomographic models are dominated by  $l = 2$  structure over the 1000–  
 178 2000 km depth range, we explore this result further using associated sensitivity kernels for  
 179 instantaneous mantle flow.



**Figure 5. Relationship between degree-two dynamic topography, geoid and  $V_S$  anomalies.** (a) Observed non-hydrostatic geoid height anomalies (Förste et al., 2008; Chambat et al., 2010). (b) Observed water-loaded dynamic topography (Davies et al., 2019). (c) Schematic radial mantle structure. (d) Normalised radial viscosity,  $\eta$ , profile (S10; Steinberger et al., 2010). (e) Spectral amplitude of  $l = 2$   $V_S$  anomalies from SEMUCB-WM1 tomographic model (French & Romanowicz, 2015). (f) Geoid kernel,  $K_N^l$ , coloured by geoid-to- $V_S$  anomaly correlation,  $r_N$ , as a function of depth. (g) Dynamic topography kernel,  $K_A^l$ , coloured by dynamic topography-to- $V_S$  anomaly correlation,  $r_A$ . (h) Geoid-to-topography ratio (GTR) kernel, coloured by  $r_N$ . Blue/red bands = values required to produce the observed GTR when thermal density anomalies are correlated/anti-correlated with the geoid.



180 The geoid-to-topography amplitude ratio (GTR) at  $l = 2$  provides a crucial con-  
 181 straint on the vertical extent of long-wavelength buoyancy anomalies associated with LLVPs.  
 182 In Figures 5a and b, we show the  $l = 2$  components of observed non-hydrostatic geoid  
 183 height anomalies and water-loaded dynamic topography, which yield an estimated GTR  
 184 of  $\sim 0.21 \pm 0.07$ . These deflections must be caused by  $l = 2$  density anomalies, with the  
 185 strongest corresponding shear-wave velocity ( $V_S$ ) anomalies found within the LLVP regions,  
 186 the mantle transition zone, and the asthenosphere (Figure 5e). These  $V_S$  anomalies are  
 187 anti-correlated with the observed geoid and dynamic topography, with the exception of the  
 188 transition zone, where  $V_S$  anomalies correlate with the geoid but remain anti-correlated or  
 189 become decorrelated with dynamic topography (Figure 5f-g).

190 Individual  $l = 2$  sensitivity kernels for the geoid, dynamic topography, and GTR  
 191 (Figure 5f-h; Appendix B) are sensitive to the choice of mantle viscosity profile (Figure 5d),  
 192 but their shape is broadly consistent for a range of published profiles (Figure S8; Forte et  
 193 al., 2010; Liu & Zhong, 2016). The  $l = 2$  GTR kernel shows that, to satisfy the observed  
 194 value of  $0.21 \pm 0.07$ , density anomalies must either anti-correlate with surface deflections in  
 195 the deep mantle (intersection with red band in Figure 5h) or positively correlate with the  
 196 geoid—while remaining negatively correlated with dynamic topography—in the transition  
 197 zone (intersection with blue band in Figure 5h). Our analyses support the conclusions of  
 198 previous studies (e.g., Hager et al., 1985) that deeper mantle structure is the dominant  
 199 contributor to the integrated GTR. These kernels also show that any  $l = 2$ , mid-mantle  
 200 ( $\sim 1000$ – $2000$  km) thermal density anomalies can only lower the GTR. A mantle density  
 201 model with LLVPs extending shallower than  $\sim 2000$  km depth (i.e.,  $\sim 900$  km above the  
 202 CMB) that fits the observed geoid will therefore inevitably overpredict long-wavelength  
 203 dynamic topography. Hence, the inversions return a preferred value of  $R_\rho \approx 0$  in the mid-  
 204 mantle. This finding provides strong evidence that long-wavelength low density anomalies  
 205 associated with LLVPs do not vertically extend beyond 900 km above the CMB, which is  
 206 consistent with recent arguments that seismically imaged  $l = 2$ , mid-mantle  $V_S$  structure  
 207 is an artefact of limited tomographic resolution (Davaille & Romanowicz, 2020). Smaller  
 208 scale density anomalies do exist in the 1000–2000 km depth interval (e.g., plumes and  
 209 slabs; French & Romanowicz, 2015; N. Simmons et al., 2015); however, instantaneous flow  
 210 sensitivity kernels for shorter wavelengths approach zero over this depth range, such that  
 211 these features have minimal impact on the geoid, surface dynamic topography and CMB  
 212 ellipticity. Indeed, geodynamic misfit changes by less than 10% when our optimised density  
 213 fields are modified to suppress  $l = 2$  structure while retaining shorter wavelength features  
 214 between 1000–2000 km depth (by instead applying a high-pass filter and setting  $R_\rho = 0.2$ ,  
 215 see Text S1.3; Karato & Karki, 2001).

#### 216 4 Compatibility with body tides and Stoneley modes

217 Despite similar, though not completely identical sensitivity to deep Earth structure  
 218 (Robson et al., 2022), previous studies based on semi-diurnal body tide and Stoneley mode  
 219 splitting observations arrive at contrasting conclusions about LLVP density structure. The  
 220 former show a clear preference for the presence of anomalously dense material, with trade-  
 221 offs between the amplitude and depth distribution of excess density (Lau et al., 2017). In  
 222 contrast, by also taking topography of the CMB into account, the latter prefer models  
 223 with integrated density anomalies in the lower 400 km that are negative, as expected for a  
 224 dominantly thermal control (Koelemeijer et al., 2017). In light of these studies, we next test  
 225 whether the mantle structure obtained from our optimal TX2011-based geodynamic model  
 226 with thermochemical variations, or its purely thermal counterpart, is most consistent with  
 227 these geodetic and seismological observations.

228 Goodness-of-fit to semi-diurnal body tide constraints is calculated following the method-  
 229 ology of Lau et al. (2017), which requires the improvement of predictions for 3D mantle  
 230 structure over a 1D reference case to be significant at the 95% level (Appendix D). The op-  
 231 timal TX2011-derived thermal model produces results that are only significant at the 93.8%

level. By contrast, the best-fitting thermochemical density model based on the same tomographic input, but with chemical heterogeneity in the base of LLVPs, yields statistically significant outcomes (95.8% significance level).

We predict Stoneley mode splitting functions by adapting the methodology of Koelemeijer et al. (2017) (Appendix D). Our revised approach has two methodological advantages over this study. Firstly, both the range and magnitude of  $R_\rho$  tested here are consistent with candidate chemical compositions in the deep mantle (as compiled by Lu et al., 2020). Secondly, by calculating the instantaneous mantle flow associated with each model, CMB deflections are dynamically consistent with each LLVP density structure rather than scaled. We find that the misfit between observed and predicted Stoneley mode splitting functions is  $\sim 20\%$  lower for the optimal TX2011-based thermochemical density model compared with its equivalent thermal model (Table S4; Figure S9). This conclusion appears to contradict the findings of Koelemeijer et al. (2017), but is readily explained by our methodological improvements, as well as the stronger  $V_S$  amplitudes at  $l = 2$  below 2500 km depth in TX2011 compared to the SP12RTS model adopted in that study (Text S1.4).

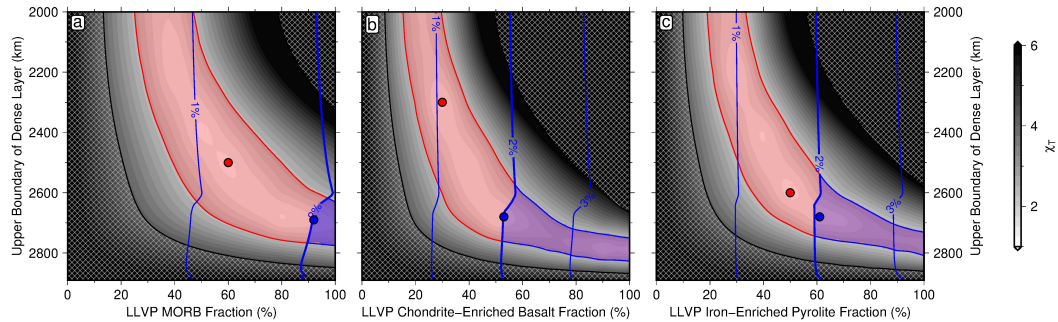
Significantly, these results indicate that the presence of anomalously dense material in the bottom  $\sim 200$  km of the LLVPs is not only compatible with available geodynamic constraints, but is also consistent with observations of Earth’s semi-diurnal body tide and Stoneley mode splitting.

## 5 Implications for lower mantle chemistry

Having established that geodynamic, seismological, and geodetic constraints provide evidence for the presence of a dense basal layer within the LLVPs, we explore the compatibility of different candidate compositions. Several hypotheses have been proposed for the formation of chemically distinct LLVP material, including: slow accumulation of basalt from subducted slabs reaching the CMB (Niu, 2018); preservation of primordial mantle material segregated during top-down crystallisation of a basal magma ocean (Labrosse et al., 2007); subduction of iron- and silicon-rich Hadean crust along with a terrestrial regolith comprising chondritic and solar-wind-implanted material (Tolstikhin & Hofmann, 2005); and pooling of dense, iron-rich melts generated in the primordial mantle transition zone (Lee et al., 2010).

We have assembled three endmembers to test the compositional range encompassed by these different scenarios: i) present-day mid-ocean ridge basalt (MORB; lowest iron, highest silicon content; Workman & Hart, 2005); ii) chondrite-enriched Hadean basalt (intermediate iron and silicon; Tolstikhin & Hofmann, 2005); iii) iron-enriched pyrolite (highest iron, lowest silicon), representing early Archaean melts generated in the transition zone or remnants of a basal magma ocean (Lee et al., 2010; Labrosse et al., 2007; Table A1; for more discussion see Text S2.1–2.2). For each of these compositions, we perform thermodynamic modelling and find that all options yield a positive density and negative shear-wave velocity anomaly with respect to ambient pyrolitic mantle at deep mantle temperatures and pressures ( $\sim 2000$ – $4000$  K;  $\sim 110$ – $140$  GPa; Figure S10; Connolly, 2005; Stixrude & Lithgow-Bertelloni, 2011). The amplitude of these anomalies vary, with modern basaltic material generating the weakest anomalies, while the most iron-rich primordial components produce the strongest anomalies (Workman & Hart, 2005; Lee et al., 2010).

The relatively modest excess density below 2700 km recovered in our initial geodynamic inversions ( $\overline{\delta\rho_c} = 0.4$ – $1.6\%$ ) is consistent with mechanical mixtures comprising 20–70% pyrolite and 80–30% modern MORB, or 50–90% pyrolite and 50–10% of either iron-rich primordial component. This excess density, however, falls below the  $\sim 2$ – $4\%$  threshold required for long-term preservation of intra-LLVP chemical heterogeneity (Tackley, 2012; Mulyukova et al., 2015; Jones et al., 2020). We therefore explore how a trade-off between the thickness of the basal layer and its excess density affects the fit to the geodynamic and seismic constraints, and which of the proposed chemical compositions are most compatible.



**Figure 6. Combined misfit to geodynamic and Stoneley mode observations as a function of mantle composition.** (a) Combined total misfit ( $\chi_T$ ) as a function of MORB fraction within the LLVPs (Workman & Hart, 2005). Material outside the LLVPs is assumed to be pyrolitic. Hatched region = models with peak-to-valley  $l = 2$  CMB topography exceeding  $\pm 4.7$  km maximum constraint (Koelemeijer, 2021); red circle = best-fitting model; red shading = models with misfit less than double that of global minimum; thin blue contours = compositional density difference between dense layer material and ambient mantle; bold blue contour = lower limit of suggested  $\sim 2\text{--}4\%$  compositional density threshold for long-term preservation of intra-LLVP chemical heterogeneity (Tackley, 2012; Mulyukova et al., 2015; Jones et al., 2020); blue circle = best-fitting model with intrinsic density anomaly above preservation threshold; blue shading = models with misfit less than double that of global minimum and compositional density anomaly above preservation threshold. (b) Same for primordial material (chondrite-enriched basalt; Tolstikhin & Hofmann, 2005). (c) Same for primordial material (iron-enriched pyrolite; Lee et al., 2010).

282 Instantaneous flow calculations are repeated with density models constructed from the  
 283 thermodynamic predictions for different combinations of chemical components within and  
 284 outside the LLVPs (Appendices A and B). Mantle material is modelled as a mechanical  
 285 mixture of pyrolite and each candidate composition, with density anomalies set to zero  
 286 between 1000–2000 km depth based on the geodynamic inversion results. We find a strong  
 287 trade-off between the anomalous density of the basal LLVP region and its thickness, with  
 288 similar misfit to geodynamic observables obtained for thin, highly enriched versus thicker,  
 289 less chemically distinct basal layers (Figure S16). Although results are dependent on the  
 290 radial mantle viscosity profile, optimal fits are generally obtained for thinner, more enriched  
 291 layers, irrespective of whether anomalously dense material within the LLVPs is assumed to  
 292 be basaltic or primordial. Best-fitting models for each chemical component yield similar  
 293 misfit values, with optimal layer thicknesses of  $\sim 200$  km.

294 Combining geodynamic and Stoneley mode misfit into a joint misfit function does not  
 295 significantly reduce the trade-off between basal layer thickness and density (Appendix D;  
 296 Figures S12, S13, and S15). Nevertheless, while each endmember composition can generate  
 297 density models that satisfy the 2–4% excess density threshold for long-term chemical hetero-  
 298 geneity preservation (Tackley, 2012; Mulyukova et al., 2015; Jones et al., 2020), the two pri-  
 299 mordial candidates yield a  $\sim 10\%$  reduction in joint misfit to Stoneley mode and geodynamic  
 300 observations compared with recycled MORB (Figure 6). Irrespective of whether the TX2011  
 301 or S40RTS tomographic model is used to generate density structure, optimal chondrite-  
 302 enriched basaltic configurations give  $\sim 5\text{--}10\%$  lower misfit than their iron-enriched pyrolitic  
 303 counterpart, indicating that a 100–200 km-thick layer, mainly composed of sequestered  
 304 Hadean crust, is most consistent with available data (Text S2.3; Figures 6 and S13–S15).

305 Although the uncertainty inherent to thermodynamic estimations of  $V_S$  and density  
 306 means that our conclusion regarding basal layer composition is not definitive (Connolly &  
 307 Khan, 2016), the presence of Hadean crust in these regions is consistent with several in-  
 308 dependent constraints. Firstly, time-dependent thermochemical convection studies suggest  
 309 that subducted,  $\sim 10$  km-thick present-day oceanic crust is easily re-entrained, whereas  
 310 an early-formed proto-crust with greater thickness and higher iron content could be more  
 311 readily preserved within the base of LLVPs (M. Li & McNamara, 2013; M. Li et al., 2014).  
 312 Secondly, the elevated  $\text{SiO}_2$  content of the primordial basaltic composition compared with  
 313 iron-enriched pyrolite helps to explain the observed spatial decorrelation between  $V_\phi$  and  
 314  $V_S$  in the lowermost mantle, provided that bridgmanite is at least partially replaced by  
 315 post-perovskite within this depth range (Su & Dziewonski, 1997; Hernlund & Houser, 2008;  
 316 Koelmeijer et al., 2018; Figure S10). Thirdly, the less extreme reduction in  $V_S$  at lower-  
 317 most mantle conditions for primordial basalt ( $\sim 2\%$ ), compared to iron-enriched pyrolite  
 318 ( $\sim 3\%$ ), is more compatible with the relatively modest  $V_S$  gradients that have been inferred  
 319 across LLVP boundaries (Hernlund & Houser, 2008; Davies et al., 2012; Deschamps et al.,  
 320 2012; Ward et al., 2020). Finally, when comparing observed and predicted  $V_S$ ,  $V_P$  and  $V_\phi$   
 321 signatures for a wide range of candidate LLVP compositions, Vilella et al. (2021) found that  
 322 seismic constraints necessitate minimal quantities of ferropericlase ( $<6$  vol%) and poten-  
 323 tially large proportions of calcium-perovskite (up to  $\sim 35$  vol%), consistent with expected  
 324 phase assemblages for basaltic material at deep mantle conditions. Optimal compositions  
 325 found by this study also feature elevated  $\text{Al}_2\text{O}_3$  contents (3–13 wt%) and oxidation states  
 326 ( $\text{Fe}^{3+}/\sum\text{Fe} > 0.3$ ), which can be attributed to the addition of chondritic material and chem-  
 327 ical partitioning during a shallow melting event (McKay et al., 1991; Walter, 1998; Zega et  
 328 al., 2003; Herzberg, 2016; Zhang et al., 2017; Table A1).

329 For all the reasons listed above, we conclude that the most likely candidate for the  
 330 chemically distinct, 100–200 km-thick basal layer is Hadean basaltic material combined with  
 331 solar wind-implanted chondritic regolith (Tolstikhin & Hofmann, 2005). Nevertheless, more  
 332 iron-rich and silicon-poor primordial endmembers, or mixtures of these components, cannot  
 333 currently be discounted given the uncertainties associated with lower mantle seismic tomo-  
 334 graphic imaging, viscosity structure, and the inference of physical properties from seismic  
 335 observations. We also note that the geodynamic, seismological, and geodetic constraints  
 336 we use here are only sensitive to long-wavelength density variations. As a result, the dense  
 337 material we identify may be unevenly distributed within the basal layers and potentially  
 338 concentrated beneath broad thermochemical plumes (e.g., Davaille & Romanowicz, 2020;  
 339 Lu et al., 2020).

340 Sites of past and present intraplate volcanism have been shown to spatially correlate  
 341 with LLVPs, leading to speculation that anomalous isotope ratios of basalts erupted in these  
 342 settings may originate from these deep mantle reservoirs (Hart, 1984; Castillo, 1988; White,  
 343 2015). These isotopic signatures include high  $\text{He}^3/\text{He}^4$  ratios and positive  $\mu^{182\text{W}}$  anomalies.  
 344 Since all  $\text{He}^3$  is of primordial origin and the decay of  $^{182}\text{Hf}$  to  $^{182}\text{W}$  has a half-life of only  
 345  $\sim 9$  Myr, these chemical signals point to the presence of an early-formed and largely isolated  
 346 (i.e., relatively undegassed) geochemical reservoir. Our finding that the basal  $\sim 100$ – $200$  km  
 347 of the LLVPs most likely contain iron- and silicon-enriched primordial material, rather than  
 348 accumulations of more youthful MORB, suggests that these regions are the source of these  
 349 anomalies and that additional geochemical deviations are therefore derived from other reser-  
 350 voirs (Mundl-Petermeier et al., 2020; Gleeson et al., 2021; Day et al., 2022; Tucker et al.,  
 351 2022). This inference is further supported by the agreement between the approximate mass  
 352 of these layers ( $3 - 6 \times 10^{22}$  kg) and that of the primordial Earth reservoir inferred from  
 353  $\text{He}^3/\text{He}^4$  ratios ( $\sim 6.2 \times 10^{22}$  kg; Tolstikhin & Hofmann, 2005). Finally, our finding that  
 354 LLVP basal layers likely contain chondrite-enriched basalt, coupled with their relative thin-  
 355 ness, is also consistent with geodynamic studies investigating the origin of systematic  $\mu^{182\text{W}}$   
 356 differences between ocean island basalts and flood basalts erupted in large igneous provinces  
 357 (Jones et al., 2019). Clearly further integration of geochemical and geophysical constraints  
 358 is needed to confirm whether these thin basal layers represent Hadean crust, remnants of

359 magmatic processes within the early Earth’s interior, or a combination of both. Our pro-  
 360 posed model of Earth structure does, however, already provide a self-consistent explanation  
 361 of a full range of geodynamic, geodetic, seismological, and geochemical constraints.

362 Both the presence of dense primordial material within LLVPs and the limited vertical  
 363 extent of their associated buoyancy ( $\leq 900$  km above the CMB) have important implications  
 364 for existing predictions of mantle evolution, reducing the amplitude and slowing the rate of  
 365 change of surface dynamic topography. By adopting this structure and validating its associ-  
 366 ated mantle flow field against evidence for continent-scale uplift and subsidence encoded in  
 367 the geological record, our understanding of Earth’s internal dynamics can be greatly refined,  
 368 allowing impacts on landscape evolution and palaeoclimatic shifts to be determined with  
 369 unprecedented fidelity.

## 370 6 Conclusions

371 Determining the thermochemical properties and vertical extent of LLVP-related buoy-  
 372 ancy anomalies is of fundamental importance to solving a range of outstanding controver-  
 373 sies in Earth sciences, including the formation of mantle plumes, the origin of anomalous  
 374 geochemical signatures in the basalts they produce, and the rate at which convectively sup-  
 375 ported topography grows and decays. By taking a multi-pronged approach that integrates  
 376 a full range of geodynamic, geodetic and seismological data with numerical models, we are  
 377 able to place valuable new constraints on LLVP structure. Firstly, by using recent mea-  
 378 surements of Earth’s dynamic topography, CMB ellipticity, and geoid to determine optimal  
 379 models of mantle flow, we find that—irrespective of assumed tomographic and rheologic  
 380 configuration—anomalously dense material is concentrated within the basal  $\sim 200$  km of  
 381 LLVPs. Secondly, we conclude that buoyancy variations associated with these seismically  
 382 imaged features extend no more than  $\sim 900$  km above the core-mantle boundary. Thirdly, we  
 383 show that the apparent disagreement between LLVP buoyancy structures previously inferred  
 384 from Stoneley mode and body tide inversions can be resolved using these optimised thermo-  
 385 chemical models. Finally, by comparing an ensemble of thermodynamically self-consistent  
 386 density models that cover a range of possible recycled and primordial compositions to a full  
 387 suite of geodynamic, seismological, and geochemical constraints, we demonstrate that the  
 388 dense basal material within the LLVPs likely comprises remnants of Hadean crust and chon-  
 389 dritic regolith. These results confirm that basal sections of LLVPs are potential reservoirs  
 390 for the primordial isotope signatures observed in oceanic island basalts. Our work further  
 391 suggests that long-wavelength, convectively-driven topography evolves relatively slowly and  
 392 is lower amplitude than would be expected if LLVPs were purely thermal features.

## 393 Appendix A Mantle density models

394 We develop two classes of mantle density models; the first based on inversion of geo-  
 395 dynamic data, the second derived using thermodynamic forward modelling of proposed  
 396 chemical compositions. To generate the first class of density models, we separate the mantle  
 397 into six layers: 0–400 km (UUM = upper upper mantle), 400–670 km (LUM = lower upper  
 398 mantle), 670–1000 km (UMM = upper mid-mantle), 1000–2000 km (LMM = lower mid-  
 399 mantle), 2000–2700 km (ULM = upper lower mantle), and 2700–2891 km (LLM = lower  
 400 lower mantle). Density in the UMM layer is determined from SLNAAFSa (Hoggard et al.,  
 401 2020), which is a version of the SL2013sv (Schaeffer & Lebedev, 2013) upper mantle model  
 402 into which the regional updates SL2013NA in North America (Schaeffer & Lebedev, 2014),  
 403 AF2019 in Africa (Celli, Lebedev, Schaeffer, Ravenna, & Gaina, 2020), and SA2019 in South  
 404 America and the South Atlantic Ocean (Celli, Lebedev, Schaeffer, & Gaina, 2020) have  
 405 been incorporated. The baseline model, SL2013sv, has been shown to produce topographic  
 406 predictions that are in good agreement with residual depth measurements, even at relatively  
 407 short wavelengths ( $\sim 1000$  km; F. D. Richards et al., 2020).

408 Seismic velocities are converted into density within the UMM layer using an anelastic  
 409 parameterisation following the methodology of F. D. Richards et al. (2020). This approach  
 410 allows self-consistent mapping between seismic velocities and temperature, density, and  
 411 viscosity variations, while correcting for discrepancies between tomographic models that  
 412 result from parameterisation choices rather than true Earth structure. Optimal param-  
 413 eters determined for SLNAAFSA are:  $\mu_0 = 75.9$  GPa;  $\frac{\partial\mu}{\partial T} = -17.9$  MPa  $^{\circ}\text{C}^{-1}$ ;  $\frac{\partial\mu}{\partial P} = 2.54$ ;  
 414  $\eta_r = 10^{23.0}$  Pa s;  $E_a = 489$  kJ mol $^{-1}$ ;  $V_a = 0.63$  cm $^3$  mol $^{-1}$ ; and  $\frac{\partial T_s}{\partial z} = 0.931$   $^{\circ}\text{C}$  km $^{-1}$ . We  
 415 assume that continental lithosphere, delineated by the  $T = 1200$   $^{\circ}\text{C}$  isothermal surface, has  
 416 neutral buoyancy and set density in these regions equal to the average density of all external  
 417 material at the relevant depth in order to eliminate any direct dynamic topographic contri-  
 418 bution. This assumption is based on heat flow measurements, xenolith geochemistry, seis-  
 419 mic velocity, gravity, and topography observations that suggest compositional and thermal  
 420 density contributions approximately balance each other within the continental lithosphere  
 421 (Jordan, 1978; Shapiro et al., 1999).

422 Deeper than 300 km, seismic velocity perturbations from whole-mantle tomographic  
 423 models LLNL-G3D-JPS (N. Simmons et al., 2015), S40RTS (Ritsema et al., 2011), SAVANI  
 424 (Auer et al., 2014), SEMUCB-WM1 (French & Romanowicz, 2015), and TX2011 (Grand,  
 425 2002) are converted to density assuming constant  $R_\rho = \partial\ln\rho/\partial\ln V_S$  values within each layer  
 426 and that average density is equal to PREM (Dziewonski & Anderson, 1981). To ensure  
 427 smooth transitions in density anomalies between the two input density parameterisations,  
 428 we take their weighted average between 300 km and 400 km, beyond which the sensitivity  
 429 of the surface wave-dominated upper mantle model tends to zero. Weighting coefficients  
 430 of the respective tomographic models,  $w_{UM}$  and  $w_{WM}$ , vary linearly between 1 and 0 over  
 431 this depth range and are combined according to  $w_{UM} = 1 - w_{WM}$ .  $R_\rho$  is fixed at 0.15 for  
 432 the whole-mantle model between 300–400 km, based on the mean value within this layer  
 433 inferred from SLNAAFSA.

434 The lower mantle layers, ULM and LLM, are laterally subdivided into regions outside  
 435 (OULM and OLLM), and within the LLVPs (LULM and LLLM), each delineated using  
 436 the -0.65%  $V_S$  anomaly contour of the whole-mantle tomographic model under investigation  
 437 (Burke et al., 2008). Outside the LLVPs,  $R_\rho$  varies as  $R_\rho = [0.1, 0.2, \dots, 0.4]$  with the exception  
 438 of the LMM layer (1000–2000 km), where a minimum bound on  $R_\rho$  of 0.0 is adopted allowing  
 439 for limited mid-mantle seismic resolution and the potential presence of artefacts due to  
 440 vertical smearing. Within the LLVPs, we apply a constant compositional density anomaly  
 441 such that  $\delta\rho(z) = R_\rho(i)\delta V_S(z) + \delta\rho_c(i)$ , where  $\delta\rho_c(i)$  is the intrinsic compositional density  
 442 difference between LLVP material and ambient mantle ( $z$  is depth,  $i$  is the layer index, i.e.,  
 443 ULM or LLM). Note that, in contrast to studies that employ negative  $R_\rho$  values (Moulik  
 444 & Ekström, 2016; Koelemeijer et al., 2017; Lau et al., 2017), this approach maximises  
 445 intra-LLVP density around the edges of the low-velocity regions rather than within their  
 446 central portions, and therefore assumes that, within each domain, internal  $V_S$  variations are  
 447 controlled by temperature in the usual manner (Figure S7). This configuration is therefore  
 448 consistent with the hypothesis that sharp compositional contrasts are responsible for strong  
 449 lateral gradients in  $V_S$  across the LLVP boundaries (Ni et al., 2002). For our thermochemical  
 450 models,  $\delta\rho_c$  varies as  $[0., 0.2, \dots, 2.0]\%$  within the LULM and LLLM regions, yielding a total  
 451 of  $\sim 2 \times 10^5$  input density structures. Note that, in the Supporting Information we also test  
 452 the effect of: a) parameterising excess LLVP density using negative  $R_\rho$  values instead of an  
 453 intrinsic density contrast and b) parameterising excess LLVP density using  $\delta\rho_c$  values, but  
 454 shifting the ULM-LLM boundary to 2800 km (see Text S1.2).

455 The second class of density models are created to investigate likely chemical compo-  
 456 sitions of the LLVPs. We generate a suite of density structures based on thermodynamic  
 457 modelling of key candidate compositions and  $V_S$  variation from tomographic models (Ta-  
 458 ble A1). For a given composition, *Perple\_X* is used alongside the thermodynamic database  
 459 of Stixrude & Lithgow-Bertelloni (2011) to generate a lookup table of anharmonic shear-  
 460 wave velocities and densities by varying temperature as [300, 350, ..4500] K and pressure as

| Composition | SiO <sub>2</sub> (%) | MgO (%) | FeO (%) | CaO (%) | Al <sub>2</sub> O <sub>3</sub> (%) | Na <sub>2</sub> O (%) | Reference                  |
|-------------|----------------------|---------|---------|---------|------------------------------------|-----------------------|----------------------------|
| Pyrolite    | 38.71                | 49.85   | 6.17    | 2.94    | 2.22                               | 0.11                  | Workman & Hart, 2005       |
| MORB        | 51.75                | 14.94   | 7.06    | 13.88   | 10.19                              | 2.18                  | Workman & Hart, 2005       |
| CEB         | 48.47                | 20.00   | 11.28   | 10.59   | 8.16                               | 1.50                  | Tolstikhin & Hofmann, 2005 |
| FSP         | 40.15                | 41.98   | 12.90   | 2.82    | 1.92                               | 0.23                  | Lee et al., 2010           |

**Table A1. Molar oxide ratios for different mantle compositional endmembers.** MORB = present-day mid-ocean ridge basalt; CEB = chondrite-enriched basalt; FSP = iron-enriched pyrolite.

461 [0., 0.1, ...140] GPa (Text S2.1). At each depth, temperature-dependent discontinuities in  
462 density and seismic velocity caused by phase transitions are smoothed by adopting the median  
463 temperature derivative across a  $\pm 500^\circ\text{C}$  swath either side of the geotherm. Smoothed  
464 anharmonic velocities are then corrected for anelasticity using a  $Q$  profile determined using  
465 the approach of Matas & Bukowinski (2007), as outlined in Lu et al. (2020) (Text S2.2;  
466 Figure S11). Having smoothed and corrected the  $V_S$  lookup table, velocities from a given  
467 seismic tomographic model can be converted into temperature at each depth, with values  
468 adjusted by a constant offset to ensure mean temperatures are consistent with the mantle  
469 geotherm. These temperatures are then used to extract the corresponding buoyancy  
470 structure from the smoothed density lookup table. In cases where compositions are not  
471 equivalent to a particular endmember, properties appropriate for a mechanical mixture of  
472 the two components are calculated using the Voigt-Reuss-Hill approximation to average the  
473 elastic moduli. When the composition of the LLVP is distinct from ambient mantle, temper-  
474 atures and densities are determined separately for the two components and then combined  
475 into a single array, with the boundary corresponding to the -0.65%  $V_S$  anomaly contour  
476 (Burke et al., 2008). All models assume that the range of possible mantle compositions  
477 is some combination of pyrolite and a specific dense component; either mid-ocean ridge  
478 basalt (Workman & Hart, 2005), chondrite-enriched basalt (Tolstikhin & Hofmann, 2005),  
479 or iron-enriched pyrolite (Lee et al., 2010). For each component, we generate models for  
480 compositional enrichments of [0, 10, ..., 100]% and for upper boundaries of the dense layer  
481 between 2000 km and 2800 km in 100 km increments, as well as testing 2850 km.

482 In the upper 300 km of the mantle, density structure is identical to the first class  
483 of models. Below 400 km, densities are taken directly from the thermodynamically self-  
484 consistent parameterisation described above, whilst between 300 km and 400 km depth,  
485 densities derived from the two parameterisations are smoothly merged by taking their  
486 weighted average, as described for the first class of models. Since optimal thermal and  
487 thermochemical density models recovered from geodynamic inversions consistently find that  
488  $R_\rho(\text{LMM}) \sim 0$ , density anomalies in the 1000–2000 km depth interval are set to zero for all  
489 models, although by using a high-pass filter to remove degree-two structure, we also test the  
490 effect of including only small-scale density anomalies in this depth region, described further  
491 in the Supporting Information (Text S1.3; Figure S14).

## 492 Appendix B Mantle flow simulations

493 Using the suite of thermal and thermochemical mantle density models, we predict  
494 surface and CMB dynamic topography and geoid undulations for  $1 \leq l \leq 30$  using an  
495 instantaneous flow kernel methodology. As Earth’s viscosity structure is uncertain, we assess  
496 the sensitivity of our mantle flow results to three different radial profiles that are constrained  
497 by geoid, heat flow and glacial isostatic adjustment observations: S10 (Steinberger et al.,  
498 2010); F10V1 (Forte et al., 2010); and F10V2 (Forte et al., 2010).

499 To calculate instantaneous mantle flow, we exploit the sensitivity kernel methodol-  
 500 ogy originally implemented by Hager & O’Connell (1979) and M. A. Richards & Hager  
 501 (1984), extended by Corrieu et al. (1995) to account for the effects of compressibility and  
 502 self-gravitation. This approach applies the propagator matrix technique to solve the equa-  
 503 tions governing conservation of mass and momentum within a highly viscous spherical shell,  
 504 alongside Poisson’s equation for gravity, to generate kernels describing the linear relation-  
 505 ship between geodynamic observables (dynamic topography, geoid and CMB topography)  
 506 and laterally varying density anomalies across the mantle. We impose free-slip surface and  
 507 CMB boundary conditions. For each assumed viscosity profile, the resulting sensitivity ker-  
 508 nels vary as a function of depth and the spherical harmonic degree under consideration.  
 509 Dynamic topography  $\delta A^{lm}$  can then be determined using

$$\delta A^{lm} = \frac{1}{\Delta\rho_0} \int_{R_C}^{R_\oplus} K_A^l(r) \delta\rho^{lm}(r) dr \quad (\text{B1})$$

510 where  $K_A^l$  is the dynamic topography kernel,  $r$  is radius,  $\Delta\rho_0$  is the density difference  
 511 between the uppermost mantle ( $\rho_0 = 3380 \text{ kg m}^{-3}$ ; Dziewonski & Anderson, 1981) and water  
 512 ( $\rho_w = 1030 \text{ kg m}^{-3}$ ),  $l$  and  $m$  are spherical harmonic degree and order,  $R_\oplus = 6371 \text{ km}$  and  
 513  $R_C = 3480 \text{ km}$  are the radii of the Earth and CMB, respectively, and  $\delta\rho^{lm}(r)$  represents  
 514 the driving density anomalies in the spherical harmonic expansion. The geoid,  $\delta N^{lm}$ , is  
 515 calculated using

$$\delta N^{lm} = \frac{4\pi\gamma R_\oplus}{(2l+1)g_{R_\oplus}} \int_{R_C}^{R_\oplus} K_N^l(r) \delta\rho^{lm}(r) dr \quad (\text{B2})$$

516 where  $K_N^l$  is the geoid kernel,  $g_{R_\oplus}$  is surface gravity and  $\gamma$  is the gravitational constant.  
 517 CMB topography,  $\delta C^{lm}$ , is determined according to

$$\delta C^{lm} = -\frac{1}{\Delta\rho_C} \int_{R_C}^{R_\oplus} K_C^l(r) \delta\rho^{lm}(r) dr \quad (\text{B3})$$

518 where  $K_C^l$  is the CMB topography kernel and  $\Delta\rho_C$  is the density difference between the  
 519 lowermost mantle ( $\rho_C = 5570 \text{ kg m}^{-3}$ ) and the uppermost outer core ( $\rho_{OC} = 9900 \text{ kg m}^{-3}$ ;  
 520 Dziewonski & Anderson, 1981).

521 Applying this kernel formalism permits rapid calculation of key observables, enabling  
 522 the more complete exploration of parameter space that is central to this study. This method,  
 523 however, cannot incorporate lateral viscosity variations (LVVs). While LVVs are undoubt-  
 524 edly present within the Earth, numerous studies conclude that they generate minimal dif-  
 525 ferences in the geodynamical observations we explore here compared with those resulting  
 526 from variability in density inputs derived from different tomographic models (Moucha et  
 527 al., 2007; Ghosh et al., 2010; Yang & Gurnis, 2016). We therefore anticipate that our main  
 528 conclusions remain valid for reasonable amplitudes of LVV.

## 529 Appendix C Misfit to geodynamic observations

We assess model performance using a combined misfit function to assess compatibility  
 with geoid, dynamic topography and excess CMB ellipticity constraints. Following previous  
 studies (Steinberger & Calderwood, 2006; N. A. Simmons et al., 2009), we define the misfit to  
 geoid and dynamic topography based on variance reduction (VR), a proxy for the proportion  
 of observed signal explained by a given model prediction. Geoid misfit,  $\chi_N$ , is defined to be  
 equivalent to  $1 - \text{VR}_N$ , where  $\text{VR}_N$  represents geoid variance reduction, and is calculated  
 globally using

$$\chi_N = \frac{\sum_{l=2}^{l_{max}} \sum_{m=-l}^l (N_c^{lm} - N_o^{lm})^2}{\sum_{l=2}^{l_{max}} \sum_{m=-l}^l (N_o^{lm})^2} \quad (\text{C1})$$

where  $N^{lm}$  terms represent spherical harmonic coefficients of observed (subscript  $o$ ) and  
 predicted (subscript  $c$ ) geoid, and  $l_{max} = 30$  is the maximum spherical harmonic degree.



Dynamic topography misfit,  $\chi_A$ , is defined analogously to  $\chi_N$  (i.e.,  $\chi_A = 1 - \text{VR}_A$ ). However, since accurate residual depth measurements only exist at specific oceanic locations, rather than compare spherical harmonic coefficients, we instead determine this value in the spatial domain according to

$$\chi_A = \frac{\sum_{n_A=1}^{N_A} \left[ (A_c^i - A_o^i) - \overline{(A_c^i - A_o^i)} \right]^2}{\sum_{n_A=1}^{N_A} (A_o^i - \overline{A_o^i})^2} \quad (\text{C2})$$

where  $A^i$  terms are predicted and observed dynamic topography at  $N_A = 2278$  geographic locations (Hoggard et al., 2017), and values are weighted by the surface area of the  $1^\circ$  bin associated with each data point in order to correct for latitudinal variation in sampling density. Since excess CMB ellipticity is defined using a single spherical harmonic coefficient, rather than using a variance reduction-based misfit definition, we use the expression

$$\chi_C = \sqrt{\left( \frac{C_c^{20} - C_o^{20}}{\sigma_{C_o^{20}}} \right)^2} \quad (\text{C3})$$

for this component, which is similar to previous studies (Steinberger & Holme, 2008; N. A. Simons et al., 2009).  $C^{20}$  terms represent the  $l = 2, m = 0$  coefficient of observed and modelled core-mantle boundary topography, and  $\sigma_{C_o^{20}} = 100$  m based on the range of reported values (Gwinn et al., 1986; Mathews et al., 2002; Dehant et al., 2017). Finally, we sum each of these three components into a combined geodynamic misfit function,

$$\chi_G = \chi_N + \chi_A + \chi_C \quad (\text{C4})$$

530 In Figure 2, we present optimal results for the S10 viscosity profile (Steinberger et  
 531 al., 2010) and TX2011 tomographic model (Grand, 2002), whilst Figures S1–S6 display  
 532 results for other combinations. We select this tomographic model as it generates geodynamic  
 533 predictions with the lowest overall misfit. We choose the S10 (Steinberger et al., 2010)  
 534 viscosity profile over F10V1 (Forte et al., 2010)—despite the latter yielding lower misfits—  
 535 since it does not include a very low viscosity ( $7 \times 10^{19}$  Pa s) layer at the base of the transition  
 536 zone, which is considered to be controversial since it requires the entire region to be nearly  
 537 water-saturated ( $\sim 1.5\%$ ; Fei et al., 2017). Nevertheless, we acknowledge that a recent joint  
 538 analysis of geodynamic and seismic tomographic models is consistent with the presence of  
 539 a low-viscosity layer at 660 km depth (Rudolph et al., 2021).

## 540 Appendix D Body tide and Stoneley mode predictions

Modelling of Earth’s body tidal response requires models of 3D elastic, 3D density, and 1D anelastic structure (Lau et al., 2017). In the upper 400 km of the mantle, 3D elastic structure is determined using the calibrated parameterisation of SLNAAFSA to remove anelastic reductions in  $V_S$  from the seismic tomographic model, leaving only anharmonic  $V_S$  variations ( $V_S^{anh}$ ). Below 300 km,  $V_S^{anh}$  is derived from the tomographic values,  $V_S^{anel}$ , using radial changes in shear attenuation,  $Q_S^{-1}$ , from PREM and the expression

$$V_S^{anel} = V_S^{anh} \left[ 1 - \frac{Q_S^{-1}}{2 \tan(\pi\alpha/2)} \right] \quad (\text{D1})$$

where  $\alpha = 0.15$  (Dziewonski & Anderson, 1981; Karato, 1993; Widmer et al., 1992). While the resulting 3D  $V_S^{anh}$  model constrains the unrelaxed shear modulus, unrelaxed bulk modulus variations are obtained from  $V_\phi^{anh}$ , assuming that

$$R_b = \frac{\partial \ln V_\phi}{\partial \ln V_S} \approx \frac{\partial \ln V_\phi^{anh}}{\partial \ln V_S^{anh}} = 0.05 \quad (\text{D2})$$

541 and the radial  $V_\phi^{anh}$  profile can be determined using the same  $V_S$ – $V_\phi$  scaling as PREM  
 542 (Dziewonski & Anderson, 1981). The 1D anelastic structure applied to determine elastic

543 modulus dispersion at the 12-hour period of the M2 body tide adopts the mean value of  
 544  $Q_S^{-1}$  obtained from the calibrated parameterisation of SLNAAFSA at depths above 400 km,  
 545 and that of PREM at greater depths.

546 With the Earth model specified, the body tide response is computed using full-coupling  
 547 normal mode perturbation theory, with shear and bulk moduli dispersion calculated at tidal  
 548 frequencies using IERS standards (Widmer et al., 1992; Lau et al., 2015). Following Lau et  
 549 al. (2017), the fit between the predicted and observed in-phase M2 body tide displacement  
 550 is assessed at the sites of GPS stations by determining whether inclusion of 3D elastic and  
 551 density structure significantly enhances coherence between the two fields compared with  
 552 a baseline 1D model (PREM; Dziewonski & Anderson, 1981). The 3D Earth model is  
 553 only considered to yield a statistically significant improvement if the correlation obtained  
 554 between ‘raw’ and ‘corrected’ GPS residuals exceeds that obtained for the 1D model at  
 555 the 95% significance level, accounting for correlation between GPS estimates due to the  
 556 uneven spatial distribution of receivers. Raw residuals represent observed M2 body tide  
 557 displacements minus those predicted for the 1D model. Corrected residuals also account for  
 558 the effects of Moho and CMB excess ellipticity, Earth rotation and ocean tidal loading, and,  
 559 in the 3D model case, incorporate an additional correction for differences in the body tide  
 560 displacement predicted using 3D versus 1D structure.

561 To predict Stoneley mode splitting functions, 3D variations in  $V_S$ ,  $V_P$  and CMB to-  
 562 pography must be specified in addition to the density model (Resovsky & Ritzwoller, 1998).  
 563  $V_S$  anomalies are drawn directly from the tomographic model used to construct a given  
 564 density model, while  $V_P$  is determined by scaling  $V_S$  anomalies using a constant value of  
 565  $R_P = \partial \ln V_P / \partial \ln V_S = 0.5$  (Ritsema et al., 2004). We do not consider seismic anisotropy.  
 566 CMB topography is determined self-consistently using instantaneous flow simulations for  
 567 each density and viscosity model combination.

For a specified input velocity, density and topography model, Stoneley mode splitting  
 coefficients,  $\mathcal{C}^{st}$  can be calculated using the expression

$$\mathcal{C}^{st} = \int_{R_C}^{R_\oplus} d \ln \mathbf{M}^{st}(r) \cdot \mathbf{K}_M^s(r) r^2 dr + d \ln \mathcal{C}^{st} K_C^s \quad (\text{D3})$$

568 where  $d \ln \mathbf{M}^{st}(r)$  represents the prescribed 3D  $V_S$ ,  $V_P$ , and density heterogeneity at angular  
 569 degree,  $s$ , order,  $t$ , and radius,  $r$ .  $\mathbf{K}_M^s(r)$  are the relevant sensitivity kernels calculated using  
 570 PREM (Woodhouse, 1980; Dziewonski & Anderson, 1981),  $d \ln \mathcal{C}^{st}$  is the CMB topography  
 571 (the discontinuity most important for Stoneley modes), and  $K_C^s$  is the associated sensitivity  
 572 kernel.

The misfit between predicted and observed Stoneley mode splitting functions,  $\chi_S$  is

$$\chi_S = \frac{1}{N_S} \sum_{n_S=1}^{N_S} \frac{\sum_{s=2}^{s_{max}} \sum_{t=-s}^s (\mathcal{C}_c^{st} - \mathcal{C}_o^{st})^2}{\sum_{s=2}^{s_{max}} \sum_{t=-s}^s (\mathcal{C}_o^{st})^2} \quad (\text{D4})$$

573 where  $N_S = 9$  is the number of individual Stoneley modes investigated, the second sum-  
 574 mation term includes only even degree terms, where  $s_{max}$  is the maximum order. In most  
 575 calculations  $s_{max} = 2$ ; however, we also test the impact of setting  $s_{max}$  to the maximum  
 576 degree at which splitting function measurements are available for a particular mode, as well  
 577 as the consequences of adopting different misfit criteria (Text S1.4; Table S4) and ignoring  
 578 CMB topography (Text S2.3; Figure S12).

We combine  $\chi_S$  and  $\chi_G$  to yield a joint total misfit function,  $\chi_T$ , using

$$\chi_T = w_G \chi_G + w_S \chi_S \quad (\text{D5})$$

579 where  $w_G = 0.5$  and  $w_S = 5$ . These weightings result in misfit values with comparable  
 580 global minima.

## Acknowledgments

F.D.R. acknowledges support from the Imperial College Research Fellowship Scheme and the Schmidt Science Fellows program, in partnership with the Rhodes Trust. M.J.H. acknowledges support from the National Aeronautics and Space Administration (grant NNX17AE17G), the Donors of the American Chemical Society Petroleum Research Fund (59062-DNI8), and Geoscience Australia. M.J.H. and S.G. are supported by the G-ADOPT project, which is funded by the Australian Research Data Commons (<https://doi.org/10.47486/PL031>). P.K. acknowledges support from a Royal Society University Research Fellowship (URF\R1\180377). H.C.P.L. acknowledges support from NSF (EAR-192386). We thank D. Al-Attar, J. Austermann, L. Colli, D. R. Davies, R. Hawkins, M. Sambridge, Y. Takei, A. Valentine, N. White and J. Winterbourne for their help. Figures were prepared using Generic Mapping Tools software.

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