Figure 1.



Figure 2.



Figure 3.



Figure 4.



Figure 5.



Figure 6.



Figure 7.



Figure 8.



b)

Α



Figure 9.

"primary" plume

warm thermal

boundary layer \

hot thermal boundary layer

"secondary" plumes

LLSVP shallow domain (warm basaltic material)

LLSVP bottom domain or "DDD" (hot primordial material) abrupt mid-mantle / seismic gradients

> abrupt internal sub-vertical seismic gradients

> > abrupt lateral seismic gradients

Compositional layering within the Large Low Shear-wave Velocity Provinces in the lower mantle

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13

14 Abstract

15 The large low shear-wave velocity provinces (LLSVP) are thermochemical anomalies in the

16 deep Earth's mantle, thousands of km wide and \sim 1,800 km high. This study explores the

17 hypothesis that the LLSVPs are compositionally subdivided into two domains: a primordial

18 bottom domain near the core-mantle boundary and a basaltic shallow domain extending from

19 1,100~2,300 km depth. This hypothesis reconciles published observations in that it predicts that

20 the two domains have different physical properties (bulk-sound vs. shear-wave speed vs. density

anomalies), the transition in seismic velocities separating them is abrupt, and both domains

remain seismically distinct from the ambient mantle. We here report underside reflections from

the top of the LLSVP shallow domain, supporting a compositional origin. By exploring a suite of

two-dimensional geodynamic models, we constrain the conditions under which well-separated

25 "double-layered" piles with realistic geometry can persist for billions of years. Results show that

long-term separation requires density differences of $\sim 100 \text{ kg/m}^3$ between LLSVP materials,

27 providing a constraint for origin and composition. The models further predict short-lived

28 "secondary" plumelets to rise from LLSVP roofs and to entrain basaltic material that has evolved

in the lower mantle. Long-lived, vigorous "primary" plumes instead rise from LLSVP margins

30 and entrain a mix of materials, including small fractions of primordial material. These

31 predictions are consistent with the locations of hotspots relative to LLSVPs, and address the

32 geochemical and geochronological record of (oceanic) hotspot volcanism. The study of large-

33 scale heterogeneity within LLSVPs has important implications for our understanding of the

evolution and composition of the mantle.

35 **1. Introduction**

Thermochemical structures in the deep mantle bear witness of the Earth's thermal and chemical 36 37 evolution, and reflect large-scale mantle convection patterns. Seismic shear-wave and P-wave tomography reveals two antipodal large low shear-wave velocity provinces (LLSVP) in the 38 lowermost mantle, one beneath the Pacific and the other beneath Africa (e.g., Dziewonski et al., 39 40 2010). LLSVP locations are spatially correlated with ancient and modern hotspots, and hence 41 thought to play a key role in the generation of mantle plumes (Torsvik et al., 2006; Burke et al., 2008; Torsvik et al., 2010; Austermann et al., 2014; Davies et al., 2015; Doubrovine et al., 42 2016). Indeed, both LLSVPs are associated with broad dynamic topography anomalies at the 43 surface ("Superswells") that have been linked to large-scale mantle upwelling. LLSVPs have 44 been identified as potential geochemical reservoirs for materials that are sampled by hotspots 45 (e.g., Christensen and Hofmann, 1994). 46

Each of the LLSVPs is thousands of km wide and extends from the core-mantle boundary (CMB, 47 at 2,900 km depth) to mid-mantle (i.e., ~1,100 km) depths. As shown by Figure 1, such a large 48 depth extent of the LLSVPs is supported by seismic tomography models (e.g., Tanaka et al., 49 2009; Ritsema et al., 2011; Cottaar and Lekic, 2016), but somewhat remains a semantic issue 50 that depends on the definition of LLSVPs (see below). Using cluster analysis, Cottaar and Lekic 51 (2016) demonstrate that five different shear-wave velocity models agree well with each other in 52 terms of the geometry and depth extent of "slow clusters" (i.e. LLSVPs according to our 53 definition here; see their Figs. 4-5). Based on observations of abrupt lateral gradients in seismic 54 55 velocity across their boundaries (Ni et al., 2002; Ni and Helmberger, 2003; To et al., 2005; He 56 and Wen, 2009; Kawai and Geller, 2010; Sun and Miller, 2013; Thorne et al., 2013), anticorrelation between shear-wave and bulk-sound velocity (v_s and v_{ϕ}) anomalies (Su and 57 Dziewonski, 1997; Masters et al., 2000; Romanowicz, 2001; Trampert et al., 2004; Mosca et al., 58 2012; Koelemeijer et al., 2016; Tesoniero et al., 2016), as well as decorrelation of v_s with density 59 anomalies (Ishii and Tromp, 1999; Simmons et al., 2010; Mosca et al., 2012; Moulik and 60 Ekström, 2016), a purely thermal origin of LLSVPs remains unlikely. In turn, LLSVPs are 61 commonly interpreted as being hot but compositionally dense piles that are shaped by ambient-62 mantle convection (Davaille, 1999; McNamara and Zhong, 2005; Tan and Gurnis, 2007; 63 64 Deschamps and Tackley, 2008; Steinberger and Torsvik, 2012).

Two end-member scenarios have been proposed for LLSVP composition and origin. First, (1) 65 66 LLSVPs may dominantly consist of subducted oceanic crust (or MORB) that has largely segregated from harzburgite to accumulate in the deep mantle over geologic timescales 67 (Christensen and Hofmann, 1994; Xie and Tackley, 2004; Nakagawa and Tackley, 2005; 68 Brandenburg and van Keken, 2007). Depending on the efficiency of segregation, such a "basaltic 69 material" may also contain some small fraction of harzburgite. Second, (2) LLSVPs may be 70 71 composed of "primordial" materials, formed early in Earth's history (Deschamps et al., 2012; Li 72 et al., 2014; Nakagawa and Tackley, 2014). For example, compositional fractionation during 73 magma-ocean crystallization may have created large-scale density anomalies that are too strong

to be fully homogenized during billions of years of mantle convection (Labrosse et al., 2007;

75 Brown et al., 2014). Similarly, subducted (or foundered) Hadean proto-crust may have persisted

at the base of the mantle to contribute to the formation of LLSVPs (Boyet and Carlson, 2005;

77 Tolstikhin et al., 2006). Noble-gas isotope signatures of hotspot lavas as well as well gases

support the existence of primordial reservoirs somewhere in the mantle (*Mukhopadhyay*, 2012;

79 *Peto et al.*, 2013; *Caracausi et al.*, 2016), possibly spatially related to plume-source regions.

Furthermore, seismic evidence indicates that compositional heterogeneity is present within the 80 LLSVPs (Fig. 1). For example, the relatively shallow domain of the LLSVP is characterized by 81 moderately negative v_s (about -0.5% to -1% on average) in seismic tomography models, whereas 82 83 the bottom domain is characterized by much greater negative anomalies (Hernlund and Houser, 2008; Lekic et al., 2012). The robustness of this observation is indicated by an excellent 84 85 agreement among tomography models (Cottaar and Lekic, 2016), and an extensive illumination of LLSVPs by S-waves. The average transitional depth between domains is at ~2,300 km, or 86 ~600 km above the CMB. Abrupt vertical velocity gradients (in both v_p and v_s) have been 87 detected by waveform analyses in a similar depth range (2,200~2,500 km) (He and Wen, 2009; 88 Kawai and Geller, 2010; Sun and Miller, 2013; Frost and Rost, 2014), suggesting the transition 89 between domains reflects a compositional boundary. Hereinafter, we refer to the LLSVP bottom 90 domain as "Distinct Deep Domain", or DDD. Note that in the literature there is some ambiguity 91 in terms of the definition of the LLSVP. While the definition of the LLSVP in some papers (e.g., 92 Hernlund and Houser, 2008; Deschamps et al., 2012) is equivalent to that of the DDDs here, we 93 here refer to the LLSVPs in a broader sense (i.e. extending from the CMB to ~1,100 km depth; 94 see Fig. 1). For example, LLSVPs make up ~8% of the mantle's volume according to our 95 definition (Cottaar and Lekic, 2016); the DDDs make up 2%-3% (Hernlund and Houser, 2008). 96

97 If these systematic changes of seismic properties across LLSVPs indeed indicate an internal compositional boundary at 2,200~2,500 km depth, then any further study may yield insight into 98 the origin of materials that evolve deep in the mantle, and are recycled by upwelling plumes. For 99 example, the LLSVP shallow domain may be made up of basaltic materials or hot pyrolite (i.e., 100 101 purely thermal origin). DDDs may host primordial materials, or a mix of primordial and basaltic materials (Tackley, 2012). The presence of primordial materials within DDDs is supported by the 102 observed anti-correlation between v_s and v_{ϕ} , which is difficult to be explained by the presence of 103 basalt (and high-pressure polymorphs) alone (Deschamps et al., 2012). A composite primordial-104 basaltic reservoir in the mantle can further account for the geochemical signature of plume-fed 105 magmatism at major hotspots (e.g., Garapic et al., 2015). Whereas geodynamic studies of a 106 dense layer in the deep mantle have significantly advanced our understanding of LLSVPs 107 (McNamara and Zhong, 2005; Tan and Gurnis, 2007; Bower et al., 2013; Bull et al., 2014; Li et 108 al., 2014; Nakagawa and Tackley, 2014), heterogeneity within this layer has not been studied 109 110 systematically.

In this study, we investigate the geodynamical, geochemical and seismological implications of 111 large-scale compositional heterogeneity within LLSVPs. We use high-resolution two-112 dimensional geodynamic models to explore the conditions under which compositional layering 113 within thermochemical structures can be preserved over geologic timescales. LLSVP 114 115 compositional models are evaluated by comparing model predictions with published seismic observations, as well as with newly identified out-of-plane seismic reflections in the mid-Pacific. 116 Finally, we compare the predictions of our geodynamic models in terms of plume generation in 117 the lower mantle with the geochemical and geographical record of intraplate hotspot volcanism. 118

119

120 **2. Numerical Methods**

To model heat and material transport through the mantle, we solve the conservation equations of 121 mass, momentum and energy with the incompressible Boussinesq approximation using the finite-122 element code CitcomCU (Moresi and Gurnis, 1996; Zhong, 2006). We use a significantly 123 extended version of CitcomCU that can robustly solve for thermochemical convection (van 124 Hunen et al., 2005; Ballmer et al., 2009; 2010). Compositional properties are advected by 125 passive markers, or "tracers" (Gerva and Yuen, 2003). Any non-linear feedback effects of 126 thermochemical convection are addressed by using a second-order Runge-Kutta time-integration 127 128 scheme (Ballmer et al., 2009).

129 The rectangular two-dimensional numerical model domain is $z_{box} = 2,900$ km deep and $x_{box} =$

130 8,700 km wide, with a resolution of 320 x 1024 elements. Refinement of the vertical dimension

of the grid in the upper mantle slightly enhances the resolution in the upper mantle (i.e., 8.25 km)

132 compared to the lower mantle (i.e., 9.333 km).

Velocity boundary conditions involve free-slip on all boundaries except for the top, where velocities are set to zero (Fig. 2). No flow into or out of the box is allowed. Imposing a no-slip condition at the top reduces computational time, while having no significant impact on results. Tests show that the choice of the top velocity boundary condition neither affects the root-mean square velocities in the box, nor the distribution of materials after several billion years model time.

139 Thermal boundary conditions involve a fixed potential temperature of $T_m = 2,000$ °C at the bottom, and $T_0 = 0$ °C at the top. These conditions impose a superadiabatic temperature 140 difference of T_m between the CMB and surface. The bottom thermal boundary condition 141 corresponds to a "real" CMB temperature of ~3,000 °C, depending on the assumed geotherm 142 (which is not prescribed here). Such a CMB temperature is well within, but near the low end of 143 the range of estimates (Lay et al., 2008), but according to recent mineral-physics constraints any 144 higher values would likely require large-scale melting in the deepest mantle (Nomura et al., 145 2014), which is inconsistent with seismic data. Geotherms with relatively low T_m are also 146

147 generally consistent with one-dimensional seismic models such as PREM (*Deschamps and* 148 *Trampert*, 2004; *Mosca et al.*, 2012). As the modeled mantle is purely heated from the bottom 149 with no internal heating (and as the CMB area remains overestimated in Cartesian models of 150 mantle convection), the modelled CMB heat-flux is an upper bound and hence so are the density 151 anomalies required to stabilize thermochemical layers in the deep mantle (i.e., despite our 152 conservative choice of T_m).

153 Initial thermal conditions involve a mid-mantle potential temperature of 1,333.3 °C with 154 boundary layers at the top and bottom. Thermal boundary layers are computed as 80-Myr half-155 space cooling profiles. A small thermal random noise is added to the initial temperature field. In 156 any case, we find that model results are insensitive to the initial thermal condition.

The compositional field involves three major components. (1) Pyrolite is the typical upper-157 158 mantle rock (Workman and Hart, 2005), similar in composition to lherzolite (and high-pressure polymorphs). We refer to (2) "basaltic material" as a fine-scale mixture of mostly mafic (e.g., 159 high-pressure polymorphs of MORB) and some ultramafic mantle rocks (e.g., lherzolite or 160 harzburgite). We refer to (2) "primordial material" as being formed during the first ~100 Myrs of 161 Earth's history, and to have remained mostly unprocessed by subsequent mantle melting or 162 metasomatism, but do not impose a specific composition (see discussion section). Whereas the 163 models are designed to simulate these three materials (according to our final interpretations, see 164 below), our results only depend on the material properties modelled. 165

The compositional field is initially three-layered and stably stratified (Figure 2). The primordial 166 layer initially extends from the CMB to 250 km above the CMB, the basaltic layer extends from 167 250 km above the CMB to 2,100 km depth, and pyrolite makes up most of the rest of the mantle. 168 In the sublithospheric mantle and lithosphere, the composition is all pyrolite at ~ 100 km depth 169 and then transitions to all harzburgite at the surface (Fig. 2). This transition represents the 170 depletion of pyrolite due to partial melting as calculated from a separate numerical model of 171 mid-ocean ridge melting according to Ballmer et al. (2009). Note however that depleted pyrolite 172 173 largely remains in the nearly stationary lithosphere through model evolution; its presence does not have any important effects on model results. 174

By initially imposing the modeled materials as layers, we focus on studying the stability and 175 entrainment of basaltic and primordial materials in the lower mantle. While an initial condition 176 with a layer of primordial material near the base of the mantle is realistic (i.e., following e.g. a 177 global-scale gravitational cumulate overturn (Elkins-Tanton et al., 2005), or basal-magma ocean 178 179 freezing (Labrosse et al., 2007)), MORB is more likely to have progressively accumulated in the lower mantle as a consequence of deep subduction. Since the mechanisms for MORB-180 harzburgite separation remain ill-constrained, we choose not to explicitly model the processes of 181 deep subduction and segregation. Uncertainties in the efficiency of this segregation are reflected 182 by the density anomaly of the initially imposed layer of basaltic material, which is varied as a 183 free model parameter (see below). Variable densities of the basaltic material thereby reflect the 184

content of ultramafic components within the dominantly basaltic mechanical mixture. In
addition, these densities reflect the specific composition in major oxides of the mafic component
(i.e., MORB) itself, which may have important effects on dynamics (*Nakagawa et al.*, 2010).

In our thermochemical models, lateral density variations due to thermal and compositionalanomalies drive mantle flow:

190

$$\rho = \rho_0 - \alpha (T - T_m) \rho_0 + X_{PRI} \Delta \rho_{PRI} + X_{BAS} \Delta \rho_{BAS} + F \Delta \rho_F, \tag{1}$$

191

with ρ density; ρ_0 reference density; T temperature; α thermal expansivity; F depletion of 192 pyrolite; X_{BAS} and X_{PRI} volume fractions of basaltic and primordial material, respectively; and 193 $\Delta \rho_{BAS}$, $\Delta \rho_{PRI}$ and $\Delta \rho_F$ the density anomalies related to these two materials and depleted pyrolite, 194 respectively. Note that $\Delta \rho_{BAS}$ and $\Delta \rho_{PRI}$ are positive, whereas $\Delta \rho_F$ is negative (-100 kg/m³). 195 According to the objectives of this study, we carefully account for the depth dependency of 196 thermal expansivity α , as well as of the density anomalies related to basaltic and primordial 197 material ($\Delta \rho_{BAS}$ and $\Delta \rho_{PRI}$) in the lower mantle (Fig. 3). α is fixed in the upper mantle at 3.0·10⁻⁵ 198 K^{-1} and linearly decreases with depth in the lower mantle (from 2.5 $\cdot 10^{-5}$ K⁻¹ at 660 km depth to 199 1.25·10⁻⁵ K⁻¹ at the CMB), generally consistent with Tosi et al. (2013). In the upper mantle, $\Delta \rho_{BAS}$ 200 and $\Delta \rho_{PRI}$ are set to $\Delta \rho_{BAS,UM}$ and $\Delta \rho_{PRI,UM}$, respectively, except for the depth range of 300 km to 201 410 km, where $\Delta \rho_{BAS}$ is set to $2\Delta \rho_{BAS,UM}$ (Aoki and Takahashi, 2004). In the lower mantle, $\Delta \rho_{BAS}$ 202 and $\Delta \rho_{PRI}$ linearly decrease from $\Delta \rho_{BAS,UM}$ to $\Delta \rho_{BAS,CMB}$ and $\Delta \rho_{PRI,UM}$ to $\Delta \rho_{PRI,UM}$, respectively, 203 except for the depth range of 660-720 km where $\Delta \rho_{BAS}$ is set to $-3\Delta \rho_{BAS,UM}$ (*Hirose et al.*, 1999; 204 Xu et al., 2008). In this depth range, $\Delta \rho_F$ is also reversed to +100 kg/m³. $\Delta \rho_{BAS,UM}$, $\Delta \rho_{BAS,CMB}$, 205 $\Delta \rho_{PRLUM}$ and $\Delta \rho_{PRLCMB}$ are free model parameters. Accordingly, we explore the effects of 206 variations in density as well as compressibility (or $\partial \rho / \partial z$) of both basaltic and primordial 207 materials on thermochemical mantle convection. 208

In our simplified models, mantle viscosity η depends on temperature and depth only: 210

$$\eta = \eta_0 \exp\left(\frac{E^* + \rho_0 g z V^*}{RT} - \frac{E^*}{RT_m}\right),\tag{2}$$

211

with η_0, g, z, E^*, V^* and R reference viscosity, gravity acceleration, depth, activation energy, 212 activation volume and the ideal gas constant, respectively. Applying even just our conservative 213 choices for E^* and V^* , viscosity contrasts over the full mantle temperature and depth range reach 214 eight and five orders of magnitude, respectively. The total viscosity contrast across the model 215 domain is cut off (at six orders of magnitude) to ensure numerical stability, and to limit the 216 strength of the upper lithosphere, which in nature deforms plastically. The specific cutoffs (at 217 $1.4 \cdot 10^{17}$ Pa·s, and $1.4 \cdot 10^{23}$ Pa·s) are chosen such that mantle upwellings are better resolved than 218 downwellings. Downwellings are rather diffuse and not slab-like, but note that the rheology of 219

downwellings in the deep mantle, i.e. where they interact with thermochemical piles, remains poorly understood. A viscosity jump at 660 km depth is not explicitly modeled, but the imposed dependency of viscosity on depth (and temperature) creates a realistic average viscosity profile (Fig. 3a) (*Rudolph et al.*, 2015). According to the effective upper-mantle and lower-mantle viscosities in our models, the effective Rayleigh numbers in are $\sim 2.6 \cdot 10^8$ and $\sim 1.2 \cdot 10^6$, respectively. All other parameters are chosen to represent the conditions of the lower mantle (Table 1), the dynamics of which we focus on in this study.

In the model suite, we specifically explore ~70 cases with different $\Delta \rho_{BAS,UM}$, $\Delta \rho_{BAS,CMB}$, $\Delta \rho_{PRI,UM}$ and $\Delta \rho_{PRI,CMB}$ (see Table 1, Fig. 3b). The initial condition (e.g. initial volumes of basaltic and primordial materials; Fig. 2) and other parameters (see methods) remain fixed. Thereby, we focus on investigating the effects of the intrinsic density anomalies of basaltic and primordial materials relative to the (less dense) pyrolite, as well as the slopes of their depth-dependencies in the lower mantle:

- 233 $\partial \Delta \rho_{BAS} / \partial z = (\Delta \rho_{BAS,UM} \Delta \rho_{BAS,CMB}) / h_{LM}$
- 234 and $\partial \Delta \rho_{PRI} / \partial z = (\Delta \rho_{PRI,UM} \Delta \rho_{PRI,CMB}) / h_{LM}$

with lower mantle height $h_{LM} = 2,900 \text{ km} - 660 \text{ km}$. We chose to vary the above four parameters, because they are ill-constrained due to mineral-physics uncertainties (*Deschamps et al.*, 2012) and have been shown to have first-order effects on thermochemical convection (*Tan and Gurnis*, 2005; 2007). Note that $\partial \Delta \rho_{BAS}/\partial z$ and $\partial \Delta \rho_{PRI}/\partial z$ are directly related to (and thus representative of) relevant physical properties. They are generally proportional to the compressibilities of basaltic and primordial materials in the lower mantle, respectively, and inversely proportional to their bulk moduli.

Geodynamic models were run for simulation times of at least 3 Gyr, to approach a statistical steady state. We refrain from running the models for much longer timescales (e.g. ~4.5 Gyrs), because the style of surface tectonics and mantle convection in the early Earth remains poorly understood. Also given the boundary conditions (see above) and fixed parameters (Table 1), which are inspired by modern-mantle values, our numerical experiments represent near steadystate models of modern-mantle dynamics rather than mantle evolution models.

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3. Results of Numerical Models

As a function of model parameters, four regimes of thermochemical convection in the deep mantle are manifested (Fig. 4). In regime I, the primordial (and basaltic) material covers most of the CMB in a nearly continuous layer. In regime II, thermochemical piles emerge mostly consisting of primordial material at the bottom plus variably small volumes of basaltic material at the top, but there is significant mixing of basaltic and pyrolytic material. In regime III, thermochemical piles also emerge, but separation between basaltic and primordial material within these piles does not persist for geological timescales. In regime IV, the basaltic and primordial layers together form a "double-layered" thermochemical pile (leaving large areas of the core-mantle boundary uncovered) with two separate compositional domains. The regimes are described in detail in sections 3.1-3.2 and Figures 5-7. The criteria used to distinguish between these regimes are the coverage of the CMB by primordial material, and the persistence of coherent domains of primordial and basaltic materials after 3 Gyrs model time (see Table 2).

The sensitivity of these different regimes on model parameters is visualized in Figure 4a as a 262 regime map in terms of two key quantities: (1) A measure of $\Delta \rho_{PRI}$ in the deep mantle, here 263 evaluated at 2,300 km depth (i.e., near DDD tops; $\Delta \rho_{PRL,2300} = 0.268 \Delta \rho_{PRL,660} + 0.732 \Delta \rho_{PRL,CMB}$), 264 and (2) a measure of $\Delta \rho_{BAS}$ in the mid-mantle, here evaluated at 1,000 km depth (i.e., near 265 LLSVP tops; $\Delta \rho_{BAS,1000} = 0.848 \Delta \rho_{BAS,660} + 0.152 \Delta \rho_{BAS,CMB}$). High $\Delta \rho_{PRI}$ in the deep mantle (i.e., 266 high $\Delta \rho_{PRI,2300}$) promotes efficient settling of primordial material at the CMB (regime I). For low-267 268 to-moderate $\Delta \rho_{BAS}$ in the mid-mantle (i.e., low-to-moderate $\Delta \rho_{BAS,1000}$), the basaltic layer is readily entrained, and thus partially or completely lost over timescales of hundreds of millions to 269 billions of years (regime II). For low $\Delta \rho_{PRI,2300}$, mixing between the primordial and basaltic 270 layers occurs (regime III), mostly promoted by a small difference between $\Delta \rho_{PRI}$ and $\Delta \rho_{BAS}$ neat 271 the CMB (Fig. 4b). For the remaining parameter space with intermediate $\Delta \rho_{PRI}$ and $\Delta \rho_{BAS}$, 272 "double-layered" thermochemical piles persist for >3 Gyrs model time (regime IV). 273

274 <u>3.1 Thermochemical convection in the deep mantle (regimes I-III):</u>

Figure 5a shows an example case for regime I (case X5LH). In regime I, the drag exerted by 275 mantle convection is insufficient to significantly pile up the hot primordial material due to high 276 $\Delta \rho_{PRL2300}$ (and $\Delta \rho_{PRLCMB}$). Instead, the primordial material forms an almost continuous layer at 277 the CMB with some topography (heights ranging from ~150 km to ~600 km), and the basaltic 278 material piles up on top of this layer. In some cases, even the basaltic material covers part of the 279 CMB. Regime I is manifested for $\Delta \rho_{PRI,2300} > \sim 180 \text{ kg/m}^3$ and high $\Delta \rho_{BAS}$ (grey circles in Fig. 280 4a). High $\Delta \rho_{BAS}$ in addition to high $\Delta \rho_{PRI}$ are required, because at low $\Delta \rho_{BAS}$ thermochemical 281 convection switches to regime II, in which the basaltic load is removed from the primordial layer 282 due to entrainment (see below) such that CMB coverage is ultimately reduced. 283

In regime II, basaltic material is entrained by ambient mantle convection and progressively 284 eroded from the thermochemical piles (see Fig. 5b-c). Thereby, the rate of erosion is a function 285 of model parameter $\Delta \rho_{BAS,1000}$. In sub-regime IIa ($\Delta \rho_{BAS,1000} < -80$ kg/m³), positive (i.e. 286 destabilizing) thermal buoyancy overcomes negative compositional buoyancy, and strong 287 erosion occurs due to gravitational instability. For example, we observe that large chunks of the 288 basaltic layer episodically rise as a whole in case 74KF (see Fig. 5b; red circles in Fig. 4a). In 289 sub-regime IIb ($\Delta \rho_{BAS,1000} < \sim 90 \text{ kg/m}^3$), moderate erosion occurs, mainly due to entrainment by 290 plumes, and a significant amount of basaltic material survives even after 3 Gyrs model time (e.g., 291 case 94KF in Fig. 5c; yellow circles in Fig. 4a). This surviving material forms a layer on top of 292

the primordial material. In both sub-regimes, any eroded basaltic material undergoes convective mixing to be dispersed through the mantle. In many cases, it tends to accumulate in the transition zone, which acts as a "density trap" for basalt (*Nakagawa and Buffett*, 2005; *Ballmer et al.*, 2015b).

In regime III, mixing between primordial and basaltic materials occurs due to a relatively small 297 density difference between those two materials in the deep mantle. For $\Delta \rho_{PRL2300} - \Delta \rho_{BAS,2300} <$ 298 $\sim 100 \text{ kg/m}^3$, the thermal buoyancy of hot primordial material is sufficient to overcome its 299 intrinsic negative buoyancy, at least relative to the basaltic material (Fig. 5d). Figure 4b shows 300 that $\Delta \rho_{PRI,2300} - \Delta \rho_{RAS,2300}$ controls the rate of mixing (or the amount of primordial material that 301 can remain relatively "pure" after 3 Gyrs model time). Depending on parameters, we find that 302 303 thermochemical structures in regime III range from partially mixed piles (sub-regime IIIb; see example case X5JB in Fig. 5d; light blue circles in Fig. 4) to fully mixed piles with a more-or-304 less homogenous distribution of basaltic and primordial material throughout the whole pile (sub-305 regime IIIa; dark blue circles in Fig. 4 for $\Delta \rho_{PRL2300} - \Delta \rho_{BAS,2300} < \sim 90$ kg/m³). Whereas relatively 306 pure primordial material in regimes I, II and IV is usually sufficiently dense to avoid significant 307 erosion, a mix of primordial and basaltic materials (or previously termed "basal mélange" 308 (Tackley, 2012)) is commonly entrained by plumes. Therefore, the efficiency of mixing between 309 primordial and basaltic materials (regime III vs. regimes I/II/IV) has important implications for 310 the storage timescales of materials in the deep mantle (see discussion section). 311

312 <u>3.2 Double-layered thermochemical piles (regime IV):</u>

For moderate $\Delta \rho_{PRL2300}$ and $\Delta \rho_{BAS,1000}$ (Regime IV; Fig. 4a), mixing rates between all the three 313 314 materials modeled are significantly smaller than for regimes II and III (Table 2). Accordingly, only limited mixing between materials occurs over 3 Gyrs model time (e.g. reference case A5LE, 315 see Fig. 6a). The well-preserved basaltic and primordial materials form composite "double-316 layered" thermochemical piles surrounded by ambient-mantle material (Fig. 4a). The primordial 317 material forms the bottom layer of the double-layered pile. This layer displays significant 318 319 topography extending from the CMB to about 300-700 km above the CMB. It can intermittently peak at heights of ~1,000 km above the CMB, or higher. The basaltic material accumulates on 320 top of the primordial layer like a huge dome reaching depths of ~1,100 km (or heights of ~1,800 321 km above the CMB). This vertical extent is time-dependent: due to the competition between 322 323 thermal and compositional buoyancy forces, as well as variable drag exerted by ambient-mantle convection, the double-layered thermochemical pile oscillates as a whole (see also Davaille et al. 324 (1999)). The basaltic layer is generally hotter than the ambient mantle but cooler than the 325 primordial layer (Fig. 6b). 326

Figure 7 shows snapshots of net density anomalies for the reference case, combining the positive thermal and negative compositional contributions. These net density anomalies (and not the intrinsic density anomalies $\Delta \rho_{BAS}$ and $\Delta \rho_{PRI}$ alone) ultimately control mantle flow and (together with bulk and shear moduli) seismic velocities. For example, warm colors in Figure 7 mark

domains that are relatively buoyant (compared to cold colors). Whereas the bottom primordial 331 layer of the LLSVP is overall significantly denser (by about 50 kg/m³ for cases in regime IV) 332 than the global average, most of the shallow basaltic layer is only slightly denser than the global 333 average (by $\sim 15 \text{ kg/m}^3$). These model predictions are well within the range of normal-mode 334 constraints (Ishii and Tromp, 1999; Trampert et al., 2004; Moulik and Ekström, 2016; 335 Koelemeijer et al., in review). Only the very top of the basaltic layer displays a somewhat 336 stronger net-density anomaly (~30 kg/m³), because $\Delta \rho_{BAS}$ increases upwards ($\Delta \rho_{BAS,UM}$ > 337 $\Delta \rho_{BAS CMB}$ in most of the cases modeled, consistent with mineral-physics data (Xu et al., 2008; 338 Ricolleau et al., 2010; Deschamps et al., 2012). These vertical changes of net density anomaly 339 within the basaltic layer account for its dome-like shape (see Tan and Gurnis (2005; 2007)). In 340 contrast, vertical changes of net density anomaly across the primordial layer are small (at least 341 compared to the global average), which thus forms ridge-like piles. The configuration and shape 342 of primordial ridge-like piles is strongly time-dependent and sensitive to ambient-mantle flow, 343 344 consistent with previous work (e.g., McNamara et al., 2010). A thin thermal boundary layer with strongly negative net density anomaly forms at the CMB (i.e., outside the piles), as well as along 345 the flanks of the hot primordial piles. Other thermal boundary layers, somewhat less pronounced, 346 form at the top of the basaltic dome, and along the internal boundary of the composite 347 348 thermochemical pile.

Two different types of mantle plumes rise from two different thermal boundary layers (see Figs. 349 6-7 and Movies S1-S3). Vigorous plumes rise out of the hot thermal boundary layers along the 350 flanks of the double-layered thermochemical pile. The bases of these plumes are relatively 351 stationary over long timescales (100s of Myrs), as they are anchored by one of the peripheral 352 ridges of primordial material. They move at rates of less than ~0.3 km/Myr (i.e. relative to 353 LLSVPs, which themselves are inferred to be stationary (Torsvik et al., 2010)). Less vigorous, 354 and somewhat cooler plumes rise from the warm thermal boundary layer at the top of basaltic 355 domes. They are usually short-lived, and their bases move swiftly relative to the LLSVPs (i.e., at 356 rates of up to several km/Myr). 357

In addition, a major plume pulse occasionally rises out of the thermochemical pile itself (see Figures 7e-f). In the models, a plume pulse is triggered by a combination of ambient mantle flow and build-up of heat within (or near) the pile. Such a pulse can deliver a lot of heat (and potentially basaltic material) into the upper mantle and may support large-scale asthenospheric melting, perhaps to sustain flood-basalt volcanism (see *Lin and van Keken*, 2005; *Sobolev et al.*, 2011b).

364

4. Comparison of model predictions with seismic constraints

366 Our geodynamic models predict that hot and compositionally distinct rock forms large piles at 367 the base of the mantle, similar in geometry to LLSVPs as imaged by seismic tomography

(Cottaar and Lekic, 2016). In regime I, thermochemical piles are predicted to largely cover the 368 CMB, inconsistent with the footprint of LLSVPs constrained by tomography (i.e., 20%~30% of 369 CMB area, or ~50% in a cross-section along a great circle through both LLSVPs). In regimes IIa 370 and IIIa, piles are predicted to be internally homogeneous in composition. This type of 371 372 thermochemical convection in the deep mantle, whether containing almost purely primordial piles (IIa) or well-mixed basaltic-primordial piles (IIIa), has been extensively discussed in the 373 literature (see references in Tackley, 2012; Hernlund and McNamara, 2015). In all other cases, 374 moderately heterogeneous (incipient mixing, regime IIIb) or strongly heterogenous piles are 375 predicted to form. Strongly heterogeneous piles display well-separated layers of basaltic and 376 primordial materials (regimes IIb and IV). 377

Pronounced compositional heterogeneity within LLSVPs, as predicted for regimes IIb and IV, is supported by seismic constraints. Reflections deep within the Pacific LLSVP cannot be explained by the iso-chemical perovskite to post-perovskite phase transition and hence indicate compositional heterogeneity (*Cobden and Thomas*, 2013). Analysis of P-waveforms supports heterogeneity by revealing complex seismic structure near both the eastern and western edges of the Pacific anomaly (*Konishi et al.*, 2014; *Tanaka et al.*, 2015). Also, some regions deep within the LLSVPs appear much slower than expected for purely thermal effects (*To et al.*, 2016).

At somewhat shallower depths, abrupt vertical gradients in v_s may be indicative of large-scale 385 sub-horizontal compositional layering within LLSVPs. These vertical gradients retrieved from 386 waveform-modeling studies occur at heights of ~400 to ~700 km above the CMB beneath the 387 Pacific (Takeuchi et al., 2008; He and Wen, 2009), as well as the western flank of the African 388 LLSVP (Sun and Miller, 2013), and at up to ~1,200 km above the CMB beneath South/East 389 Africa (Ni and Helmberger, 2003; Wang and Wen, 2007). Abrupt vertical gradients are here 390 interpreted to occur across interfaces internal to the larger LLSVP domains (i.e., the top of the 391 392 DDD), because they are often located well within LLSVPs, which extend from the CMB to ~1.800 km above the CMB (Figure 1). This large vertical extent of LLSVPs (or "slow clusters") 393 and large degree of internal variability is supported by cluster analysis of five recent tomography 394 models (Cottaar and Lekic, 2016). Also of note is that abrupt vertical gradients roughly coincide 395 396 with the depth range of pronounced radial changes in LLSVP seismic properties (i.e., constrained by tomography): in terms of average v_s of slow clusters (*Lekic et al.*, 2012), decorrelation of v_s 397 and density (Mosca et al., 2012; Moulik and Ekström, 2016), as well as anti-correlation between 398 v_s and v_{ϕ} (Masters et al., 2000; Romanowicz, 2001; Koelemeijer et al., 2016; Tesoniero et al., 399 2016). 400

We interpret these changes in seismic properties to reflect a compositional boundary within the LLSVPs. Such heterogeneity within LLSVPs may be attributed to various (compositional) models. For example, (1) the LLSVP shallow domains may be composed of basaltic materials (i.e., rock assemblages with a high content of MORB), and the bottom domain (or DDD) of

405 primordial materials, or (2) vice-versa. Alternatively, (3) the LLSVP shallow domain may be

406 made up of hot pyrolite (i.e., purely thermal origin), and the DDD of primordial and/or basaltic407 materials.

A primordial origin of DDDs such as in scenarios (1) and (3) above is supported by seismic 408 constraints. The observed anti-correlation between v_s and v_{ϕ} in general (Deschamps et al., 2012), 409 and its restriction to the DDDs (Koelemeijer et al., 2016; Moulik and Ekström, 2016; Tesoniero 410 411 et al., 2016) in particular, is best explained by the presence of materials enhanced in Fe-rich bridgmanite, potentially with primordial origin. Based on the spatial distribution of the anti-412 correlation (Koelemeijer et al., 2016; Moulik and Ekström, 2016; Tesoniero et al., 2016), an 413 alternative iso-chemical explanation for the anti-correlation appears require that post-perovskite 414 is stable within the hot DDDs. In turn, Fe-rich (Mg,Fe)SiO₃ bridgmanitic materials can account 415 416 for the observed anomalously high densities and v_{ϕ} in DDDs through the combined effects of Fe on density, and those of (Mg,Fe)SiO₃ on bulk modulus and thus v_{ϕ} (Deschamps et al., 2012; 417 418 Wolf et al., 2015). A relatively high intrinsic bulk modulus can account for the observed anticorrelation, and is consistent with the parameter range spanned by our preferred models (which 419 420 display positive $\partial \Delta \rho_{PRI}/\partial z$, and thus are more compressible than pyrolite). The formation of materials strongly enhanced in bridgmanite has been indeed linked to primordial processes such 421 as magma-ocean crystallization (Elkins-Tanton, 2008), since Mg-rich bridgmanite is the magma-422 ocean liquidus phase over a wide pressure range (de Koker et al., 2013; Tateno et al., 2014), but 423 a mechanism for subsequent Fe-enrichment remains elusive. Other viable (primordial) 424 candidates for DDD composition involve foundered CaSiO3-rich proto-crust (Kawai and 425 Tsuchiya, 2014) as well as anomalously reduced bridgmanitic materials (Gu et al., 2016). 426

To test whether LLSVP shallow domains are dominantly basaltic (scenario (1) above) as in 427 regimes IIb/IV, or pyrolytic in composition (scenario (3) above) as in regime IIa, we analyze 428 seismic array data. Note that seismic tomography is insufficient to distinguish between these two 429 430 end-members because of a nearly perfect trade-off between temperature and basalt content when predicting seismic velocities at mid-mantle depths (Wentzcovitch et al., 2010; Deschamps et al., 431 2012). We focus on the vertical component of arrivals in western North America that are related 432 to events in the western Pacific. We detected signals arriving at the array from a different 433 434 direction than the main phases of the events. Using the directivity information given by the backazimuth and slowness as well as the travel-time of each signal, the ray-paths and reflection 435 points are reconstructed by back-tracing through a one-dimensional velocity model (we use 436 ak135 (Kennett et al., 1995)). The data processing is explained in detail in Schumacher and 437 Thomas (2016). Figure 8a shows the locations of the events (red stars), the reference stations 438 439 (black triangles), and the reflection points (circles, depth indicated by color) which are able to explain the measured backazimuth, slowness and traveltime of our out-of-plane signals. 440

Two reflection points are located in the mid mantle below the Pacific at a depth of ~1,500 km. They can be explained by P-to-P underside reflections off the top of the LLSVP shallow domain as predicted by mantle-convection models in regimes IIb and IV. With respect to uncertainties in the back-tracing routine as well as seismic tomography, the two reflection points agree with the upper bound of the low velocity region in the S40RTS tomography model (Fig. 8b). Another two
reflection points in the same region are located deeper, well within the interior of the LLSVP, as
imaged by tomography, in a depth of around 2250km and 2550km. The observed signals could
be caused by P-to-P underside reflections off an internal compositional domain boundary, such
as between the DDD and LLSVP shallow domain.

450 The underside reflections retrieved here thus lend credibility to a double-layered model for LLSVP composition, e.g. with a deep primordial domain and a shallow basaltic domain. In 451 particular, the two reflections at $\sim 1,500$ km depth appear to be at odds with a purely-thermal 452 LLSVP shallow domain, indicating a sub-horizontal compositional boundary such as between 453 basaltic materials and ambient-mantle pyrolite. Note that a purely-thermal shallow domain is 454 455 expected to be structurally dominated by the presence of sub-vertical plumes. Thus, the underside reflections retrieved here corroborate a (thermo-)chemical origin for the whole 456 457 LLSVPs (deep plus shallow domains) with a total volume of ~8% of the mantle (Cottaar and Lekic, 2016). However, future systematic studies of underside reflections are certainly needed to 458 459 support this interpretation, and to rule out any alternative scenarios. For example, we cannot exclude the possibility that individual reflections are simply caused by the marble-cake structure 460 of the mantle. 461

Nevertheless, mostly based on the geometry of reflection points, both near the roof and within 462 LLSVPs, we prefer a scenario with hot and compositionally "double-layered" thermochemical 463 piles (Fig. 8). As another line of evidence, note that a reservoir for the long-term (>3 Gyr) 464 storage of subducted (basaltic) materials somewhere in the (lower) mantle is required by hotspot-465 lava isotope geochemistry (Cabral et al., 2013). If the DDDs are indeed primodial in origin (see 466 above), then the LLSVP shallow domains are perhaps the best candidate to host this reservoir. 467 Along these lines, the predictions of cases in regimes IIb and IV (Figs. 5c, 6) with composite 468 469 piles of primordial (bottom) and basaltic (shallow domains) materials are most consistent with observations. In an alternative explanation, both LLSVP shallow domains and DDDs host 470 primordial materials, but with a distinct major-element composition in each domain (i.e., 471 different kinds of primordial materials, for example recording different processes). While this 472 473 scenario is fully consistent with the seismic observations presented here, it does not offer an opportunity for the long-term storage of subducted materials. In any case, as our model results 474 exclusively depend on the material properties modelled, predictions for regimes IIb and IV are 475 also applicable to this alternative scenario. 476

By visual analysis, cases in regime IV appear to be in better agreement with seismic observations than those in regime IIb, because the volume of basaltic material preserved in the lower mantle is greater. Piles in regime IV extend upward from the CMB to depths of ~1,100 km depth, similar to depths inferred for LLSVP roofs (i.e., on the basis of cluster analysis of, and radial correlation functions of global seismic-tomography models (*Rudolph et al.*, 2015; *Cottaar and Lekic*, 2016); also see Fig. 1). In contrast, thermochemical piles in regime IIb do not extend as much above the CMB due to progressive erosion of basaltic material. Note however that seismic tomography

might somewhat overestimate the height of the piles, and more importantly that our geodynamic 484 models do not consider the replenishment of basalt by deep subduction (there is no explicit 485 subduction of basalt in our models). Partial erosion of the basaltic layer over 3 Gyr model time 486 such as predicted by cases in regime IIb can indeed be well compensated by subduction and 487 488 accumulation of MORB. For modern-style plate tectonics, MORB subduction on the order of $D_{CRUST} \cdot L_{SUBD} \cdot V_{SUBD}$ (with crustal thickness $D_{CRUST} \approx 7$ km, the length of all subduction zones 489 $L_{SUBD} \approx 60,000$ km and the average speed of plate convergence $V_{SUBD} \approx 50$ km/Myr) can 490 potentially replenish up to $\sim 7\%$ of the volume of the mantle (i.e., similar to the volume of 491 492 LLSVPs) per billion year. Considering this flux of subducted MORB, cases in regime IIb with moderate erosion of basaltic materials are equally or perhaps even more realistic than those in 493 regime IV. A mix of ancient and young (i.e., recently replenished) basaltic materials in the 494 lower-mantle source region of mantle plumes is consistent with isotope geochemical constraints 495 (Sobolev et al., 2011a; Cabral et al., 2013). 496

497 Another difference between regimes IIb and IV is in the slopes of the pile's sides, which are generally less steep in regime IIb than in regime IV. This difference can be understood if one 498 recalls that regime IIb manifests at lower $\Delta \rho_{BAS,1000}$ (Fig. 4a), and thus usually also at lower 499 $\Delta \rho_{BAS,1000} - \Delta \rho_{BAS,2300}$ (Table 2) than regime IV. The weaker depth-dependence of $\Delta \rho_{BAS}$ acts to 500 reduce the slope of the piles, because the (positive) pile density anomaly (or negative buoyancy) 501 in the deep mantle becomes larger at a given density anomaly at the top of the pile, or in other 502 words, because of a reduced "metastability" (Tan and Gurnis, 2007). Cluster analysis of v_s 503 tomographic models shows that LLSVPs consist of aggregations of meso-scale piles that exhibit 504 a wide range of morphologies, from gently or moderately steep slopes, to near vertical or even 505 overhanging (Cottaar and Lekic, 2016). This range of topographies is highly consistent with 506 findings from studies of P waves that report regionally varying LLSVP slopes (Frost and Rost, 507 2014). Such regional variations are indeed expected for LLSVPs with pronounced (lateral) 508 compositional heterogeneity. 509

510

511 **5. Discussion**

512 The above seismic constraints lend credibility to our models in regimes IIb and IV with composite "double-layered" thermochemical piles (Figs. 6-7). These two regimes are restricted 513 to a rather small parameter range in Figure 4, thus tightly bracketing model parameters. The 514 related bounds for basalt densities in the deep mantle and $\partial \Delta \rho_{BAS}/\partial z$ are generally consistent with 515 mineral-physics estimates (Ricolleau et al., 2010; Deschamps et al., 2012). In terms of the 516 density difference between (primordial?) DDD materials and pyrolite $\Delta \rho_{PRI}$, the models yield 517 bounds of about 160 kg/m³ to 190 kg/m³. These latter bounds correspond to relevant buoyancy 518 numbers $B = \Delta \rho / \alpha T_m \rho_0$ of ~1.1 to ~1.3 at the α and T_m assumed here. These B can potentially be 519 used to constrain DDD composition. Even tighter bounds arise from our models for the density 520 521 difference between basalt and (primordial) DDD materials deep in the mantle (100~120 kg/m³)

and corresponding B (0.7 \sim 0.8). Thus, better mineral-physics constraints on basalt density under 522 the relevant high pressure-temperature conditions may help to constrain the density, composition 523 and origin of DDDs. Note however that the above bounds may be sensitive to parameter choices 524 and model assumptions that have not been systematically explored here. For example, we did not 525 526 consider the effects of intrinsic viscosity variations between the materials, which should 527 influence mixing between materials and thermochemical-pile shapes (e.g., Bower et al., 2013), and thus density anomalies required for a specific regime. Also, we do not explore the rheology 528 of subducted slabs in the deep mantle systematically, or even the history of subduction, which 529 both affect the coupling between the pile(s) and the ambient mantle, and hence pile shapes (e.g., 530 Steinberger and Torsvik, 2012; Bull et al., 2014). In regimes IIa and IV, two distinct types of 531 plumes emerge from the LLSVPs (also see chapter 3.2). Hot and stationary plumes rise from the 532 LLSVP margins; somewhat cooler, episodic "plumelets" from LLSVP roofs. These predictions 533 can reconcile characteristic differences between "primary" and "secondary" plumes, as inferred 534 from a range of geophysical and geochemical observations (Courtillot et al., 2003). While 535 primary plumes are thought to ascend from the CMB, secondary plumes (or plumelets) are 536 thought to episodically rise from a thermal boundary layer in the mid-mantle (Davaille, 1999; 537 Tanaka et al., 2009; Saki et al., 2015), similar to our model predictions (Fig. 9). For example, 538 age-distance patterns as well as isotope geochemical signatures of South Pacific hotspots (i.e. 539 directly above the Pacific LLSVP) indicate episodic short-lived volcanism (≤20 Myrs) (Clouard 540 and Bonneville, 2005; Jackson et al., 2010), whereas e.g. Hawaii, Réunion and Iceland plumes 541 display much longer-lived volcanism (~100 Myrs or more). Accordingly, the rise of plumes both 542 from the margins as well as the roofs of double-layered LLSVPs as predicted by our models can 543 account for the diversity among hotspot volcanism worldwide, for example in terms of plume 544 excess temperatures and buoyancy fluxes (Sleep, 1990; Herzberg et al., 2007; Ballmer et al., 545 2015c). That the rise of plumes is not restricted to LLSVP margins (cf. Burke et al., 2008), but 546 547 also involves LLSVP roofs, is moreover consistent with statistical assessments of hotspot locations (Austermann et al., 2014; Davies et al., 2015). 548

549 The predictions of our models in terms of entrainment of materials in the lower mantle have implications for our understanding of ocean-island basalt (OIB) geochemistry. On average, 550 plumes in our models continuously entrain (and deliver into the asthenosphere) rather small 551 fractions of primordial materials (i.e., $\sim 3\%$ in regime IIb / $\sim 2\%$ in regime IV) in addition to 552 moderate fractions of basaltic material (~14% / ~8%). Entrainment of basaltic and primordial 553 material strongly varies between individual plumes, and over time for the same plume (see also 554 Deschamps et al., 2011; Williams et al., 2015). Both basaltic and primordial materials are more 555 efficiently carried by hot "primary" plumes than by warm "secondary" plumes, consistent with 556 geochemical observations (Courtillot et al., 2003; Sobolev et al., 2007). Since these dense 557 materials are more fusible than pyrolite, and are usually entrained into the hottest core of the 558 plume (Jones et al., 2016), they are expected to be over-represented in mantle melting and 559 560 associated hotspot lavas (Phipps Morgan, 1999; Ito and Mahoney, 2005; Ballmer et al., 2013). Strong but variable contributions of basaltic (i.e., mafic) materials are indeed consistent with 561

olivine major-element geochemistry of OIB (Sobolev et al., 2005; 2007; Herzberg, 2011). For 562 example, some primary hotspots such as Hawaii or Iceland have much more strongly mafic 563 sources than MORBs (Sobolev et al., 2007). In turn, if noble gases are indeed stored within the 564 primordial DDDs, then they should be more efficiently sampled by primary than by secondary 565 plumes. This prediction is consistent with the highest ³He/⁴He ratios being sampled at e.g. 566 Iceland and Hawaii hotspots. In more general terms, the predicted composite source for OIB 567 volcanism helps to understand the large variability spanned by isotope geochemical data, as well 568 as the intimate association of primordial and recycled geochemical signatures (Zindler and Hart, 569 1986; Hofmann, 1997; Koppers and Staudigel, 2005; Garapic et al., 2015). 570

A composite source for OIB volcanism may further provide an explanation for the geochemical 571 asymmetry as is observed across various oceanic hotspot tracks (Huang et al., 2011; Weis et al., 572 2011; Pavne et al., 2013; Jackson et al., 2014; Hoernle et al., 2015). For example, the Hawaiian 573 hotspot is separated into the "Loa" and "Kea" trends with distinct isotopic signatures and 574 contributions from mafic materials in mantle melting (Abouchami et al., 2005; Sobolev et al., 575 2005). Our calculations in preferred regimes IIb and IV predict "primary" plumes to rise from 576 the margins of double-layered piles and thus to entrain distinct types of materials on either side 577 of the plume (Fig. 7a). On the side facing away from the LLSVP, the plumes tend to entrain hot 578 and mostly pyrolytic material from the thermal boundary layer just above the CMB. On the other 579 side, they are predicted to entrain warm and partly basaltic material (plus some primordial 580 581 material) from the thermal boundary layer just above the LLSVP flank (including some LLSVP 582 material). These model predictions involve a thermal as well as a compositional gradient across major plumes. Thus, they may provide an explanation for the relationship of geochemical 583 asymmetry across hotspots with the underlying geometry of LLSVPs. However, since 584 superposition of thermal effects and compositional effects during plume ascent (in terms of 585 buoyancy, and fusibility during hotspot melting) may obscure the deep origin of these patterns 586 (Ballmer et al., 2015a; Jones et al., 2016), we motivate petrological studies of lateral thermal 587 variations across hotspots (cf. Xu et al., 2014), in addition to those of lateral isotopic variations. 588

Double-layering of materials within LLSVPs may moreover provide an explanation for the poor 589 expression of primordial reservoir(s) in hotspot lavas. Whereas ancient noble-gas signatures have 590 been well documented in OIB (Mukhopadhyay, 2012), indicating the persistence of reservoir(s) 591 that have formed within ~100 Myrs of Earth accretion, any such evidence from other isotopic 592 systems in modern OIB is still lacking. For example, the ¹⁸²Hf-¹⁸²W and ¹⁴⁶Sm-¹⁴²Nd systems 593 should record any early differentiation of the Earth, but until recently any ¹⁸²W or ¹⁴²Nd 594 595 anomalies have only been retrieved from high-precision analyses of Archean igneous rocks (see references in Brown et al., 2014). The lack of evidence from both these systems for modern OIB 596 suggests that any primordial reservoirs, if they indeed persist in the mantle, are rather well 597 598 hidden. Double-layering of LLSVPs with a bottom layer of primordial material that is shielded 599 by an overlying basaltic layer provides good conditions to promote persistence of primordial materials. Note that our models predict plumes to entrain only very small fractions of primordial 600

601 material, and that high-precision 182 W or 142 Nd measurements are yet only available for a small 602 subset of OIBs. Indeed, recent high-precision 182 W analyses indicate a primordial contribution to 603 Ontong-Java and North-Atlantic flood basalts (*Rizo et al.*, 2016), supporting our interpretations.

Mapping the internal LLSVP compositional boundary may thus quantify the volume of 604 primordial geochemical reservoirs, with implications for the Earth's chemical budget as well as 605 the distribution of radioactive isotopes through the mantle. According to our hypothesis (Fig. 9), 606 the volume(s) of any (enriched) primordial reservoirs correspond to that of the DDDs, which 607 have been estimated at 2-3% of the mantle's volume (Hernlund and Houser, 2008). Primordial 608 reservoir(s) enriched in iron (and thus possibly enriched in other incompatible elements) are 609 viable candidates for DDD composition, because iron-enrichment is required to reconcile DDD 610 611 physical properties (see above). This interpretation does not rule out any (depleted) primordial reservoir(s) to be dispersed through the ambient mantle (Becker et al., 1999). While the 612 613 requirement for the survival of a large or strongly-enriched primordial reservoir from early silicate-silicate fractionation (i.e., "early enriched reservoir" (EER) (Boyet and Carlson, 2005)) 614 has been removed by a better understanding of ¹⁴²Nd nucleosynthetic anomalies across the solar 615 nebula (Bouvier and Boyet, 2016; Burkhardt et al., 2016), a moderately-enriched primordial 616 reservoir similar in size to that of the DDDs cannot be ruled out at the current precision of ¹⁴²Nd 617 analyses. Moreover, the survival of a primordial reservoir that records early metal-silicate 618 fractionation due to core formation is evident by the recovered ¹⁸²W anomalies (see above; *Rizo* 619 620 et al., 2016). Independent of the specific origin and composition of any primordial reservoir(s), 621 we speculate that the African LLSVP may contain more primordial (and less basaltic) material (similar to regime IIb?) compared to the Pacific LLSVP (regime IV?), because abrupt gradients 622 tend to be closer to tomographically-inferred LLSVP boundaries (see detailed discussion in 623 Cottaar and Lekic, 2016). Such a possible difference in their make-up may account for 624 differences in shapes and volumes of these two major LLSVPs, and is consistent with the recent 625 history of basalt subduction. 626

We show that layering of materials within LLSVPs with a primordial bottom domain (or DDD) 627 628 and a basaltic shallow domain is geodynamically viable and can reconcile a range of seismic observations (see sections 3-4). LLSVP bottom layer(s) may host iron-enriched cumulates that 629 have been formed in the upper mantle during the late stages of magma-ocean crystallization, and 630 then foundered to the base of the mantle (Elkins-Tanton, 2008; Brown et al., 2014). 631 Alternatively, primordial heterogeneity may have originated from incomplete core-mantle 632 equilibration during multi-stage planetary accretion and core formation (Kaminski and Javov, 633 634 2013). The accumulation of subducted MORB on top of such primordial materials likely requires an efficient mechanism of basalt-harzburgite separation somewhere in the mantle. Separation 635 would be driven by the density difference between both these lithologies, but should only be 636 637 efficient in a region of low viscosities, for example in the mantle transition zone (Motoki and 638 Ballmer, 2015) or near the CMB (Tackley, 2011), where the stabilization of post-perovskite may reduce viscosity (Ammann et al., 2010). Quantitative integration of seismology, geodynamics 639

and mineral-physics approaches is needed to further constrain the heterogeneous compositionand origin of LLSVPs.

642

643 **6.** Conclusion

644 We here combine two key seismic observations to motivate a geodynamic study of 645 compositional layering within thermochemical piles. First, seismic tomography models agree well with each other, in that LLSVPs are broad features that extend from the CMB upward to 646 ~1,000 km depth (Fig. 1) (Cottaar and Lekic, 2016). Second, abrupt vertical gradients of seismic 647 wave speed as well as pronounced radial changes of LLSVP seismic properties (in terms of 648 average v_s , and the correlations between v_s and v_{ϕ} , as well as between v_s and net density) occur at 649 650 \sim 2.300 km depth within LLSVPs (see section 4), thereby separating the LLSVPs into a shallow domain and distinct deep domain (DDD). Our geodynamic models demonstrate that two layers 651 652 with distinct composition and physical properties can avoid significant mixing with each other (and with the rest of the mantle) over billions of years, forming double-layered composite piles 653 654 in the lower mantle. The parameter space, in which limited mixing occurs and piles with geometries similar to LLSVPs are formed, can inform about the specific compositions and 655 origins of heterogeneity within LLSVPs. 656

We hypothesize that LLSVP shallow domains are mostly basaltic in composition, and DDDs are 657 primordial in origin. A composition of LLSVP shallow domains that is distinct from the ambient 658 mantle (and likely basaltic) is supported by underside reflections that we detect near LLSVP tops 659 660 (see section 4). A primordial origin of DDDs is supported by the anti-correlation between v_s and v_{ϕ} as well as the decorellation of v_s and density, as are observed in the deep lower mantle 661 (Deschamps et al., 2012; Koelemeijer et al., 2016; Moulik and Ekström, 2016). Such large-scale 662 compositional layering within LLSVPs is reconciled by another set of underside reflections that 663 664 we detect at 2250-2550 km depth, as well as by published observations of abrupt vertical gradients that occur in a similar depth range. A composite make-up of the LLSVPs overall 665 (Figure 9) is consistent with geochemical constraints for the manifestation of long-lived 666 subduction-related as well as primordial reservoirs somewhere in the (lower) mantle, which are 667 668 variably tapped by mantle plumes (see section 5). Indeed, our geodynamic models predict mantle plumes to originate from the (1) flanks as well as (2) the roofs of double-layered thermochemical 669 piles, and to entrain a mix of materials. Model predictions in terms of location, vigor, episodicity 670 and materials entrained by these two types of plumes are generally consistent with those inferred 671 from observations for (1) "primary" and (2) "secondary" plumes (Courtillot et al., 2003). Future 672 modeling efforts are required to quantitatively test geodynamic-model predictions with seismic 673 observables as well as geochemical signatures of hotspot lavas. 674

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684 8. Figures and Tables

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Figure 1: Seismic profiles through the (A) African and (B) Pacific LLSVP. Colors show average

687 S-wave anomalies across five recent tomographic models: SPani (*Tesoniero et al.*, 2016),

688 SAVANI (Auer et al., 2014), S40RTS (Ritsema et al., 2011), "ME2016" (Moulik and Ekström,

689 2016), and UCBSEM-WM1 (French and Romanowicz, 2014). Before averaging, all these

models were truncated at spherical harmonic degree 18, and had the global average shear-wave

691 speed at each depth removed. (C) Horizontal cross-section through the same average S-wave

model at 2,800 km depth with profile locations (black lines). Green and white dots mark the left

and right sides of each profile, respectively. For cross-sections through cluster analyses of the

same five tomography models, see Figures 4-5 in *Cottaar and Lekic* (2016).

695

Figure 2: Model setup with initial conditions of (A) composition and (B) temperature. Initial thermal conditions (B) are shown as a vertical profile at the left side of the model domain (x = 0km) that is representative for the whole domain.

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Figure 3: Viscosity and compositional density profiles for the reference case A5LE. (A) 700 Representative viscosity profile is shown as the horizontal average of viscosity in the reference 701 case A5LE after 3 Gyr model time, and generally consistent with post-glacial rebound, geoid and 702 slab-sinking speed constraints (Mitrovica and Forte, 1997; Čižková et al., 2012; Rudolph et al., 703 704 2015). (B) Depth-dependent compositional density profiles for the basaltic (blue) and primordial 705 (dark brown) materials are given, along with depth-dependent thermal expansivity α (red). The specific parameters $\Delta \rho_{BAS,UM}$, $\Delta \rho_{BAS,CMB}$, $\Delta \rho_{PRI,UM}$, and $\Delta \rho_{PRI,CMB}$ for reference case A5LE are 706 marked by black circles. These parameters alone define the density profiles between cases. Note 707 708 that the applied parameterization for basaltic material involves that $\Delta \rho_{BAS} = 2\Delta \rho_{BAS,UM}$ at 300-410 709 km depth and $\Delta \rho_{BAS} = -3 \Delta \rho_{BAS,UM}$ at 660-720 km depth (dashed lines).

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711 Figure 4: Regime map (A) showing the style of thermochemical convection in the lower mantle 712 for each case as a two-dimensional projection of the four-dimensional parameter space. In this 713 projection, parameters $\Delta \rho_{BAS,UM}$, $\Delta \rho_{BAS,CMB}$, $\Delta \rho_{PRI,UM}$, and $\Delta \rho_{PRI,CMB}$, are expressed in terms of the densities of basaltic material at 1,000 km depth ($\Delta \rho_{BAS,1000}$), and of primordial material at 714 2,300 km depth ($\Delta \rho_{PRL,2300}$); see text and Figure 3. Numerical experiments can be classified into 715 six different (major and subsidiary) regimes (colors). Filled circles mark cases that are shown in 716 detail in Figs. 5-7. (B) Percentage of nearly pure primordial material after 3 Gyrs model time as a 717 function of the density difference between primordial and basaltic materials at 2300 km depth. 718 Preferred regimes IIb and IV (see section 4) require density differences of greater than ~100 719 kg/m^3 . 720

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Figure 5: Snapshots of mantle composition predicted by cases (A) X5LH (B) 74KF, (C) 94KF
and (D) X5JB after ~3 Gyrs model time. These cases are representative of regimes I, IIa, IIb and
III, respectively. Primordial and basaltic materials are denoted white and green, respectively
(ambient-mantle pyrolite is blue). All other colors represent mixtures between the materials.
Contours of potential temperature (dashed) are spaced at intervals of 200 K.

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Figure 6: Snapshots of (A) composition and (B) potential temperature for the reference case A5LE (regime IV) after 3 Gyrs model time. For color scale and description of dashed lines in (A), see Figure 5 caption. Dashed lines in (B) roughly delineate compositional domain boundaries contouring mechanical mixtures of 25% primordial with 75% basaltic materials, and 75% basaltic material with 25% pyrolite (see dashed tickmarks in Figure-5-colorscale). For animated versions of panels (A) and (B), see Movies S1 and S2, respectively.

Figure 7: Snapshots of net-density anomaly for the reference case A5LE (regime IV). For an animated version, see Movie S3. Net density anomalies $\Delta \rho = \rho - \rho_0$ are computed from Eq. (1), and the radial average is removed. Positively and negatively buoyant mantle rocks are visualized by warm and cold colors, respectively. For description of dashed lines, see Figure 5 caption.

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Figure 8: Reflection points located in the lower mantle beneath the Pacific. (A) Stars and triangles represent the location of events and stations, respectively. The great circle paths are shown as black lines. Plate boundaries (red) are taken from Bird (2003). Reflection points are displayed by circles and their depth is indicated by color (red to blue for shallow to deep points, respectively). (B) The cross-section shows the reflection points as black circles superimposed onto the shear wave tomography model S40RTS by Ritsema et al. (2011). 745

Figure 9: Summary cartoon. LLSVPs are proposed to be compositionally subdivided into two 746 747 domains, a "primordial" bottom domain and a shallow domain made up of a mix of mostly mafic lithologies (i.e, subducted and accumulated MORB) plus some ultramafic lithologies (tiger 748 stripes). Abrupt seismic gradients, as are found by waveform-modeling studies, are here 749 interpreted to occur across the boundaries between compositional domains, at some of which 750 751 seismic reflections are also observed (see section 4). Seismic reflections from the LLSVP top boundary are not expected to be coherent (dashed line). Blobs qualitatively mark the entrainment 752 of LLSVP materials by plumes. 753

Table 1: key model parameters. The bottom four rows report the free parameters of this study

(bold) that have been systematically explored within the reported range. (^a) For definition of
 depth-dependent parameters, see text and Figure 3b.

Parameter	symbol	value, range
depth of the numerical domain	Z_{box}	2,900 km
width of the numerical domain	x_{box}	8,700 km
surface temperature	T_0	0 °C
mantle reference potential temperature	T_m	2,000 °C
mantle reference density	$ ho_0$	$4,500 \text{ kg/m}^3$
mantle reference viscosity	η_0	$1.4 \cdot 10^{21} \text{ Pa} \cdot \text{s}$
activation volume	V^*	$1.36114 \cdot 10^{-6} \text{ m}^{3}/\text{mol}$
activation energy	<i>E</i> *	47,549.97 J/mol
density anomaly related to depletion of pyrolite	Δho_F	a
excess density of basaltic material	$\Delta \rho_{BAS}$	a
excess density of primordial material	$\Delta \rho_{PRI}$	a
thermal expansivity	α	a
gravity acceleration	g	$9.8 \text{ m}^2/\text{s}$
thermal diffusivity	к	$2.5 \cdot 10^{-6} \text{ m}^2/\text{s}$
upper-mantle excess density of basaltic material	$\Delta \rho_{BAS,UM}$	$70 \text{ kg/m}^3 - 120 \text{ kg/m}^3$
excess density of basaltic material at CMB	$\Delta \rho_{BAS,CMB}$	$40 \text{ kg/m}^3 - 80 \text{ kg/m}^3$
upper-mantle excess density of primordial material	$\Delta \rho_{PRI,UM}$	$160 \text{ kg/m}^3 - 230 \text{ kg/m}^3$
excess density of primordial material at CMB	$\Delta \rho_{PRI,CMB}$	$120 \text{ kg/m}^3 - 200 \text{ kg/m}^3$

757

Table 2: Summary of the results for all 67 cases (reference case A5LE is marked by asterisk). 759 Input parameters are reported in italics [kg/m³]. Important output variables (also used as 760 diagnostic criteria to distinguish between regimes) are given in bold [%]. The "survival rates" of 761 basaltic and primordial materials, surv_{BAS} and surv_{PRI}, are computed as the volumes of mantle 762 material with concentrations of basaltic and primordial material exceeding 80% after 3 Ga model 763 time divided by the initial volumes. The area of the CMB covered by primordial material after 3 764 Ga model time $covrg_{CMB}$ is reported in the last column. $covrg_{CMB}$, $surv_{BAS}$ and $surv_{PRI}$ are used as 765 criteria for discrimination between regimes (see first column). A small subset of cases (^a) have 766 not been run long enough to reach 3 Ga model time, but note that regime discrimination for these 767 cases is unambiguous upon visual inspection of results. 768

regime	case	$\Delta ho_{BAS,660}$	$\Delta \rho_{BAS,CMB}$	$\Delta ho_{PRI,660}$	$\Delta \rho_{PRI,CMB}$	SURV BAS	surv _{PRI}	covrg _{CMB}
Ι	97JH	90	70	200	180	52.0	63.3	74.2
Ι	X5LH	100	50	220	180	67.7	69.5	89.2
Ι	X6KG	100	60	210	170	61.0	65.7	73.3
Ι	A5LG	110	50	220	170	76.0	80.1	80.3
Ι	A6JH	110	60	200	180	59.5	77.8	76
Ι	A6LH	110	60	220	180	68.5	71.0	67.2
Ι	A7KG	110	70	210	170	80.5	69.5	62.5
Ι	A7LG	110	70	220	170	74.2	69.8	80.7
Ι	A8LH	110	80	220	180	62.7	75.4	83.9
Ι	B6KF	120	60	210	160	66.0	63.8	92.6
Ι	B6KH	120	60	210	180	85.2	79.4	60
Ι	B6MH	120	60	230	180	80.1	77.3	65.7
Ι	B8KI	120	80	210	190	85.1	82.6	100
Ι	B8MI	120	80	230	190	82.4	82.1	95.3
IIa	00JG	0	0	200	170	N/A	100	48
IIa	74HD	70	40	180	140	0.1	65.4	45.3
IIa	74JE	70	40	200	150	a	a	a
IIa	74JF	70	40	200	160	0.0	99.1	36.3
IIa	74KF	70	40	210	160	4.1	90.4	44.8
IIa	77HH	70	70	180	180	0.0	88.0	60.6
IIa	94JE	90	40	200	150	0.2	63.8	27.1
IIa/III	87KF	80	70	210	160	0.0	24.7	40
IIa/III	88JF	80	80	200	160	0.5	24.3	31.1
IIb	88JJ	80	80	200	200	15.2	63.6	54.5
IIb	94KF	90	40	210	160	25.6	55.2	45.1
IIb	94LG	90	40	220	170	12.1	88.5	35.2
IIb	94MH	90	40	230	180	36.5	56.3	57
IIb	95JF	90	50	200	160	11.1	50.8	28.5
IIb	95KG	90	50	210	170	18.3	62.5	52
IIb	96KF	90	60	210	160	1.0	60.3	38.6
IIb	97IH	90	70	190	180	12.2	65.8	30.4

IIb	X5JE	100	50	200	150	20.5	50.6	27.2
IIb	X5JG	100	50	200	170	25.3	56.4	38.1
IIb	X5KG	100	50	210	170	44.7	69.0	62
IIb	X5KH	100	50	210	180	53.3	71.5	53.7
IIb	X5LI	100	50	220	190	60.9	67.6	62.7
IIb/III	X4JE	100	40	200	150	23.4	37.4	36.2
IIIa	88FB	90	40	160	120	2.4	7.5	38.8
IIIa	94HB	90	40	180	120	0.4	2.4	19.7
IIIa	94JB	90	40	200	120	2.8	18.8	24.4
IIIa	X4HB	100	40	180	120	a	a	a
IIIa	X5HB	100	50	180	120	a	a	a
IIIa	A5JB	110	50	200	120	12.2	5.8	26.2
IIIa	A6HB	110	60	180	120	a	a	a
IIIa	A6HE	110	60	180	150	38.9	18.3	34.4
IIIa	A6JB	110	60	200	120	15.1	1.5	13.6
IIIb	94HE	90	40	180	150	11.1	20.5	14.6
IIIb	X4HD	100	40	180	140	23.4	22.0	31.9
IIIb	X5IE	100	50	190	150	43.5	45.1	32.2
IIIb	X5JB	100	50	200	120	27.0	30.1	43
IIIb	X5JD	100	50	200	140	36.5	40.0	55
IIIb	A5JE	110	50	200	150	63.8	50.0	34.8
IIIb	A6JE	110	60	200	150	48.8	42.1	48.4
IIIb	A7JF	110	70	200	160	50.8	35.9	42.9
IV	X5HG	100	50	180	170	43.9	53.5	43.9
IV	X5IF	100	50	190	160	50.5	52.2	48
IV	X5KE	100	50	210	150	43.1	63.3	70.6
IV	X6JF	100	60	200	160	54.4	49.3	65.7
IV	X6KF	100	60 70	210	160	53.7	60.1	66
	X/IG	100	/0	190	1/0	47.1	63.8	59.7
	X/JH	100	/0	200	180	72.3	61.3	66.2
	A5JG	110	50	200	1/0	67.8	65.5	61
	*A5LE	110	50	220	150	66.5	61.0	55.9
	ASLF	110	50 60	220	160	65.4	67.5	49
	AOJG	110	0U 60	200	170	04.2	04.U 76 1	55.0 55.0
	A0LG DCL C	110	00	220	170	03.2	/0.1	55.5
11	ROLG	120	00	220	I/U	ð1./	/0.2	57.9

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