

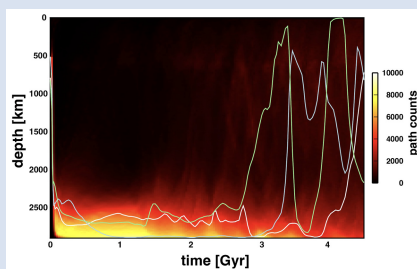
Core-mantle chemical interaction *via* convection within thermochemical piles

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Abstract



Dense thermochemical piles, widely believed to manifest themselves as large low-velocity shear provinces observed above the core-mantle boundary, are gravitationally stable against the ambient mantle, but convection can still happen within the piles. This “inter-pile convection” has two competing effects on core-mantle chemical interaction: one is to reduce the residence time of mantle material at the core-mantle boundary, and the other is to enhance the transfer of core-affected material. Based on convection diagnostics from the numerical simulations of thermochemical piles, we discuss how to constrain the likely formation mechanism of thermochemical piles using mantle geochemistry.

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Introduction

The silicate mantle and the metallic core interact both thermally and chemically at the core-mantle boundary (CMB). Thermal interaction manifests as core heat flux, which has long been studied by numerous authors because of its importance for plume dynamics (e.g., Sleep, 1990), the geodynamo (e.g., Nimmo, 2015), and Earth’s thermal evolution (e.g., Korenaga, 2008). By contrast, less attention has traditionally been paid to chemical interactions, largely because chemical diffusion can be slower than thermal diffusion by many orders of magnitude and other possible transfer mechanisms are not well understood (e.g., Porcelli and Halliday, 2001; Walker and Walker, 2005). However, recent years have witnessed a growing number of observational, experimental, and theoretical studies that consider the possibility of core-mantle chemical interaction to explain the geochemistry of ocean island basalts (OIBs) (e.g., Mundl *et al.*, 2017; Rizo *et al.*, 2019; Yoshino *et al.*, 2020; Olson and Sharp, 2022; Ferrick and Korenaga, 2023b; Deng and Du, 2023; Horton *et al.*, 2023). For example, negative $\mu^{182}\text{W}$ anomalies reported for OIB (e.g., Mundl *et al.*, 2017), especially in conjunction with their negative correlation with $^3\text{He}/^4\text{He}$, are difficult to explain without some interactions with the core, which is considered to have a strongly negative $\mu^{182}\text{W}$ value.

The CMB region is physically and chemically heterogeneous, as indicated by the presence of large low shear-velocity provinces (LLSVPs) and ultra-low velocity zones (ULVZs) (e.g., Garnero *et al.*, 2016). The LLSVPs appear to be the source regions for most large igneous provinces (Burke and Torsvik, 2004), which in turn often serve as the precursors of hot spot magmatism (Richards *et al.*, 1989). Thus, the nature of core-mantle chemical interaction at the LLSVPs becomes important

when considering how possible core signatures can be imparted to the OIB source mantle. Based on their apparent longevity (Torsvik *et al.*, 2014), the LLSVPs have been suggested to represent thermochemical piles compositionally denser than the ambient mantle. Whereas this presumed intrinsic density anomaly can stabilise thermochemical piles against the ambient mantle, convection can still take place within the piles themselves. The purpose of this paper is to investigate the potential role of this convection within a thermochemical pile, or “inter-pile convection”, in core-mantle chemical interaction. ULVZs may contribute to the geochemistry of the OIB source mantle, but given their global distribution and highly variable characteristics (e.g., Hansen *et al.*, 2023), their role in hot spot magmatism is not very clear, and it is beyond the scope of this paper. Core-mantle interaction affects both the ambient mantle and thermochemical piles, but core signatures imparted to the former would be quickly diluted by convective mixing with the bulk mantle. By contrast, dense piles could potentially reside on the CMB for billions of years and acquire strong core signatures, and despite its limited surface coverage on the CMB (~30 %), they could play a disproportionate role in creating chemical heterogeneities in the mantle. Because of inter-pile convection, however, their longevity does not necessarily translate to strong core signals in the geochemistry of OIB source mantle.

Inter-pile convection has two competing effects. Without it, core-mantle chemical interactions would affect only the lowermost portion of the piles, most of which would remain inaccessible. With convection, the base of the piles can be brought directly to the top of the piles, facilitating sampling by mantle plumes. At the same time, convective mixing within piles dilutes the effect of core-mantle chemical interactions. Imparting core signatures on the OIB source mantle, therefore, depends on

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how directly inter-pile convection can bring materials from the bottom to the top and side boundaries, where mixing with the ambient mantle allows the entrainment of dense pile materials. Quantifying the efficiency of such convective transfer is, however, not straightforward because the volume of thermochemical piles can be time dependent; they could have been smaller if they represent accumulated subducted materials (e.g., Christensen and Hofmann, 1994) or larger if they are primordial anomalies (e.g., Korenaga and Marchi, 2023). This fundamentally transient nature of inter-pile convection makes it difficult to build a generic model. In this study, therefore, we opt to present a case study by utilising our earlier numerical model of thermochemical piles (Korenaga and Marchi, 2023). Even though this model is based on a particular hypothesis for pile formation, our approach of characterising the nature of inter-pile convection is sufficiently general, allowing us to discuss core-mantle chemical interaction in the presence of inter-pile convection.

Convection Diagnostics with Dense Tracers

Our convection model was designed to study the fate of a metal-impregnated layer formed in the mid-mantle resulting from a large late accretion impact (Korenaga and Marchi, 2023) (Fig. 1a). The metal-impregnated layer contains $\sim 1 \times 10^{22}$ kg of metal (corresponding to $\sim 1\text{--}2\%$ density anomalies), and it first

sinks to the CMB by the Rayleigh-Taylor instability (Fig. 1b). The layer is then gradually entrained by mantle convection, increasing the abundance of highly siderophile elements (HSE) in the ambient mantle (Fig. 1c–f). Our hypothesis is that the LLSVPs represent the residue of this metal-impregnated layer. The details of our convection modelling are given in the Supplementary Information. Even though this convection model was originally built to test the feasibility of our hypothesis, our modelling results can also be utilised to discuss inter-pile convection in a more general context.

The fate of the metal-impregnated layer is tracked by dense tracers that represent metallic components, and their trajectories contain valuable information on the nature of inter-pile convection (Fig. 1g). As the metal-impregnated layer initially descends to the CMB, most of dense tracers become part of thermochemical piles, and they circulate within the piles before being entrained by the ambient mantle. We focus on the dense tracers that initially sank to the bottom 10% of the mantle and monitor their trajectories up to the time of their entrainment, t_e (defined as the time when a tracer enters the upper half of the mantle). The criterion of the bottom 10% is chosen to easily identify dense particles that constitute long lived piles. For each trajectory, we measure the number of extrema (N_x) of its vertical coordinate as well as the minimum distance to the CMB. The transit time scale, τ_T , or the average period of particle motion within convection, is estimated as $2t_e/(N_x)$ (Fig. 2a).

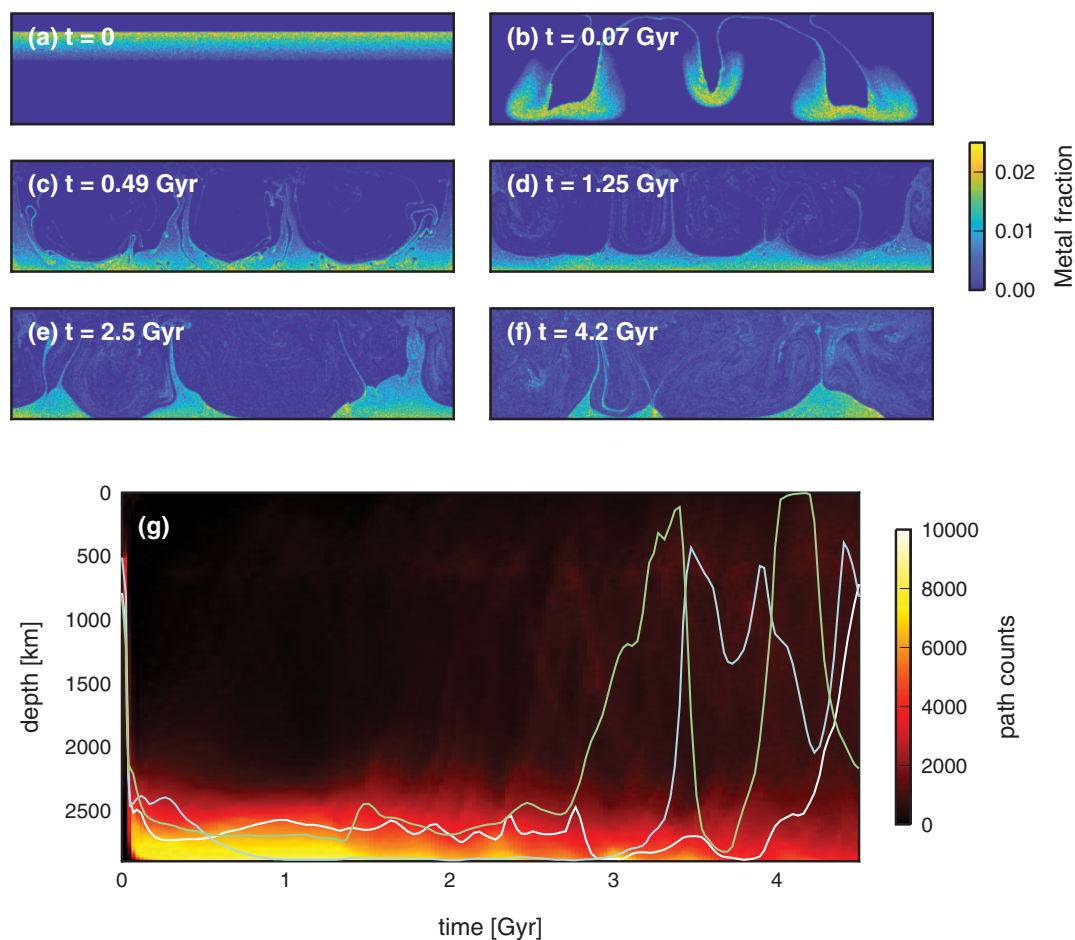


Figure 1 (a–f) Snapshots of the composition field for the case starting with a laterally homogeneous, 750 km thick metal-impregnated layer with the total metal of 10^{22} kg, and with $Ra = 3 \times 10^7$, non-dimensional internal heating of 25, and the Frank-Kamenetskii parameter $\theta = 4.61$. See Figure S-1 for the corresponding temperature field. (g) Summary of vertical trajectories of dense tracers, with three sample trajectories highlighted.

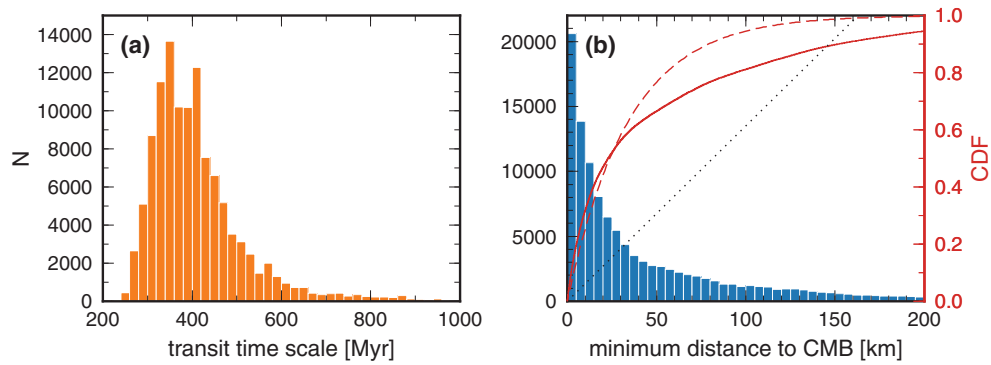


Figure 2 Histograms of (a) transit time scale and (b) minimum distance to CMB, measured from the trajectories of dense tracers for the case shown in Figure 1. Also shown in (b) are the corresponding cumulative distribution function (CDF) in red, a fitted exponential CDF in red-dashed, and a reference uniform CDF in dotted.

To quantify how directly inter-pile convection can bring core signatures to its boundaries, we need to know the fate of mantle parcels that directly touch the CMB, and our inference is based on the statistics of the minimum distance to the CMB for dense tracers (Fig. 2b). The distribution of the minimum distance is usually similar to the exponential distribution. In thermal convection, the delamination of the bottom boundary layer is the source of upwelling, and simply bringing the bottom boundary layer to the top of the pile would result in a relatively uniform distribution of the minimum distance, within the length scale corresponding to the thickness of the boundary layer. We define the amplification factor as a measure of deviations from this expected uniform distribution (see the [Supplementary Information](#) for details). The amplification factor of three, for example, means that entraining the mantle parcels that have directly touched the CMB is three times more likely than expected from the simple upwelling of a bottom boundary layer.

As expected, the transit time scale is basically a function of the Rayleigh number (Fig. 3a), and a higher Rayleigh number (*i.e.* lower viscosity) results in a shorter transit time scale. The transit time scale does not vary much as a function of the entrainment ratio, which measures the fraction of entrained tracers among the dense tracers that initially sank to the bottom 10 % of the mantle. The statistics of the amplification factor are more variable (Fig. 3b). Considerable scatters are seen at low entrainment ratios because of the overpopulation of dense tracers in the piles. In these cases, dense tracers located near the CMB are unlikely to ever be entrained, and thus the distribution of the minimum distance for the entrained tracers does not

necessarily take its maximum at zero distance. For relatively high entrainment ratios (>0.5), the amplification factor is consistently ~ 3 to ~ 4 .

Discussion: Origins of Thermochemical Piles

The characterisation of inter-pile convection allows us to evaluate the strengths and the weaknesses of the existing hypotheses put forward for the origin of thermochemical piles. The diagnostics of inter-pile convection given in the previous section are based on one scenario for pile formation, but the main points of our discussion should remain valid as long as inter-pile convection is concerned. Here we focus on whether a given hypothesis can explain the correlation between the He and W isotopes observed in OIBs (Mundl *et al.*, 2017; Mundl-Petermeier *et al.*, 2020). A recent modelling study suggests that both He and W isotope signatures can be explained by core-mantle chemical interaction (Ferrick and Korenaga, 2023b), but this model treats the OIB source mantle (or equivalently, thermochemical piles) as a static reservoir lying on the CMB. Based on experiments on W diffusivity through lower mantle minerals (Yoshino *et al.*, 2020), the W isotopic signature of the core can diffuse into the bottom ~ 20 km of the mantle over four billion years, but this time scale for chemical interaction is much longer than the transit time scale (Fig. 3a), which is a more appropriate diffusion time scale in the presence of inter-pile convection. With a residence time of only 200 Myr, for example, the diffusion length would be

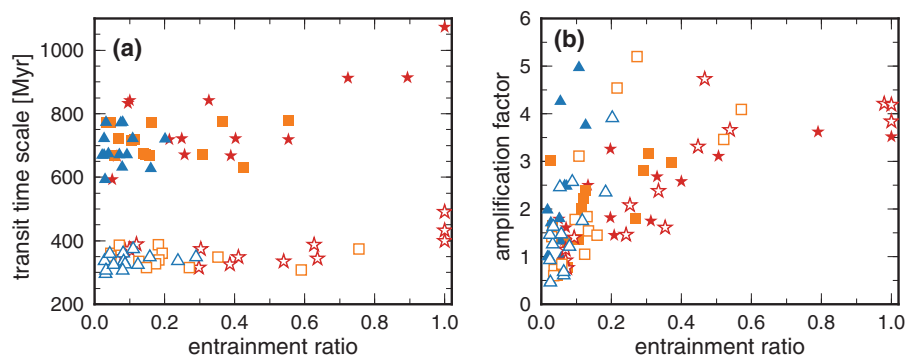


Figure 3 Summary of convection diagnostics for all model runs: (a) transit time scale and (b) amplification factor. Solid and open symbols for $Ra = 10^7$ and 3×10^7 , respectively, and stars, squares, and triangles denote the cases with the Frank-Kamenetskii parameter θ of 4.61, 5.70, and 6.91, respectively.

reduced to ~ 4.5 km. At the same time, inter-pile convection can efficiently deliver this thin core-affected layer to the OIB source mantle, so it is still possible to reproduce the observed isotope correlation by core-mantle interaction. In this case, scatters in the observed correlation are likely caused by the stochastic nature of convective mass transfer, instead of differences in the sampled depth as originally proposed (Ferrick and Korenaga, 2023b).

Inter-pile convection can transport core signatures over a few hundred kilometres, but whether such mass transfer would be quantitatively sufficient still depends on the efficiency of diffusive interaction across the CMB. Consider, for example, the Hawaiian plume, which is the most volumetrically significant plume with its plume buoyancy flux estimated to be $\sim 5 \times 10^3 \text{ kg s}^{-1}$ (Sleep, 1990; King and Adam, 2014). By dividing the buoyancy flux with the product of thermal expansivity ($3 \times 10^{-5} \text{ K}^{-1}$) and a temperature contrast (200 K), we obtain the corresponding mass flux of $\sim 8 \times 10^5 \text{ kg s}^{-1}$. If the average $\mu^{182}\text{W}$ anomaly of Hawaiian basalts is -5 (Willhite *et al.*, 2024) and the core $\mu^{182}\text{W}$ anomaly is -220 , $\sim 2.3\%$ of the source mantle must diffusively interact with the core. To provide such a source mantle for the duration of 20 Myr, for example, the mass of core-affected material needs to be $\sim 10^{11} \text{ kg}$. This translates to the thickness of core-affected layer of $\sim 670 \text{ m}$, with the density of the lowermost mantle (5500 kg m^{-3}) and the radius of the plume stem area of 500 km. For the residence time along the CMB of 200 Myr, this diffusion length scale requires W diffusivity to be greater than $1.7 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$. Higher W diffusivities have been suggested for the CMB conditions ($\sim 8 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$) (Yoshino *et al.*, 2020), but this involves considerable extrapolations from the experimental conditions of 1600–1900 K and 25 GPa to the CMB conditions of $\sim 4000 \text{ K}$ and 130 GPa. Thus, if the actual W diffusivity at the CMB conditions were too low, core-mantle interaction would be insufficient to explain the observed W isotopic signatures. The same consideration applies to the He isotopic signatures, but the situation is different from imparting core W isotopic signatures, which involves only isotope exchange across the CMB with no net W flux. On the other hand, the transfer of core He isotopic signatures depends on the net diffusional flux of He across the CMB, which is proportional not only to the He diffusivity but also to the He concentration in the core, both of which are poorly constrained (Olson and Sharp, 2022).

Thus, W diffusivity at the CMB conditions, along with the transit time scale of inter-pile convection, may be the key to distinguishing different formation hypotheses for thermochemical piles. If the W diffusivity is too low to explain the magnitude of hot spot magmatism with W isotope anomalies, pile formation mechanisms without involving metallic components (*e.g.*, Christensen and Hofmann, 1994; Yuan *et al.*, 2023) would be difficult to explain the observed W isotope signatures. By contrast, W diffusivity becomes irrelevant for pile formation by late accretion (Korenaga and Marchi, 2023) because the core-like W isotope signatures ($\mu^{182}\text{W}$ of -220) are already embedded in the piles from the outset. Note that, contrary to our earlier conjecture (Korenaga and Marchi, 2023), pile formation by late accretion can still satisfy the observed absence of correlation between $\mu^{182}\text{W}$ and HSE concentrations (Mundl *et al.*, 2017). To be entrained and become part of the OIB source mantle, the density anomaly of pile materials must be compensated by the thermal buoyancy of hot ambient mantle, and as a result, pile materials can constitute only $\sim 10\%$ of the OIB source mantle (Ferrick and Korenaga, 2023b). Mixing with the ambient mantle during entrainment thus sufficiently dilutes the effect of metallic components. A major challenge for the late accretion-based hypothesis is instead how to explain the correlation between He and W isotope anomalies. Because

W isotope signatures would be sufficiently strong everywhere within the piles, the magnitude of He isotope signatures need to be similarly ubiquitous, requiring efficient ^3He transport from the core to the mantle. The slopes of the correlations between $\mu^{182}\text{W}$ and $^3\text{He}/^4\text{He}$ vary from plume to plume (or even within a single plume), and some plume-derived systems (*e.g.*, Baffin Bay) are characterised by very high $^3\text{He}/^4\text{He}$ but no W anomaly (*e.g.*, Mundl-Petermeier *et al.*, 2020; Willhite *et al.*, 2024). It is to be seen whether such complexity of observations can be explained by the combination of core-mantle interaction and inter-pile convection.

The threshold diffusivity for W is suggested to be on the order of $10^{-11} \text{ m}^2 \text{ s}^{-1}$ in the above, but there are a few important assumptions, the validity of which requires more careful analyses of hot spot magmatism. First, we need to accurately estimate the spatial and temporal average of a W isotope anomaly for a given mantle plume. If very negative $\mu^{182}\text{W}$ signatures are spatially or temporally localised features, the required volume of core-affected materials could be substantially reduced. Second, plume buoyancy fluxes themselves are subject to non-trivial uncertainties (*e.g.*, King and Adam, 2014). It is also important to consider various types of diffusive interactions. For example, the present day core-side temperature of the CMB is close to the mantle solidus, so the lowermost part of the mantle is likely to have been partially molten when the core was hotter in the past. This tendency is even more important for the piles because they are generally hotter than the ambient mantle. Diffusive transport of core signatures would be rapid through a partially molten mantle. Though it seems difficult to maintain a partially molten state as melt is denser than the coexisting solid phase at the CMB conditions (*e.g.*, Nomura *et al.*, 2011) and likely drains downward, it may be possible to avoid downward percolation by convective stirring (Hernlund and Jellinek, 2010).

Direct mass transfer from the core can also be treated as a diffusive process by defining an 'effective' diffusion coefficient. Whereas the penetration of molten iron into the lower mantle by morphological instability may be unlikely (Yoshino, 2019), MgO exsolution from a cooling core can still happen if the core was initially saturated with Mg (Deng and Du, 2023). This mechanism is important for our discussion because efficient exsolution could result in a high effective W diffusivity, but its efficacy is still uncertain. One important point is that a substantial fraction of an impactor's core would directly merge with the target's core (Marchi *et al.*, 2018), *i.e.* without chemically equilibrating with the target's mantle. Thus, the early core is not necessarily saturated with Mg, and the weak temperature dependence of Mg solubility in the core (Du *et al.*, 2019) further reduces the likelihood of MgO exsolution. MgO exsolution was originally proposed to explain the early geodynamo (O'Rourke and Stevenson, 2016), but it is also possible to explain the early geodynamo without invoking MgO exsolution (Ferrick and Korenaga, 2023a).

Delineating the origin of thermochemical piles using mantle geochemistry will thus require further observational, experimental, and theoretical efforts. It is hoped that our exploration of inter-pile convection will serve as a useful theoretical reference when assimilating the outcomes of future research.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2439>.



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