### **SUPPLEMENTARY INFORMATION**

# **Post-Mesozoic Rapid Increase of Seawater Mg/Ca due to Enhanced Mantle-Seawater Interaction**

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## **SUPPLEMENTARY METHODS**

#### **1. Modeling the global ridge system**

We simulated mantle dynamics beneath the global mid-ocean ridge system in order to estimate the extent to which mantle peridotites can react with seawater in the ocean basins. Peridotite-seawater reactions can take place at temperatures below 500°C, and are responsible for a variety of chemical exchanges (e.g. Mg,  $H_2O$ ,  $CO_2$ ) between solid earth, hydrosphere and biosphere. We divided the global ridge system in 80 sectors each 2048x1024 km, partially overlapping to avoid edge effects. Plate boundaries were digitized from satellite-derived global gravity maps<sup>1</sup> (release 18.1) in Mercator projection at a scale of 1:1000000 (standard parallel:  $0^{\circ}$  N), with the exception of Gakkel Ridge that was digitized from the map of ref. 2 in Lambert Conformal Conic projection (standard parallels: 84°N and 86°N). Assuming a plate spreading velocity for each sector by averaging spreading rates and directions obtained by Euler vectors of ref. 3, we calculated passive mantle flow, thermal structure and melt production beneath each ridge segment (Tab. S1) taking advantage of Comput-ER (Computing in Emilia Romagna), a common-distributed computing infrastructure.

Comput-ER is based mainly on commodity farms and includes g-Lite GRID and cloud/WNoDeS services. It has been set up within a multidisciplinary context in order to support high-CPU demanding and data-driven applications. In fact, a GRID environment offers a large number of storage and computing resources, and advanced high level job submission services. The submission of jobs on different resource centers reduces consistently processing time. The key solution is based on the jobs execution as loose parallel applications, splitting the input data in several parts, and using a single part of input for the elaboration on each node of the GRID. Once all the input data have been processed, it is possible to merge the global output summing up the results of each elaboration. To exploit this possibility we used the parametric job submission feature of the g-Lite WMS in

order to maximize the resource usage and to optimize the management of the computational tasks.

Sub-lithospheric mantle flow and mantle thermal structure were obtained from the solution of Stokes and heat equations by semi-analytical-pseudo-spectral<sup>4</sup> and finite difference techniques<sup>5</sup>, respectively. Depth of isotherms and melt production below a generic ridge-transform-ridge domain are related to (a) spreading rate; (b) length of transform offset and of spreading segments; (c) potential temperature; (d) mantle composition and (e) rheology. The entire sequence of computing programs, requiring as input spreading velocity, plate boundary geometry, mantle potential temperature at the base of the model and mantle source composition, runs on a single computing node (2 Gb RAM, standard OS software) in 24-48 hours and produces 750 Mb of output data. The GRID infrastructure allows the exploration of a wide range of input parameters. Thus, we used a script to submit in a single instance *n-*jobs (one for each mid-ocean ridge sector), achieving *n* solutions within approximately the time of one single run.

#### **1.1 Mantle flow and velocity field**

Fluid-mechanical calculations on mantle flow and melting at spreading centers, lead to models either where mantle upwelling and melting are caused solely by plate separation at ridge axis (passive model), or where a dynamic component of mantle flow is required, driven by thermal and compositional buoyancy. In passive flow models the mantle flow pattern is shaped dominantly by viscous drag from rigid plates moving apart, with melt generated in a broad upwelling region. Mantle flow velocity is affected by plate geometry close to ridge axis, and the upwelling velocity can exceed the half spreading rate when the plate thickens rapidly with distance from ridge axis<sup>5-8</sup>. Dynamic flow models require buoyancy forces and low viscosities in the melting region. In these models mantle upwells close to the spreading axis in a region only a few kilometres wide; upwelling is much faster than the half spreading rate, and melt moves vertically primarily due to its buoyancy<sup>9-11</sup>.

Focusing of upwelling as predicted by "dynamic" flow models is not clearly seen, implying that flow is mostly passive and driven by plate motion<sup>6</sup>. Thus, we attempted to estimate the mantle thermal structure beneath the global ridge system, adopting mantle passive flow models (Figs S1 and S2). We considered a steady-state mantle flow induced by motion of the overlying rigid plates in an incompressible, homogeneous, isoviscous mantle. The steady-state three-dimensional passive mantle flow has been solved via the Fourier pseudo-spectral technique outlined in ref. 4. We modelled plate thickening passive flow beneath an accretionary plate boundary geometry in a computational frame 2048x1024 km wide and 150 km deep (1x1 km spaced grid points for each 1 km depth increment). The base of the lithosphere, assumed to correspond to the depth of the 800 °C isotherm, was obtained iteratively solving the steady-state advection-diffusion heat equation, starting from the thin plates solution.

#### **1.2 Melting Model**

We carried out numerical experiments to estimate mean degree of melting, mean pressure of melting, and mean composition of the aggregate melt beneath a spreading axis. We modelled melt generation, including the effect of water on the peridotite solidus $^{12}$ , using a modified parameterization of experimental data developed by ref. 13, adding a pressuredependent H<sub>2</sub>O bulk distribution coefficient<sup>14</sup>. We varied the H<sub>2</sub>O content of the upper mantle from 0.005 to 0.025 wt%; mantle mineral assemblages for garnet, spinel, and plagioclase peridotite facies are from ref . 15. REE distribution coefficients and source contents are from ref. 16. We assumed that mineral proportions in the transition zone between 85 and 60 km, vary linearly from pure garnet peridotite to pure spinel peridotite. Moreover, we assumed pure-fractional melting and complete melt extraction, but we neglected both latent heat of fusion by freezing of melt, and hydrothermal cooling.

Given that H<sub>2</sub>O is about as incompatible as  $Ce^{17}$ , its concentration in the aggregate liquid is inversely proportional to the mean extent of melting. Thus, the water content of the basaltic glasses for a given degree of melting (i.e.  $Na<sub>8</sub>$ ) constrains the amount of water contained in the mantle source of mid-ocean ridge basalts. The amount of water and  $Na<sub>2</sub>O$ contained in the aggregated melt has been estimated by varying mean degree of melting (spreading rate and/or mantle potential temperature) and mantle source water content. We assumed a water content of 0.02 wt%, because it fits best the value inferred from global basaltic glasses data, obtained from our unpublished results and from the Petrological Database of the Ocean Floor (PETDB, http://www.petdb.org/) for different spreading rates away from hot spots.

Assuming the melting model outlined above, a plate-thickening passive mantle flow and a temperature of  $0^{\circ}$ C at the surface and a constant mantle temperature at 150 km depth (base of our model), ranging from 1300 °C to 1450 °C, we have calculated that the crustal thickness at different spreading rates measured in mid-ocean ridge segments over the global ridge system can be explained best by a mantle potential temperature of 1350 °C at the base model (Fig. S3). The crustal thickness  $H_c$  was calculated at any along-axis locations  $y_0$  of a ridge segment by:

$$
H_c(y_0) = \frac{\rho_m \dot{M}(y_0)}{\rho_c U_0}
$$
\n(S1)

where  $U_0$  is the average sea-floor half-spreading rate of the ridge segment;  $\rho_m$  and  $\rho_c$  are mantle and crustal densities, and  $\dot{M}$  is the total volume of melt production per unit time per unit length of ridge. *M* is computed by integrating the melt production rate *m* at along axis location  $y_0$ , over the cross-sectional area *R* in which melting occurs (i.e., over the region where  $\dot{m}$  is positive):

$$
M(y_0) = \iint_R m(x, y_0, z) dx dz
$$
 (S2).

The melt production rate *m* at any location  $(x, y, z)$  beneath the ridge is given by:

$$
m(x, y, z) = \mathbf{v}(x, y, z) \cdot \nabla F(x, y, z)
$$
 (S3)

where **v** are the mantle flow velocities and *F* is the total amount of melting<sup>12,13</sup>. The mean crustal thickness  $H_c^i$  of the *i-th* ridge segments is obtained by:

$$
\overline{H_c^i} = \frac{\int_0^{L_i} H_c(y_0) dy_0}{L_i} \tag{S4}
$$

where  $L_i$  is the length of the *i-th* ridge segment. The mean global crustal thickness is computed by a weighted average over all the segments of the ridge system:

$$
\overline{H_c} = \frac{\sum_{i=1}^{N} \overline{H}_c^i L_i}{\sum_{i=1}^{N} L_i}
$$
\n(S5)

where *N* is the total number of ridge segments.

Figure S3 shows that our predicted crustal thickness values  $(H_c^i)$  are lower than those inferred from seismic refraction experiments<sup>18-22</sup> at slow- ultraslow-spreading ridges, and higher than those inferred at fast ridges. This is probably due to the inclusion of the serpentinized mantle in the inferred crustal thickness derived from seismic experiments at slow spreading ridges given the similarity of seismic velocities between lower crust and serpentinites. The assumption of complete melt extraction in predicting crustal thickness may be not valid at fast spreading ridges.

#### **1.3 Volume of mantle rocks that can interact yearly with seawater at T < 150 °C**

The amount of cold fluids brought down into the mantle during circulation below ridges depends on the thermal structure of the ridge, which is strongly influenced by rock permeability and spreading rate<sup>23</sup>. Serpentinization itself may affect rock porosity and permeability, causing large volume changes of mantle rocks and therefore rock fracturing.

In our calculations, the thermal structure beneath a segment of the global ridge system depends only on spreading rate and plate boundary geometry; in fact, we neglected latent heat by freezing of melt and hydrothermal cooling. Release of latent heat of fusion by melt freezing rises the isotherms within the crust, while hydrothermal circulation deepens the isotherms. Thus, the two processes, both acting in the shallow portion of the subridge oceanic lithosphere, tend to balance each other. Moreover, latent heat of fusion is low at ultraslow spreading ridges, where melt production is scarce or absent and where mantle peridotite/seawater interactions are likely to occur. Since we are interested in determining the depths of the 150 °C isotherm, where Mg-release by MORP-seawater reactions may occur, we have neglected hydrothermal cooling in order to be conservative. The along-axis depthdistribution of the 150 °C isotherm was obtained by averaging depths from 10 km-wide across-axis sections. The integral of differences between isotherm depth and crustal thickness (where they are positive) times the full spreading rate gives us an estimate of the volume of mantle-derived mid-ocean peridotites (MORP) that can interact yearly with seawater at a temperature below 150°C at each ridge segment. Figure S4 shows two examples of predicted isotherm depths and crustal thickness along segments of the equatorial Mid Atlantic Ridge offset by long offset transform faults. Mantle-derived peridotite can interact with seawater, other than close to long offset transform faults, only at slow-ultraslow spreading ridges (Fig. S5 and Tab. 1).

#### **2. Mg-loss during peridotite-seawater reactions**

We have attempted to measure Mg loss in real rocks as a result of peridotite-seawater reactions (Fig. 1d). We used a set of mantle-peridotites sampled from the Vema Lithospheric Section that exposes lithosphere generated during a 26 Myr time interval at a single Mid Atlantic Ridge segment<sup>24</sup>.

We have considered 29 major element analyses of bulk rock<sup>25</sup> (Tab. S2) obtained by Xray fluorescence at the Department of Earth Sciences of the University of Pisa (Italy). They all show a significant "Loss on Ignition" (LOI), i.e., 11.5 wt%  $\pm$ 1.3, that approximates their H<sub>2</sub>O content. The primary bulk rock major elements composition of 31 samples was reconstructed (Tab. S3) from their primary modal composition and the chemical composition of their primary mineral phases from ref. 26. The rocks are spinel-equilibrated harzburgite to lherzolite. The secondary mineralogy is dominated by serpentine minerals (lizardite  $\pm$  magnetite) that can reach up to 95 % of the rock (Tab. S3). Serpentinization is dominantly pseudomorphic, with minor serpentine veining accommodating some volume expansion during serpentinization. Samples with carbonate veins have been discarded. In order to reconstruct the bulk rock primary composition (major elements) the composition of olivine needs to be assessed. However, due to the high degree of serpentinization primary unaltered olivine is preserved only in few samples. Therefore olivine composition has been estimated based on best correlations with coexisting orthopyroxene and clinopyroxene phases.

We have based our estimates on regression of compositional trends in MORP using available reliable data for slow and ultra-slow spreading ridges<sup>16,26-44</sup>. Natural samples show compositional scatter revealing that exposed MORPs are often affected by a number of postmelting processes. Accordingly, we have discarded data from samples with evidence of late magmatic interaction/impregnation, and we focused on compositional variations in "purely" residual rocks. The resulting data base includes of 130 samples analyzed for the 4 primary phases olivine, opx, cpx, and spinel. Regressions have been made for each oxide pair in order to derive the correlation between major oxides in pyroxenes and in olivine. Spinel composition shows no significant correlation with olivine composition, while Si, Mg, and Fe in both opx and cpx show significant global correlations with olivine. Regression results and correlation values are reported in Fig. S6 and table S4.

MgO in olivine has been estimated based on MgO and  $SiO<sub>2</sub>$  of opx. FeO in olivine was estimated based on  $SiO<sub>2</sub>$  and FeO in opx and  $SiO<sub>2</sub>$  in cpx. Olivine silica content was estimated based on  $SiO<sub>2</sub>$  and FeO in opx, and  $SiO<sub>2</sub>$  in cpx. Existing olivine data from the VLS treated as unknown led to oxide estimates within the 1 sigma error of the measured value. Bulk rock primary compositions have been derived by combining primary modal mineralogy and composition of the primary phases. No volume correction has been applied. Volume to weight modal transformations have a negligible effect on the resulting mode given the expected

density of the analyzed phases (Tab. 3). The specific weight difference of the main phases (olivine, opx, cpx) is too small to significantly affect the overall weight distribution.

Equation (2) of ref. 27 allows to estimate the primary  $MgO<sub>0</sub>$  content from the altered composition (on an anhydrous base) when the primary bulk composition is not available. This can be done inferring the  $MgO/SiO<sub>2</sub>$  ratio from the terrestrial array, and assuming that nothing else (particularly  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$ ) is removed or added to the rock. We calculated Mg loss for the Vema Lithospheric Section (VLS) samples using eqs 2 and 1 of ref. 27. The results (Fig. S7) show that on average 4.1 wt% MgO is extracted from those rocks, in agreement with values inferred from bulk rock reconstructions.

Figure S7 includes also samples of ref. 27 and shows that no correlation exists between estimated Mg loss and crustal age, although most of the samples are from continuous transverse ridges along transform faults. This implies probably early exposition at the seafloor of mantle-derived ultramafics close to the ridge-transform intersection (RTI), and no significant low-T Mg-loss with age.

## **3. Variations in crustal production and volume of mantle rocks that can interact with seawater at at T < 150 °C over the past 150 Ma**

Global cycles, such as sea-level changes<sup>45-46</sup>, the carbon cycle<sup>47</sup>, and seawater chemistry<sup>48</sup>, are assumed to be partly due to temporal variations of the rates of ocean-floor spreading. Based on areal distribution of present-day seafloor age with time since  $180 \text{ Ma}^{49}$ and some tectonic reconstructions<sup>50</sup> it has been suggested that spreading rate could have remained constant over the past 180 Ma. However, recent maps of present day seafloor ages<sup>51,52</sup> and new plate tectonic reconstruction models<sup>52,53</sup> established a curve of seafloor spreading rate and of oceanic crust production rate over the last 150  $Ma^{54,55}$ , suggesting a strong reduction of spreading and crustal production rate since the Santonian , i.e., 83.5 Ma (Fig. S8).

We calculated plate tectonic reconstructions relative to a fixed Africa plate, based on plate boundaries and finite Euler vectors from refs. 52 and 53, at the same Chrons of constructed seafloor spreading isochrons of ref. 53. Reconstructed positions of continents and mid-ocean ridges were obtained with the Gplates software (*http://www.gplates.org*), at

Chrons: 5o (10.9 Ma), 6o (20.1 Ma), 13y (33.1Ma), 18o (40.1Ma), 21o (47.9Ma), 25y (55.9Ma), 31y (67.7 Ma), 34y (83.5 Ma), M0 (120.4 Ma), M10 (131.9Ma), M16 (139.6 Ma), M21 (147.7 Ma) and M25 (154.3 Ma) (Fig. S9). Half spreading rate at each ridge segment mid-point was computed for each time interval, from stage poles and rotation angles for the relative motions of plates that share a specific mid-ocean ridge boundary. Stage rotations describe the motions of plate during two characteristic ages and were calculated combining the finite rotations, expressed as rotation matrices, in the Africa-fixed reference frame. The mean global spreading rate  $V<sub>s</sub>$  was calculated by a weighted average over all the segments of the ridge system:

$$
\overline{V_s} = \frac{\sum_{i=1}^{N} V_s^i L_i}{\sum_{i=1}^{N} L_i}
$$
\n(S6)

where  $V_s^i$  is the spreading rate of the *i-th* ridge segment (Fig. S8a).

Plate boundary geometries and spreading rates of the reconstructed paleo-oceans were used to estimate variations in crustal production and in volume of mantle rocks that can interact with seawater at  $T < 150$  °C over the past 150 Ma, using the same techniques described in previous sections for the current mid ocean ridge system. Results are shown in figures S8b and table S5.

#### **4. Modeling the oceanic mass balance of Mg and Ca through time**

There is strong evidence that the ocean has not been at steady state for Mg concentrations over geological time scales. Several studies have modelled the evolution of seawater composition during Phanerozoic<sup>48,56-60</sup>, or have estimated changes from fluid inclusions<sup>61-64</sup>; from pore fluid chemistry<sup>65</sup>; from marine biogenic carbonates<sup>66-68</sup>; from ridge flank hydrothermal carbonate veins<sup>69</sup>; and from Mg, B and S isotope composition<sup>70-73</sup>. While there is disagreement over the causes of seawater compositional changes, there is a general consensus that the Mg concentration in seawater has varied during Phanerozoic; in particular, that Mg/Ca has risen over the past 80 Ma as a consequence of both an increase of seawater Mg concentration and a decrease of Ca concentration. We attempt next to model the effect on Mg and Ca seawater concentration of (a) variations through time of oceanic crustal production and (b) volume of low-temperature mantle hydration related to changes in spreading rates and accretionary plate boundary geometry. The model calculations are based on the paleoreconstructions described previously. The inferred secular variations of oceanic crust production rate and of volume of low-temperature mantle hydration curves (Tab. S5) were resampled at 1 Ma step after spline interpolation (Fig. 5 and Fig. S8).

#### **4.1 Mg model**

We assume a model where secular variations of seawater Mg-content are controlled by: (1) a constant influx from rivers; (2) variable Mg-release flux from peridotite-seawater reactions; (3) variable Mg-removal flux from high-temperature hydrothermal alteration of the basaltic crust as the result of seafloor spreading rate variations; and (4) a constant Mg residualoutflow including low-temperature off-axis hydrothermal-basaltic crust interactions<sup>74-76</sup>, reaction with carbonate to form dolomite<sup>58-59</sup>, ion-exchange reactions with clays<sup>59</sup>.

Changes in the size of the oceanic Mg reservoir are thus calculated as:

$$
\frac{d\left[Mg\right]}{dt} = F_{rw}^{Mg} + F_{hyP}^{Mg}\left(t\right) - F_{hyB}^{Mg}\left(t\right) - F_{res}^{Mg}
$$
\n(S7)

where:  $F_{rw}^{Mg}$  is the constant river influx, assumed of 5.6·10<sup>12</sup> mol/yr;  $F_{hyP}^{Mg}(t)$  is the influx due to MORP-seawater reactions. We let the MORP-derived Mg flux vary through time following the estimated volume of mantle rocks that can interact yearly with seawater at  $T < 150$  °C (Tab. S5). The estimated recent flux is of  $1.15 \cdot 10^{12}$  mol/yr, assuming that 100% of MORPs that can potentially react with seawater at  $T < 150$  °C do actually react and lose 5% wt of their MgO content (i.e.,  $F_{hyp}^{Mg}(0) = 0.05 \cdot \rho_m \cdot P_m(0) / [MgO_{molar \; mass}]$ , where  $\rho_m = 3300 \text{ kg/m}^3$  is the density of mantle rocks and  $P_m(0) = 2.7769 \cdot 10^8 \text{ m}^3/\text{yr}$  is the current volume rate of MORP that interact with seawater at T<150 °C );  $F_{res}^{Mg}$  is a constant unknown Mg-residual outflow; and  $F_{h_yB}^{Mg}(t)$  is the Mg-removal flux by high-temperature hydrothermal circulation at ridge axis, that varies due to variations in the rate of seafloor production (Tab. S5). The relation between rate of

oceanic crust production and high-temperature hydrothermal flux is likely to be nonlinear, considering the complexity of the physical, chemical, and hydrological processes involved. As a first approximation, we assume that variations through time of Mg removal by the hightemperature hydrothermal depend by seawater Mg content and by an hydrothermal flux that scales linearly with variations in mid ocean ridge crustal rate production:

$$
F_{hyB}^{Mg}(t) = H_{hT}(t)[Mg]_{sw}(t) = H_{hT}(0)[1 + \alpha \frac{P_c(t) - P_c(0)}{P_c(0)}][[Mg]_{sw}(t)]
$$
\n(S8)

where  $[Mg]_{sw}(t)$  is the concentration of Mg in seawater at time *t* and  $H_{hr}(0)$  is the modern mid ocean ridge high-T hydrothermal flux, assumed at  $5·10^{13}$  kg/yr. Assumed values of  $3·10^{13}$  and 5.2 $\cdot$ 10<sup>13</sup> kg/yr (range of the estimated high-T hydrothermal flux) do not change main results.  $P_c(0) = 2.0355 \cdot 10^{10} \text{ m}^3/\text{yr}$  is the current oceanic crustal production rate;  $P_c(t)$  is the production rate through time (Tab. S5); and  $\alpha$  is a scale factor. Figure S10 shows modeled Mg inflow and outflow fluxes since 150 Ma.

#### **4.2 Ca model**

Secular variations of seawater Ca-content have been modelled assuming: a constant river influx  $F_{rw}^{Ca}$  of 1.4·10<sup>13</sup> mol/yr; a variable influx  $\hat{F}_{hyB}^{Ca}(t)$  due to Ca release during low and high temperature hydrothermal circulation at mid ocean ridges; a variable outflux  $F_{wBP}^{Ca}(t)$  related to oceanic seafloor alteration; and an unknown constant residual Ca-outflow  $F_{res}^{Ca}$  including: Cafixation due to carbonate accumulation (biogenic and inorganic), and anhydrite precipitation. Thus, changes through time of seawater Ca-content can be described by:

$$
\frac{d\left[Ca\right]}{dt} = F_{rw}^{Ca} + \hat{F}_{hyB}^{Ca}(t) - F_{wBP}^{Ca}(t) - F_{res}^{Ca}
$$
\n(S9)

with

$$
F_{wBP}^{Ca}(t) = F_{wMORB}^{Ca}(t) + F_{wMORP}^{Ca}(t)
$$
\n(S10)

where  $F_{wMORB}^{Ca}(t)$  and  $F_{wMORP}^{Ca}(t)$  are the outflow relate to alteration at seafloor of mid-oceanridge basalts (MORB) and of exposed mantle rocks, respectively. Secular variations of the net flux due to Ca-release by hydrothermal circulation and Ca-capture by MORB alteration as a result of variations of oceanic crust production (Tab. S5) can be approximated by:

$$
F_{hyB}^{Ca}(t) = \hat{F}_{hyB}^{Ca}(t) - F_{wMORB}^{Ca}(t) = \hat{F}_{hyB}^{Ca}(0)[1 + a\frac{P_c(t) - P_c(0)}{P_c(0)}] - F_{wMORB}^{Ca}(0)[1 + b\frac{P_c(t) - P_c(0)}{P_c(0)}] =
$$
  
=  $F_{hyB}^{Ca}(0)[1 + \beta\frac{P_c(t) - P_c(0)}{P_c(0)}]$  (S11)

where  $\hat{F}^{Ca}_{hyB}(0)$ ,  $F^{Ca}_{wMORB}(0)$ ,  $F^{Ca}_{hyB}(0)$  are the modern mid ocean ridge hydrothermal flux, MORB alteration flux and MORB hydrothermal-weathering net inflow, respectively; and *a, b* and β are scale factors. Estimated modern fluxes of Ca caused by hydrothermal Ca-release  $\hat{F}^{Ca}_{hyB}(0)$ and Ca-fixation during oceanic seafloor alteration  $F_{wBP}^{Ca}(0)$  are  $\lt 4.8 \cdot 10^{12}$  and 1.5-2.4 $\cdot 10^{12}$ mol/yr, respectively<sup>57</sup>. During episodes of amagmatic spreading, along slow-spreading ridges detachments and core complexes unroof the mantle through faulting, allowing exposure of peridotites at the seafloor<sup>77</sup>. These processes may occur along a significant portion of the Atlantic (up to 25%) with peridotites making up as much as 75% of the deeper rocks exposed<sup>78-81</sup>. Several stretches of ultra-slow spreading ridges, characterized by amagmatic accretion, expose mantle peridotites, as well as long-offset transform faults. We can estimate that at least 20% of the current seafloor at mid ocean ridges is floored by peridotites. Thus, the total modern Ca-fixation flux due to alteration of the oceanic seafloor (assumed at  $2.4 \cdot 10^{12}$ mol/yr) may be attributed to:

 $(0) = 80\% \ \ F_{wBP}^{Ca}(0) =$  $F_{wMORB}^{Ca} (0) = 80\% F_{wBP}^{Ca} (0) = 1.9 \cdot 10^{12}$  mol/yr and  $F_{wMORP}^{Ca} (0) = 20\% F_{wBP}^{Ca} (0) =$  $F_{wMORP}^{Ca}(0) = 20\% \ F_{wBP}^{Ca}(0) = 0.5 \cdot 10^{12} \text{ mol/yr}.$ Ophicalcites occur associated with ocean-continent transitions (W-Iberian margin, Alpine Tethys) or slow-spreading ridges and transform zones. They were never observed in the Semail (Oman) and Troodos ophiolites, believed to represent lithosphere produced at intermediate spreading rate (D. Bernoulli, pers. comm.). In ophicalcites hosted by serpentinites, dolomite is very rare and usually of secondary origin. Cements are typically calcite (probably neo-morphic

after aragonite) or aragonite<sup>82</sup>. This suggests that alteration of mantle rocks exposed at the seafloor can capture some Ca rather than Mg. Ca can be extracted from seawater following serpentinization, as observed for instance at the Lost City site  $83$ .

We assume a modern MORB hydrothermal-weathering net inflow  $F_{hyp}^{Ca}(0)$  of 1.25·10<sup>12</sup> mol/yr. Assumed values of  $0.8 \cdot 10^{12}$  and 1.5 $\cdot 10^{12}$  mol/yr (range of the estimated hydrothermal flux) do not change main results. In addition, we assume variations through time of Caremoval by MORP weathering  $F_{wMORP}^{Ca}(t)$  that scales linearly with variations in volume of mantle rocks that can interact yearly with seawater at  $T < 150$  °C (Tab. S5)

$$
F_{\text{wMORP}}^{Ca}(t) = F_{\text{wMORP}}^{Ca}(0)[1 + \gamma \frac{P_m(t) - P_m(0)}{P_m(0)}]
$$
\n(S12)

where  $P_m(0)$  is the current volume rate of MORP that interact with seawater at T<150 °C;  $P_m(t)$ is the volume rate through time (Tab. S5); and  $\gamma$  is a scale factor.

Given the above (S9) can be written as:

$$
\frac{d\left[Ca\right]}{dt} = F_{rw}^{Ca} + F_{hyB}^{Ca}(t) - F_{wMORP}^{Ca}(t) - F_{res}^{Ca}
$$
\n(S13).

Numerical solutions of eqs. (S7) and (S13) were reached by finite difference approximation (Crank–Nicolson implicit scheme) using an integration time step of 1 Ma and initial (150 Ma) Mg- and Ca-seawater concentrations ([Mg] =  $30.5$  mmol/kg H<sub>2</sub>O and [Ca] =  $24$ mmol/kg H<sub>2</sub>O), inferred from halite fluid inclusions<sup>63</sup>. The unknown residual fluxes  $F_{res}^{Mg}$  and  $F_{res}^{Ca}$  were solved iteratively to fit the modern Mg- and Ca-seawater concentrations ([Mg] = 53 mmol/kg  $H_2O$  and  $[Ca] = 10.5$  mmol/kg  $H_2O$ ).

Numerical modelling clearly show that Mg-released by low-temperature MORP-seawater reactions contributed to the sharp increase of seawater Mg-concentration around 70-60 Ma. In addition, Mg-output decreased due to a rapid lowering of oceanic crust production since 83.5 Ma (Santonian) (Fig. 1c). Decreasing Ca-release due to a lower hydrothermal flux after the Santonian decreases seawater Ca-concentration. The Mg/Ca, as a consequence of temporal variations of both Mg and Ca seawater concentrations, stayed close to 1 during the Late Cretaceous and rose up to the current value of  $\sim$ 5.1 during the last 70 Ma, with a rate that increases gradually toward the present day. The maximum rate of Mg/Ca increase recorded at  $\sim$ 20 Ma may account for the rapid change of Mg/Ca during the last 10 Ma inferred from pore fluid chemistry<sup>65</sup>.

#### **4.3 Constraints from Mg isotope ratios**

The Mg isotope budget of the oceans may provide constraints on the relative contribution of the two major sinks for Mg: hydrothermal circulation in basalt, and dolomite deposition. Following ref. 71, we considered the Mg isotope budget of the oceans through time, including the effect of Mg-released by MORP/seawater reactions, assuming the Mg-fluxes estimated by our model. Considering first the residual Mg-outflow  $F_{res}^{Mg}$  of our model, we assume here that it is due only to off-axis low-temperature hydrothermal circulation within basalts  $F_{h\nu O}^{Mg}$  and dolomite accumulation  $F_{dd}^{Mg}$  :

$$
F_{res}^{Mg} = F_{hyO}^{Mg} + F_{dol}^{Mg} \implies F_{hyO}^{Mg} = a(t)F_{res}^{Mg}; F_{dol}^{Mg} = b(t)F_{res}^{Mg}
$$
 (S14)

where *a* and *b*, are fractions of the residual flux due to off-axis hydrothermal circulation and dolomite deposition, respectively.  $a(t) + b(t) = 1 \ \forall t \in \mathfrak{R}^+$ .

The rate of change through time of seawater  $^{26}Mg/^{24}Mg$  in the oceans may be derived from the conservation equations for the individual isotopes:

$$
\frac{d\delta_{\text{sw}}^{\text{26}}(t)}{dt} = \begin{cases} F_{\text{rw}}^{\text{Mg}} [\delta_{\text{rw}}^{\text{26}} - \delta_{\text{sw}}^{\text{26}}(t)] + F_{\text{hyp}}^{\text{Mg}} (t) [\delta_{\text{hyp}}^{\text{26}} - \delta_{\text{sw}}^{\text{26}}(t)] - \\ - F_{\text{hyp}}^{\text{Mg}} (t) [\delta_{\text{hyp}}^{\text{26}} - \delta_{\text{sw}}^{\text{26}}(t)] - \\ - F_{\text{res}}^{\text{Mg}} [a(t) [\delta_{\text{hyp}}^{\text{26}} - \delta_{\text{sw}}^{\text{26}}(t)] + b(t) [\delta_{\text{dol}}^{\text{26}} - \delta_{\text{sw}}^{\text{26}}(t)] \end{cases} / N_{\text{Mg}}(t) \tag{S15}
$$

where  $\delta_{\scriptscriptstyle SW}^{\scriptscriptstyle {^{26}}Mg}$  ,  $\delta_{\scriptscriptstyle rw}^{\scriptscriptstyle {^{26}}Mg}$  $\delta_{\scriptscriptstyle \mathit{rw}}^{\scriptscriptstyle 2^6Mg}$  ,  $\delta_{\scriptscriptstyle hyp}^{\scriptscriptstyle 2^6Mg}$  $\delta^{\scriptscriptstyle{{2^6}Mg}}_{\scriptscriptstyle{hyP}}$  ,  $\delta^{\scriptscriptstyle{{2^6}Mg}}_{\scriptscriptstyle{hyB}}$  $\delta^{\scriptscriptstyle{{2^6}Mg}}_{\scriptscriptstyle{hyB}}$  ,  $\delta^{\scriptscriptstyle{{2^6}Mg}}_{\scriptscriptstyle{hyO}}$  $\delta_{\text{hyO}}^{^{26}Mg}$  and  $\delta_{\text{dol}}^{^{26}Mg}$  are the  $\delta^{26}Mg$  of seawater, of river waters, of MORP/seawater reactions related fluids, of high-temperature hydrothermal fluids, of lowtemperature off-axis hydrothermal fluids and of dolomite, respectively. *NMg is* the total number of moles of Mg in the ocean. We assume that current best estimated values for  $\delta^{26}Mg$  of rivers (-1.09), for MORP (-0.36) and dolomite (-2.0) did not change through time. We assume also no Mg isotopic fractionation during MORP/seawater reactions. In addition, we assume that no Mg is returned to the ocean from both high- and low-temperature hydrothermal circulation within basalts. This last assumption implies that  $\delta_{hyB}^{\alpha\beta} = \delta_{hyO}^{\alpha\beta}$  $\boldsymbol{\delta}_{h\mathsf{y}O}^{\mathsf{^{26}Mg}}$   $=$   $\boldsymbol{\delta}_{\mathsf{^{SW}}}^{\mathsf{^{26}Mg}}$  $\delta_{\text{sw}}^{^{26}Mg}$ , and eq. (S13) reduces:

$$
\frac{d\delta_{\rm sw}^{\rm 26\,Mg}(t)}{dt} = \begin{cases} F_{\rm rw}^{\rm Mg} \left[ \delta_{\rm rw}^{\rm 26\,Mg} - \delta_{\rm sw}^{\rm 26\,Mg}(t) \right] + F_{\rm hyp}^{\rm Mg}(t) \left[ \delta_{\rm hyp}^{\rm 26\,Mg} - \delta_{\rm sw}^{\rm 26\,Mg}(t) \right] - \left[ \left[ \delta_{\rm hyp}^{\rm 26\,Mg} - \delta_{\rm sw}^{\rm 26\,Mg}(t) \right] \right] \end{cases} / N_{\rm Mg}(t) \tag{S16}
$$

We explore next three possible scenarios:

- (i) The ocean is at steady state with respect to Mg isotopic composition;
- (ii) Non steady state scenario for  $\delta^{26}Mg$  of seawater and constant accumulation of dolomite;
- (iii) Non steady state scenario for  $\delta^{26}Mg$  of seawater as well as for dolomite.

(i) Assuming that no change of seawater  $\delta^{26}Mg$  occurred through time, eq. (S16) allows to infer variations of dolomite deposition:

$$
b(t) = \frac{\left\{F_{rw}^{Mg} \left(\delta_{rw}^{^{26}Mg} - \delta_{sw}^{^{26}Mg}\right) + F_{hyP}^{Mg} \left(t\right) \left(\delta_{hyP}^{^{26}Mg} - \delta_{sw}^{^{26}Mg}\right)\right\}}{F_{res}^{Mg} \left(\delta_{dol}^{^{26}Mg} - \delta_{sw}^{^{26}Mg}\right)}
$$
(S17)

where  $\delta_{sw}^{^{26}Mg}$  =-0.82 (ref. 71).

Figure S11a shows modelled variations of Mg-removal flux due to dolomite deposition since 150 Ma. Dolomite Mg-removal flow ranges from ~22% of the river input (implying  $F_{h\nu0}^{Mg}$  $=$  ~32% of the river input) during the Cretaceuos, to 15 % (implying  $F_{hyO}^{Mg} = \sim 39\%$  of the river input) during the Cenozoic (volume fraction of MORP interacting at low-T with seawater = 100%). Note that when the effect of Mg-released by MORP/seawater reactions is added (volume of MORP interacting at lowT with seawater  $> 0$ ), modelled fluxes and the percentage of dolomite in carbonate sediments of ref. 84 correlate.

(ii) We let seawater  $\delta^{26}Mg$  vary through time. If we assume that dolomite deposition has been constant through time, the fraction *b* of  $F_{res}^{Mg}$  can be solved iteratively from eq. (S16) by fitting the modern seawater  $\delta^{26}Mg$  value (-0.82). Dolomite Mg-removal fluxes range from 15.7% of the river input (volume fraction of MORP interacting at low-T with seawater  $= 100\%$ ) to 23% of the river input (volume fraction of MORP interacting at low-T with seawater  $= 0\%$ ), regardless of the initial value of 150 Ma seawater  $\delta^{26}Mg$  (Fig. S11b).

(iii) We let the  $\delta^{26}Mg$  of seawater and dolomite deposition vary through time. In this case  $b(t)$ is undetermined and additional constraints are needed. Several lines of evidence show that dolomite deposition prevailed during the Cretaceous, in contrast with the prevailing carbonate deposition that occurred during the Cenozoic. It has been also suggested that as much as 90% of seawater Mg is lost to a dolomite sink during enhanced periods of dolomitization<sup>56</sup>. Such a large dolomite sink would induce a huge shift in the  $\delta^{26}Mg$  of seawater toward values heavier (Fig. S11c) than those of modern seawater  $\delta^{26}Mg$  (-0.82), in contrast with data from echinoderms<sup>85</sup> suggesting that seawater  $\delta^{26}Mg$  stayed approximately constant and close to the modern value during the last 350 Ma, except for a negative excursion occurring at around 100 Ma. We conclude that the Mg isotopic chemistry is consistent (compatible) with a lowered dolomite deposition and increased MORP-seawater interaction in the Cenozoic and modern oceans.

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**Figure S1.** *Mantle thermal structure beneath a ridge-transform-ridge plate boundary and transform offset of 100 km. Depth of 150 °C (a, b) and 500 °C (e, f) isotherms for spreading rate of 7.5 and 30.0 mm/yr, respectively. c, d, g, h, across axis sections far away from the ridge transform intersection. Red dashed line: mean isotherm depth obtained by averaging depths from the across-axis 10 km-wide region (gray shaded area), where we assume the occurrence of hydrothermal seawater circulation.*



**Figure S2.** *Mantle thermal structure beneath a ridge-transform-ridge plate boundary and transform offset of 500 km. See caption of figure S1.*



**Figure S3.** *Oceanic crustal thickness versus spreading rate. Diamonds indicate mean crustal thickness predicted at each mid-ocean ridge segment. Circles, squares and triangles indicate crustal thickness inferred from seismic refraction experiments*18-22 *.*



**Figure S4.** *Crustal thickness and depth from seafloor of the 150 °C isotherm predicted at two Mid Atlantic Ridge segments dislocated by long offset transforms (Chain, Romanche and St. Paul). Shaded areas indicate the mantle region that can contribute to <150 °C MORP-seawater reactions and consequently to Mg release, i.e., the region where the difference between isotherm depth and crustal thickness is positive.*



**Figure S5.** *Volume of mantle rocks that may react with seawater at T <150 °C along present day mid-ocean ridges.* 



**Figure S6.** *Linear regression (green solid line) of olivine major oxides (SiO2, MgO and FeO) versus oxides of opx and cpx. 95% confidence intervals are also indicated (gray solid lines).*



**Figure S6.** *Mg-loss (*∆*MgO) versus crustal age. Diamonds refer to SWIR peridotites<sup>27</sup>, squares to Vema Lithosperic Section peridotites24-26. Both populations show Mg-loss in serpentinites compared to unaltered primary rocks.* 



**Figure S8.** *Spreading rate (a) and oceanic crustal production (b) temporal variations since 150 Ma.*



Figure S9. Global plate reconstructions relative to Africa-fixed reference frame from Late Oxfordian (154 Ma) to the present day. Positions of continents and mid-ocean ridges, obtained with Gplates software (http://www.gplates.org), are plotted in Mollweide projection using PLOT-MAP package<sup>86</sup>. Preserved mid-ocean ridges (double red lines) are from ref. 52, and reconstructed mid-ocean ridges (double blue lines) are from ref. 53. Half spreading rates are calculated by stage poles obtained from the finite Euler poles and rotation angles of ref. 53. Magnetic chron ages ( $o=lower$  and  $y=upper$  boundary) are from ref. 87 (Chrons 0-34) and ref. 88 (Chrons M25-M0).





**Figure S9 continues.** 



**Figure S9 continues.**



**Figure S10.** *Modeled seawater Mg fluxes since 150 Ma. a, constant river inflow and temporal variations of Mg-release flux due to low-T MORP-seawater interactions for different volumes of mantle rocks that can interact yearly with seawater at T < 150 °C. b, calculated residual removal flux and temporal changes of Mg-removal flux by high-T hydrothermal circulation in basalts for different volumes of mantle rocks that can interact yearly with seawater at T < 150 °C.* 



**Figure S11.** *Mg-removal constraints from Mg isotope ratio. a, temporal variations of Mg-removal flux due to dolomite deposition inferred at different volumes of mantle that can interact yearly with seawater at T < 150 °C, assuming steady state seawater Mg isotopic composition. b, temporal changes of seawater* δ *<sup>26</sup>Mg due to Mg-release by MORP-seawater reactions for different volumes of mantle that can interact yearly with seawater at T < 150 °C and for different initial conditions, assuming a constant flux of dolomite Mgremoval, obtained iteratively to fit the modern ocean*  $\delta^6Mg$  *of seawater. c, variations of seawater*  $\delta^6Mg$ *, assuming no Mg contribution from MORP-seawater reactions and assuming a greater Mg-removal flux due to dolomite deposition during the Cretaceous than that during the Cenozoic.* 

**Table S1.** *Volume (m<sup>3</sup>/yr) of mantle rocks (Vol*<sub> $<150°C$ </sub>) that can interact yearly with seawater at T  $<$ *150 °C estimated at each segment of the global mid-ocean ridge system. Latitude (Lat), Longitude (Long), half spreading rate (Vs) and spreading direction (as) are referred to the mid-point of each ridge segment (#). Spreading rates (mm/yr) and directions (azimuth, degree) were obtained using MORVEL angular velocities (ref. 3).*  $L_{seg}$ ,  $L_{off}$ <sup>1</sup> and  $L_{off}$ <sup>2</sup> are the lengths (km) of the ridge segment *and of the two bounding transform offsets. Hc is the mean crustal thickness (km) at ridge segment. Plate name abbreviations are as follows: AN, Antarctic; AR, Arabia; AU, Australia; CO, Cocos; CP, Capricorn; EU, Eurasia; IN, India; JF, Juan de Fuca; LW, Lwandle; NA, North America; NB, Nubia; NZ, Nazca; PA, Pacific; SA, South America; SM, Somalia and SR, Sur. Mgrel is the amount of Mg that can be released yearly to seawater from hydration of the oceanic mantle.* 

<b>Plate</b>	<b>Ridge</b>	#	Lat	Long	$\boldsymbol{V_s}$	$\alpha_{\rm s}$	$L_{\frac{seq}{}}$	1 off	$\boldsymbol{2}$ $L_{\text{off}}$	$H_c$	Vol <150℃
SM-AN	<b>SWIR</b>	$\mathbf{1}$	$-26.56300$	67.81948	6.30	354.67	423.48	0.00	29.67	1.56	7588975
SM-AN	<b>SWIR</b>	$\overline{c}$	$-27.53997$	65.82392	6.44	355.02	97.36	29.67	30.66	1.53	2057335
SM-AN	<b>SWIR</b>	$\mathbf{3}$	$-27.97186$	63.90687	6.57	355.72	253.20	30.66	28.04	1.67	4379756
SM-AN	<b>SWIR</b>	$\overline{4}$	$-28.40468$	62.48292	6.66	356.19	65.05	28.04	45.89	1.47	1620009
SM-AN	<b>SWIR</b>	$\mathbf 5$	$-28.81275$	61.80463	6.70	356.29	65.39	45.89	21.63	1.50	1554555
SM-AN	<b>SWIR</b>	6	-28.99793	61.10407	6.74	356.55	64.16	21.63	137.44	1.22	2200814
SM-AN	<b>SWIR</b>	$\overline{7}$	$-30.66291$	59.64672	6.84	356.40	254.88	137.44	50.27	1.50	5610136
SM-AN	<b>SWIR</b>	8	$-31.70996$	57.68906	6.94	356.92	131.11	50.27	203.61	1.12	4272373
SM-AN	<b>SWIR</b>	9	$-33.70055$	56.39605	7.02	356.68	131.24	203.61	66.66	1.08	4417996
SM-AN	<b>SWIR</b>	10	$-34.36110$	55.24813	7.07	357.05	60.32	66.66	44.39	1.31	1866287
SM-AN	<b>SWIR</b>	11	$-34.74418$	54.48724	7.11	357.32	66.74	44.39	62.28	1.35	1925342
SM-AN	<b>SWIR</b>	12	$-35.34749$	53.87608	7.14	357.42	44.61	62.28	0.00	1.42	1233203
LW-AN	<b>SWIR</b>	1	$-35.44980$	53.52162	7.37	0.70	21.06	0.00	76.02	1.38	762988
LW-AN	<b>SWIR</b>	$\overline{c}$	$-36.12830$	52.85346	7.36	1.13	92.58	76.02	111.63	1.13	3110012
LW-AN	<b>SWIR</b>	3	-38.00976	49.14486	7.33	3.56	576.51	111.63	141.94	1.94	7795241
LW-AN	<b>SWIR</b>	4	$-40.30028$	45.34907	7.29	6.32	138.86	141.94	40.96	1.47	3436059
LW-AN	<b>SWIR</b>	$\mathbf 5$	-40.98222	43.60392	7.26	7.61	143.34	40.96	217.88	1.32	4078443
LW-AN	<b>SWIR</b>	6	-42.79260	42.11284	7.25	8.99	47.69	217.88	130.99	0.57	2832534
LW-AN	<b>SWIR</b>	$\overline{7}$	-43.89076	40.90934	7.23	10.08	91.79	130.99	47.86	1.19	2912442
LW-AN	<b>SWIR</b>	8	$-43.39413$	39.84900	7.21	10.74	84.44	47.86	101.01	1.23	2648334
LW-AN	<b>SWIR</b>	9	-44.52763	37.40200	7.16	12.80	324.31	101.01	175.19	1.62	6629348
LW-AN	<b>SWIR</b>	10	$-46.10050$	34.31334	7.11	15.57	71.35	175.19	147.81	0.66	3551620
LW-AN	<b>SWIR</b>	11	$-47.20813$	32.93099	7.09	16.97	76.67	147.81	105.44	0.80	3362003
LW-AN	<b>SWIR</b>	12	-48.04974	31.81229	7.07	18.12	23.48	105.44	133.23	0.73	1462203
LW-AN	<b>SWIR</b>	13	-49.13129	30.54540	7.05	19.48	21.04	133.23	0.00	0.94	1128966
NB-AN	<b>SWIR</b>	$\mathbf{1}$	-49.18814	30.35341	8.20	15.76	10.22	0.00	456.57	0.84	1142165
NB-AN	<b>SWIR</b>	$\boldsymbol{2}$	-52.67615	26.74986	8.14	20.18	137.95	456.57	137.41	0.97	5787635
NB-AN	<b>SWIR</b>	3	$-52.76229$	18.76308	7.87	27.50	1012.67	137.41	31.24	2.43	3958578
NB-AN	<b>SWIR</b>	$\overline{4}$	-52.94640	10.13585	7.55	36.00	32.17	31.24	199.15	1.33	1491546
NB-AN	<b>SWIR</b>	5	-54.10965	7.81689	7.52	38.95	32.01	199.15	23.08	1.39	1431316
NB-AN	<b>SWIR</b>	6	-53.98690	7.01534	7.48	39.72	60.34	23.08	119.37	1.52	1753867
NB-AN	<b>SWIR</b>	7	$-54.45521$	5.12765	7.44	41.94	57.80	119.37	22.70	1.47	1694503
NB-AN	<b>SWIR</b>	8	-54.05075	3.84523	7.37	43.11	120.48	22.70	222.66	1.43	3359277
NB-AN	<b>SWIR</b>	9	-54.89757	0.30274	7.30	47.37	57.59	222.66	28.64	1.20	2271104
NB-AN	<b>SWIR</b>	10	-54.87546	$-0.55455$	7.27	48.31	28.47	28.64	0.00	2.22	408448

**South West Indian Ridge (SWIR)** 

 $SWIR - Vol_{< 150^{\circ}C} = 105.735 \cdot 10^6 \text{ m}^3/\text{yr} - \text{Mg}_{rel} = 4.368 \cdot 10^{11} \text{ mol/yr}$ 

 **Vol**MORB **= 118.697·10<sup>6</sup> m<sup>3</sup> /yr** 

## **Mid Atlantic Ridge (MAR)**





 $\text{MAR} - \text{Vol}_{< 150^\circ \text{C}} = 62.317 \cdot 10^6 \text{ m}^3/\text{yr} - \text{Mg}_{rel} = 2.574 \cdot 10^{11} \text{ mol/yr}$ 

$$
Vol_{MORB} = 1971.477.10^6 \text{ m}^3/\text{yr}
$$

#### **Gakkel Ridge**



 $\bf{Vol}_{\rm MORB} = 26.674 {\cdot} 10^6 \ {\rm m}^3/{\rm yr}$ 

## **America-Antarctica Ridge (AAR)**



**AAR – Vol<150°C = 16.536 10<sup>6</sup> m<sup>3</sup> /yr – Mgrel = 0.683 10<sup>11</sup> mol/yr**   $\text{Vol}_{\text{MORB}} = 18.825 \cdot 10^6 \text{ m}^3/\text{yr}$ 

## **Central Indian Ridge (CIR)**





 $CIR - Vol_{<150°C}$  = 12.734 10<sup>6</sup> m<sup>3</sup>/yr – Mg<sub>rel</sub> = 0.526 10<sup>11</sup> mol/yr

 **Vol**MORB **= 878.091·10<sup>6</sup> m<sup>3</sup> /yr** 

# **Pacific Ridge (PAC)**





 $PAC - Vol_{< 150^\circ \text{C}} = 10.520 \cdot 10^6 \text{ m}^3/\text{yr}} - \text{Mg}_{rel} = 0.435 \cdot 10^{11} \text{ mol/yr}$ 

 $\boldsymbol{Vol}_{\text{MORB}} = 11589.773 \cdot 10^6 \text{ m}^3/\text{yr}$ 

## **Nazca Ridge**



 $N$ azca – Vol<sub><150°C</sub> = 6.657 10<sup>6</sup> m<sup>3</sup>/yr – Mg<sub>rel</sub> = 0.275 10<sup>11</sup> mol/yr  **Vol**MORB **= 1020.456·10<sup>6</sup> m<sup>3</sup> /yr** 

## **South East Indian Ridge (SEIR)**







 $\text{SEIR} - \text{Vol}_{< 150^\circ \text{C}} = 4.125 \cdot 10^6 \text{ m}^3/\text{yr} - \text{Mg}_{rel} = 0.170 \cdot 10^{11} \text{ mol/yr}$ 

 **Vol**MORB **= 3938.797·10<sup>6</sup> m<sup>3</sup> /yr** 

## **Galapagos Ridge**



Galapagos – Vol<sub><150</sub> $c = 0.0$  m<sup>3</sup>/yr – Mg<sub>rel</sub> = 0.0 mol/yr  $\bf{Vol}_{\rm MORB} = 791.925 {\cdot} 10^6 \ {\sf m}^3/{\sf yr}$ 

<b>Sample</b>	<b>SiO2</b>	TiO <sub>2</sub>	Al203	<b>FeOtot</b>	<b>MnO</b>	MgO	Ca <sub>O</sub>	<b>Na20</b>	<b>K20</b>	P <sub>205</sub>	LOI	Sum
S19-02/21	39.61	0.07	1.23	7.5866	0.1	35.83	1.38	0.38	0.02	0.04	13.1	99.32
S19-02/22	40.06	0.06	2.82	8.3215	0.12	33.49	2.17	0.66	0.03	0.06	11.5	99.25
S19-02/26	37.8	0.06	0.82	10.247	0.12	35.39	0.58	0.68	0.03	0.09	13.2	99.03
S19-03/25	40.40	0.06	1.19	6.95	0.08	37.08	0.95	0.39	0.03	0.04	12.2	99.40
S19-04/64	39.6	0.03	1.31	7.6315	0.25	37.22	0.48	0.28	0.04	0.06	12.4	99.25
S19-07/23	40.42	0.12	2.68	7.04	0.19	35.87	1.36	0.30	0.04	0.05	11.3	99.39
S19-07/30	39.34	0.01	0.91	8.47	0.12	36.29	0.61	0.47	0.04	0.05	12.9	99.21
S19-12/04	39.73	0.07	2.44	12.04	0.22	32.26	3.57	0.54	0.04	0.15	7.8	98.86
S19-12/33	36.64	0.08	2.49	15.86	0.40	30.75	2.85	0.81	0.07	0.24	8.2	98.43
S19-13/36	37.13	0.04	1.20	8.18	0.10	35.42	3.51	0.37	0.02	0.05	13.2	99.23
S19-13/37	44.92	0.04	1.00	10.19	0.14	30.29	2.49	0.66	0.10	0.05	9.2	99.08
S19-15/17	39.67	0.04	2.07	9.78	0.18	33.79	1.32	0.51	0.06	0.11	11.5	99.05
S19-15/70	38.90	0.04	1.67	10.98	0.17	34.42	1.65	0.51	0.04	0.13	10.4	98.93
S19-20/85	37.98	0.10	3.00	12.18	0.35	30.54	3.02	0.55	0.05	0.15	10.9	98.79
S19-05/55	40.59	0.01	1.14	8.14	0.16	36.51	0.47	0.34	0.03	0.03	11.9	99.28
S19-05/64	41.19	0.02	1.25	6.26	0.12	38.35	0.43	0.25	0.02	0.01	11.6	99.52
S19-05/84	40.69	0.02	0.68	8.50	0.15	37.04	0.67	0.35	0.03	0.04	11.1	99.25
S19-23/39	40.48	0.02	1.36	5.95	0.09	38.81	0.16	0.21	0.02	0.02	12.4	99.54
S19-23/41	39.97	0.02	1.16	6.22	0.10	38.98	0.21	0.19	0.01	0.02	12.6	99.50
S19-23/45	40.01	0.02	1.24	8.09	0.11	37.56	0.78	0.32	0.03	0.06	11.1	99.33
S19 23/46	40.20	0.03	1.59	7.88	0.16	37.38	0.66	0.41	0.06	0.05	10.9	99.35
S19-23/49	40.51	0.03	1.22	6.93	0.11	38.05	0.23	0.45	0.05	0.03	11.8	99.41
S19-24/19	40.17	0.04	1.32	8.29	0.15	36.05	1.46	0.34	0.02	0.05	11.4	99.25
S19-25/71	40.49	0.01	0.52	8.18	0.08	36.57	0.85	0.32	0.03	0.03	12.2	99.27
S19-27/01	40.23	0.03	1.58	6.41	0.18	38.49	0.62	0.26	0.04	0.03	11.6	99.46
S19-28/23	40.32	0.02	1.39	7.39	0.24	37.04	0.97	0.29	0.03	0.04	11.6	99.31
S19-30/30	39.10	0.01	0.71	7.30	0.20	39.30	0.06	0.23	0.02	0.03	12.3	99.29
S19-30/72	39.59	0.04	1.39	6.25	0.36	38.77	0.30	0.27	0.03	0.03	12.5	99.50
S19-30/79	40.80	0.02	1.76	10.03	0.16	33.22	1.65	0.30	0.03	0.11	11.2	99.29

**Table S2.** *Bulk rock composition of samples from the Vema Lithospheric Section<sup>25</sup>. Data show a variety of serpentinized rocks, texturally ranging from porphyroclastic to mylonitic.* 

**Table S3.** *Reconstructed primary bulk rock major elements of samples from the Vema Lithospheric Section<sup>26</sup> .* 

		reconstr. bulk rock			rock mode weight		rock mode volume			estimated olivine lopx						<b>CDX</b>			sn					
sample	sum	<b>Sitot</b>	<b>Matot</b>	Fetot	ol	ODX	cpx	sp	ol	ODX	cpx	sp	Maol	Feol	Si	<b>SiO2</b>	Fe0	<b>MaO</b>	<b>SiO2</b>	Fe0	MaO	<b>SiO2</b>	Fe <sub>0</sub>	Ma <sub>0</sub>
S1902-05	97.80	43.48	45.83	8.49	0.780	0.190	0.023	0.009	0.778	0.190	0.023	0.009	50.04	9.18	40.78	55.14	6.05	33.11	51.45	2.58	16.46	0.02	13.45	17.50
S1902-07	96.48	44.66	43.71	8.11	0.678	0.280	0.032	0.013	0.675	0.279	0.032	0.013	49.96	9.20	40.85	54.74	5.84	$32.7^{\circ}$	51.49	2.17	15.73	0.02	13.27	17.32
S1904-38A	95.98	43.44	44.04	8.50	0.711	0.251	0.015	$0.02^{\circ}$	0.709	0.251	0.015	$0.02^{n}$	49.81	9.37	40.83	54.30	6.07	32.25	51.13	2.45	16.32	0.04	13.03	17.40
S1904-39	97.65	45.63	43.86	8.16	0.653	0.317	0.032	0.005	0.647	0.315	0.032	0.005	50.02	9.28	40.70	54.90	6.18	33.44	51.10	2.39	16.14	0.02	13.45	18.10
S1904-40A	97.08	43.36	45.34	8.38	0.758	0.215	0.011	0.016	0.756	0.215	0.011	0.016	50.02	9.12	40.86	55.07	5.77	32.79	51.62	2.06	16.29	0.00	13.31	17.07
S1904-42A	95.17	44.20	42.71	8.26	0.646	0.296	0.037	0.021	0.645	0.296	0.037	0.021	49.81	9.42	40.77	53.98	6.12	32.64	51.06	2.36	15.96	0.01	12.92	17.27
S1905-59	93.04	45.91	39.47	7.66	0.535	0.333	0.112	0.022	0.532	0.332	0.112	0.022	49.86	9.30	40.84	54.96	6.30	32.10	51.39	2.43	15.9	0.01	14.30	16.76
S1912-04A	90.55	44.29	38.67	7.59	0.536	0.286	0.131	0.04 <sup>4</sup>	0.537	0.288	0.132	0.041	49.93	9.25	$40.8^{\circ}$	54.90	6.10	32.5i	51.34	2.85	15.95	0.00	12.70	18.71
S1912-06	95.45	44.02	43.10	8.33	0.687	0.249	0.046	0.018	0.684	0.249	0.046	0.018	49.79	9.37	40.84	54.63	6.20	31.92	50.97	2.76	15.37	0.01	12.32	18.78
S1913-01	94.90	43.19	43.42	8.29	0.716	0.203	0.055	0.023	0.716	0.204	0.055	0.023	49.88	9.28	40.84	54.70	6.04	32.32	51.30	2.56	15.57	0.01	11.94	18.87
S1913-07A	92.72	43.89	40.70	8.13	0.599	0.294	0.070	0.033	0.600	0.295	0.070	0.033	49.71	9.46	40.83	54.06	6.18	31.85	50.85	3.17	16.16	0.00	12.90	18.97
S1923-38A	96.95	43.20	45.06	8.69	0.748	0.233	0.000	0.018	0.747	0.233		0.018	49.84	9.38	40.78	54.55	6.24	$32.4^{\circ}$	50.98	2.37	15.98	0.00	12.56	18.07
S1923-42A	96.51	42.42	45.48	8.61	0.779	0.190	0.002	0.024	0.781	0.191	0.002	0.024	49.98	9.19	40.83	55.23	6.03	32.58	51.36	2.37	16.03	0.02	12.67	18.10
S1923-84A	96.79	42.97	45.26	8.57	0.764	0.195	0.023	0.017	0.763	0.195	0.023	0.017	49.99	9.29	40.72	54.83	6.12	33.22	50.95	2.67	16.44	0.02	12.50	18.39
S1923-86A	94.95	41.90	44.25	8.80	0.739	0.204	0.012	0.036	0.745	0.206	0.012	0.036	49.90	9.39	40.71	54.88	6.50	32.7'	50.93	2.59	16.12	0.01	14.06	17.41
S1924-01A	95.23	42.73	44.20	8.31	0.758	0.146	0.074	0.020	0.758	0.146	0.074	0.020	50.05	9.21	40.74	55.22	6.18	33.27	51.38	2.45	15.56	0.02	12.55	18.84
S1924-04	96.17	45.34	42.59	8.25	0.632	0.321	0.036	0.014	0.628	0.320	0.036	0.014	49.86	9.31	40.83	54.99	6.43	32.12	51.58	3.50	15.85	0.00	12.09	19.30
S1924-12	93.02	40.99	43.22	8.81	0.744	0.155	0.045	0.042	0.752	0.157	0.046	0.043	49.71	9.58	$40.7^{\circ}$	54.08	6.60	32.25	50.55	2.48	14.80	0.00	12.70	19.17
S1927-03A	96.20	43.43	44.25	8.52	0.736	0.200	0.048	0.016	0.734	0.200	0.048	0.016	49.91	9.36	40.73	55.01	6.43	$32.6^{\circ}$	50.90	2.88	16.36	0.00	13.00	17.83
S1927-05A	95.72	43.35	43.84	8.54	0.707	0.238	0.031	0.022	0.706	0.238	0.031	0.022	49.86	9.40	40.74	54.42	6.41	32.75	51.36	2.57	$16.2^{\circ}$	0.01	12.95	17.94
S1928-09A	93.26	45.18	40.19	7.89	0.545	0.360	0.065	0.029	0.543	0.360	0.065	0.029	49.84	9.37	40.78	54.60	6.21	32.36	50.89	2.69	15.54	0.03	13.00	18.16
S1928-10	94.33	43.90	42.23	8.20	0.627	0.310	0.028	0.031	0.633	0.314	0.028	0.031	49.88	9.34	40.78	54.51	5.99	32.58	50.74	2.71	15.35	0.03	13.06	17.92
S1928-15A	93.93	44.06	41.70	8.17	0.619	0.296	0.052	0.030	0.618	0.297	0.052	0.030	49.88	9.35	40.77	54.69	6.20	32.52	50.92	2.66	15.58	0.02	13.79	17.83
S1928-19	94.67	43.37	42.97	8.34	0.677	0.253	0.037	0.029	0.679	0.254	0.037	0.029	49.91	9.32	40.78	54.92	6.17	$32.5^{\circ}$	50.86	2.63	15.75	0.03	12.94	18.09
S2209-01A	97.50	43.40	45.48	8.62	0.787	0.163	0.048	0.005	0.784	0.163	0.048	0.005	49.85	9.41	40.74	54.35	6. 15	32.72	50.72	2.76	16.8	0.02	15.13	16.29
S2220-04	98.16	43.98	45.72	8.47	0.761	0.228	0.006	0.008	0.758	0.228	0.006	0.008	50.00	9.17	40.83	55.19	5.98	$32.7^{\circ}$	51.46	2.76	16.4	0.01	13.83	17.30
S2220-06	98.76	43.68	46.45	8.63	0.802	0.187	0.013	0.003	0.798	0.186	0.013	0.003	50.00	9.22	40.78	55.30	6.13	32.72	51.09	3.01	16.45	0.04	16.08	16.57
S2221-01A	95.16	45.15	42.01	7.99	0.608	0.315	0.064	0.016	0.606	0.315	0.064	0.016	49.92	9.39	40.69	54.51	6.12	33.09	50.45	2.45	15.99	0.02	12.41	18.00
S2221-04B	96.61	43.04	44.87	8.70	0.755	0.199	0.030	0.016	0.755	0.199	0.030	0.016	49.85	9.45	40.70	54.29	6.38	32.90	50.87	2.85	16.13	0.01	13.46	17.68
S2221-05B	97.13	43.52	45.04	8.57	0.752	0.215	0.023	0.012	0.750	0.215	0.023	0.012	49.88	9.37	40.75	54.54	6. 12	32.72	50.75	2.78	15.66	0.00	12.91	17.67
P7003-23A	98.16	42.20	47.25	8.71	0.857	0.120	0.011	0.011	0.858	0.120	0.011	0.011	50.07	9.10	40.83	55.26	6.02	33.12	52.10	2.67	17.13	0.00	14.66	16.27

**Table S4.** *Regression results and correlation values. Regressions have been performed for each oxide pair in order to derive the correlation between major oxides in pyroxenes and in olivine. Plots refer to panels in figure S7.* 

		$Mg_{Ol}$			Si <sub>Ol</sub>			Fe <sub>Ol</sub>		
		Int	<b>Slope</b>	${\bf R}^2$	Int	<b>Slope</b>	${\bf R}^2$	Int	<b>Slope</b>	${\bf R}^2$
$Plot\ I\text{-}\mathbf{Mg_{opx}}$	Value	34.20	0.479	0.46	38.30	0.074	0.02	21.09	$-0.372$	0.36
	σ	1.40	0.043		1.33	0.041		1.33	0.041	
$Plot 2-Si_{\text{opx}}$	Value	28.90	0.378	0.43	33.14	0.138	0.11	29.35	$-0.369$	0.54
	σ	1.96	0.036		1.73	0.032		1.55	0.028	
$Plot 3 - Feopx$	Value	52.78	$-0.547$	0.11	42.35	$-0.289$	0.06	3.96	0.893	0.41
	σ	0.72	0.124		0.52	0.090		0.51	0.089	
$Plot 4 - Mgcpx$	Value	49.82	$-0.010$	0.01	39.94	0.041	0.01	9.85	$-0.041$	0.01
	σ	0.59	0.034		0.40	0.023		0.49	0.028	
$Plot 5-Si_{cpx}$	Value	35.66	0.271	0.24	34.15	0.126	0.11	24.79	$-0.304$	0.45
	σ	2.00	0.039		1.49	0.029		1.40	0.027	
Plot 6- $\mathrm{Fe_{cpx}}$	Value	51.23	$-0.556$	0.26	40.86	$-0.069$	0.00	7.71	0.497	0.31
	σ	0.22	0.075		0.17	0.060		0.17	0.060	

**Table S5.** *Temporal variations of spreading rate (Vs ), ridge length (excluding transform faults), mean crustal thickness (Hc ), oceanic crustal production rate, and volume (m<sup>3</sup> /yr) of mantle rocks (Vol<150°C) that can interact yearly with seawater at T<150 °C estimated along present day and paleo mid-ocean ridges since 150 Ma.* 

