

Geological storage of CO₂ within the oceanic crust by gravitational trapping

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Received 27 October 2013; accepted 4 November 2013; published 3 December 2013.

[1] The rise of atmospheric carbon dioxide (CO₂) principally due to the burning of fossil fuels is a key driver of anthropogenic climate change. Mitigation strategies include improved efficiency, using renewable energy, and capture and long-term sequestration of CO₂. Most sequestration research considers CO₂ injection into deep saline aquifers or depleted hydrocarbon reservoirs. Unconventional suggestions include CO₂ storage in the porous volcanic lavas of uppermost oceanic crust. Here we test the feasibility of injecting CO₂ into deep-sea basalts and identify sites where CO₂ should be both physically and gravitationally trapped. We use global databases to estimate pressure and temperature, hence density of CO₂ and seawater at the sediment-basement interface. At previously suggested sites on the Juan de Fuca Plate and in the eastern equatorial Pacific Ocean, CO₂ is gravitationally unstable. However, we identify five sediment-covered regions where CO₂ is denser than seawater, each sufficient for several centuries of anthropogenic CO₂ emissions. **Citation:** Marieni, C., T. J. Henstock, and D. A. H. Teagle (2013), Geological storage of CO₂ within the oceanic crust by gravitational trapping, *Geophys. Res. Lett.*, 40, 6219–6224, doi:10.1002/2013GL058220.

1. Introduction

[2] Human activities since the industrial revolution have increased atmospheric concentrations of greenhouse gases, in particular carbon dioxide (CO₂), requiring the development of mitigation strategies to minimize the effect on the global climate and potential ocean acidification [*Intergovernmental Panel on Climate Change*, 2007]. Various strategies have been proposed to reduce CO₂ emission including reducing energy demand, increasing renewable energy, and carbon capture and storage (CCS) underground. The effectiveness of geological reservoirs depends on their storage capacity, reservoir stability, risk of leakage, and the retention time [*Hawkins*, 2004; *Rochelle et al.*, 2004], with deep saline sedimentary aquifers [*Eccles and Pratson*, 2012; *House et al.*, 2006; *Levine*

et al., 2007; *Schrag*, 2009], and depleted oil and gas reservoirs [*Bachu*, 2000; *Jessen et al.*, 2005] receiving the greatest research attention. In addition, several mafic and ultramafic formations are under consideration for CO₂ storage including lava flows on Iceland [*Gislason et al.*, 2010; *Oelkers et al.*, 2008] and the Columbia River Basalts in the United States [*McGrail et al.*, 2006].

[3] This paper investigates the geological storage of CO₂ in the deep-sea basalts [*Goldberg et al.*, 2008, 2010; *Matter et al.*, 2007; *Slagle and Goldberg*, 2011] that form the uppermost igneous lavas of the oceanic crust and cover approximately 60% of Earth's surface. These formations may have advantages over other potential geological storage options: (a) large reservoir capacities; (b) low risk of postinjection leakage due to low permeability sediment blankets in some regions; (c) in situ availability of water; and (d) estimated fluid retention times greater than 500 years [*Goldberg et al.*, 2008]. The Juan de Fuca Plate (JdFP), offshore Washington State, has been the focus of conceptual studies of deep-sea basalt CCS [*Goldberg et al.*, 2008] because it is the best studied mid-ocean ridge flank with well-characterized regional thermal and hydrological regimes [*Fisher and Davis*, 2000]. The rocks on this plate are relatively young, having formed at the Juan de Fuca Ridge less than 11 Myr ago. The pillow lavas that form the upper few hundred meters of the JdFP crust have high connected porosity (>10%) [*Fisher*, 1998], and the ridge flank is blanketed by a thick (from 30 to over 700 m) sequence of hemipelagic and turbiditic sediments derived from the North American continent.

[4] Other well-studied regions of the upper oceanic crust that have been considered for CO₂ storage [*Slagle and Goldberg*, 2011] are in the eastern equatorial Pacific Ocean (eePO) at Sites 504 and 1256 located in ~7 and 15 Myr old crust, respectively. Numerous studies on hydrothermal circulation provide detailed descriptions of physical properties and the porosity of the extrusive section of the ocean crust at these sites [*Alt et al.*, 1993; *Teagle et al.*, 2006; *Wilson et al.*, 2006].

[5] Three primary trapping mechanisms for the long-term storage of carbon dioxide in seafloor basalts have been proposed: (1) *Gravitational trapping* under pressure and temperature conditions where CO₂ is more dense than seawater [*Levine et al.*, 2007]. (2) *Physical or permeability trapping*, where the presence of ≥200 m of overlying low permeability marine sediments isolate the CO₂ injected into the basalts from the oceans, so that any leakage is trapped in the sediments [*Goldberg and Slagle*, 2009]. (3) *Geochemical trapping*, where the CO₂ and water react with the basalt host rocks to form geologically stable carbonate minerals [*Matter et al.*, 2007].

[6] In this study we consider the global variability of sediment thickness, pressure (*p*), and temperature (*T*), and consequently, the density (*ρ*) of CO₂ and seawater at the sediment-basement interface of the oceanic crust, to identify

Additional supporting information may be found in the online version of this article.

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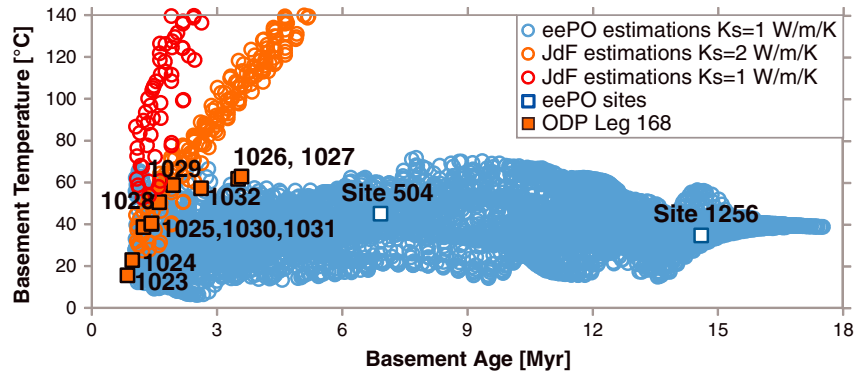


Figure 1. Comparison between estimated temperatures in the eastern equatorial Pacific Ocean (eePO) and the Juan de Fuca Plate (JdFP), and measured downhole temperatures at the sediment-basement interface. White squares: data from eePO [Alt et al., 1993; Teagle et al., 2006]; orange squares: data from JdFP [Davis et al., 1997]. Circles: estimated values in the eePO (blue), and on the JdFP (red with $K_s = 1$ W/m/K; orange with $K_s = 2$ W/m/K).

potential targets for combined gravitational and physical CO₂ sequestration. Although this is a physically robust scoping study, detailed programs of local data acquisition are imperative before any targets could be further developed.

2. Physical Parameters

[7] We have developed global maps of the density of seawater and CO₂ at the sediment-ocean crust interface. Pressure was estimated using the NOAA-gridfive sediment thickness database [Divins, 2003] combined with the General Bathymetric Chart of the Oceans (GEBCO)-gridfive world bathymetry map (both 6' × 6' grids) [IOC et al., 2003]. The anomalies in the topography (e.g., abyssal hills and seamounts) are not always detected by global altimetry and gravity analyses, but the NOAA database is the currently best available. We assume a hydrostatic load to the top of basement, using a constant seawater density of 1030 kg/m³ and a constant salinity of 35 psu (practical salinity unit) [Brown et al., 1995]; this gives a lower bound on the pressure and the CO₂ density. We used the Global Depth and Heat flow model (GDH1) [Stein and Stein, 1992] to estimate the heat flow, and then the temperature, based on the oceanic crustal age [Müller et al., 2008] (see supporting information “Text S1” for the equations). We use GDH1 because it is better for predicting the heat flow at old oceanic crustal ages than the Half Space Cooling Model (HSCM) [Turcotte and Schubert, 2002], and because the Global Heat Flow Database [Pollack et al., 1993] is sparse and irregular. Nevertheless, our GDH1-based approach still presents uncertainties due to incomplete information on sediment thicknesses, local heat flow anomalies, and the thermal properties of sediments.

[8] From the heat flow we have calculated the geothermal gradient [Heberling et al., 2010] and consequently estimated the temperature at the top of the basement (in °C), with the thermal conductivity of the sediments taken as $K_s = 1$ W/m/K [Pollack et al., 1993; Pribnow et al., 2000a] (see supporting information “Figure S2”).

[9] We have validated our estimates of temperature at the sediment-basement interface by using borehole temperature logs from Sites 504 and 1256 [Alt et al., 1993; Teagle et al., 2006] and the Juan de Fuca Plate [Davis et al., 1997] (Figure 1; supporting information “Figure S1” for comparison with HSCM). The two areas have different trends of

basement-sediment interface temperature as a function of age. In the eastern equatorial Pacific Ocean there is good agreement between estimated and measured temperature. However, on the Juan de Fuca Plate, the temperatures at most sites are better fit using the higher measured thermal conductivity ($K_s \sim 2$ W/m/K) of the local muddy and sandy turbiditic sediments [Pribnow et al., 2000b]. At Sites 1026 and 1027, the measured temperature is lower than predicted because of hydrothermal circulation [Hutnak et al., 2006; Wheat et al., 2004] linked to surrounding basement outcrops (e.g., Baby Bare) [Fisher et al., 2003]. Although there are numerous holes drilled into the oceanic crust by scientific ocean drilling (Deep Sea Drilling Project (DSDP), Ocean Drilling Program (ODP), and Integrated Ocean Drilling Program (IODP)), eePO, and JdF are the only locations where the temperatures close to the sediment-basement interface are well constrained. The validation at the eePO and JdFP gives us confidence in our calculations but emphasizes the need for verification of local physical properties.

[10] The densities of CO₂ (ρ_{CO_2}) and seawater (ρ_{seawater}) were calculated for pressures from 0 to 60 MPa and temperatures from 0 to 100°C. The CO₂ density was determined by interpolating the online National Institute of Standards and

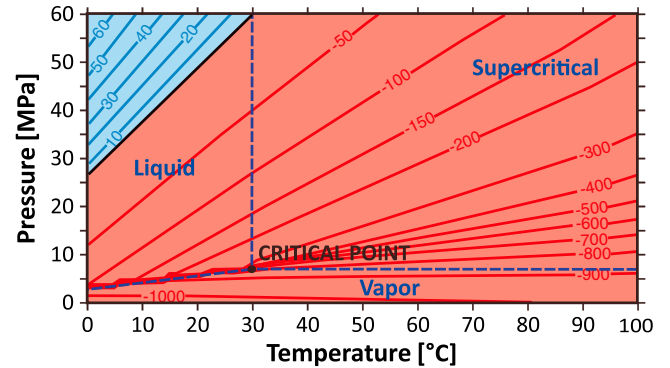


Figure 2. Density difference ($\Delta\rho = \rho_{\text{CO}_2} - \rho_{\text{seawater}}$ in kg/m³) between CO₂ and seawater density as a function of pressure between 0 and 60 MPa, and temperature between 0 and 100°C, with the phase diagram of CO₂ overlaid. Positive differences shown in blue indicate conditions for gravitational trapping.

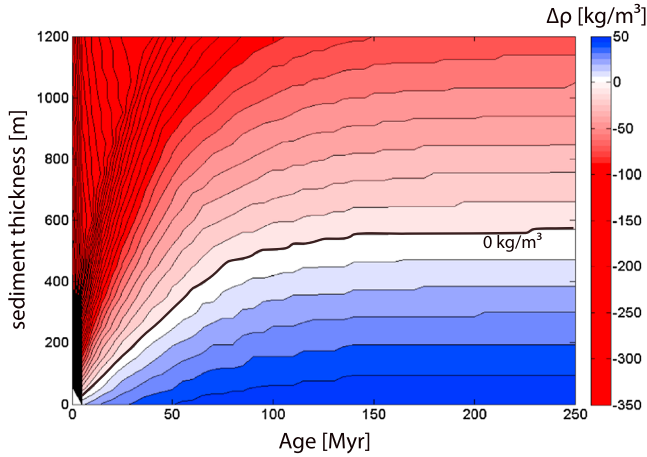


Figure 3. Density difference between CO₂ and seawater at the sediment-basement interface as a function of plate age and sediment thickness using the GDH1 model to determine both water depth and thermal conditions. Sediment thicknesses below the heavy black line show where positive density differences required for stable gravitational trapping are achieved.

Technology (NIST) database (Linstrom, P. J., and W. G. Mallard (Eds.), NIST Chemistry WebBook, NIST Standard Reference Database Number 69, National Institute of Standards and Technology, Gaithersburg Md, 20899, retrieved November 12, 2012, <http://webbook.nist.gov>), which is based on the equation of state by *Span and Wagner* [1996]. Seawater density was estimated using the SeaWater MATLAB library [Fofonoff *et al.*, 1983], assuming a constant salinity of 35 psu [Brown *et al.*, 1995]. Figure 2 shows the density difference

between CO₂ and seawater as a function of temperature and pressure together with a phase diagram of carbon dioxide. Temperatures above 100°C are not considered because the density of CO₂ is too low to allow gravitational trapping.

[11] The density difference at the sediment-basement interface at each point in our 6' × 6' global grids is calculated using the estimated temperature and pressure. We combined this with the sediment thickness map to identify locations where (1) CO₂ is denser than seawater at the sediment-basement interface (Figure 2), and (2) the sediment thickness is between 200 m and 700 m (Figure 3). We choose a minimum thickness of 200 m to ensure a continuous low permeability blanket over minor basement topography such as fault ridges or seamounts that might puncture the sediment cover and allow the egress of basement fluids. To estimate the maximum sediment thickness, we have calculated the density difference for a wide range of lithospheric ages and sediment thicknesses using the GDH1 model for both water depth and heat flow, and assuming a hydrostatic sediment column (Figure 3). Based on global average conditions, GDH1 indicates a restricted zone where gravitational trapping is possible, and that anywhere with more than ~600 m of sediments CO₂ is likely to be gravitationally unstable due to the high temperatures. Using the HSCM (see supporting information “Figure S3”), the equivalent limit is ~1000 m. Hence, we settle on an upper sediment thickness limit of 700 m.

3. Discussions

[12] Much of the upper oceanic crust does not provide suitable locations for the geological sequestration of CO₂ by gravitational and physical trapping. Gravitational trapping ($\rho_{\text{CO}_2} > \rho_{\text{seawater}}$) requires pressures higher than 25 MPa

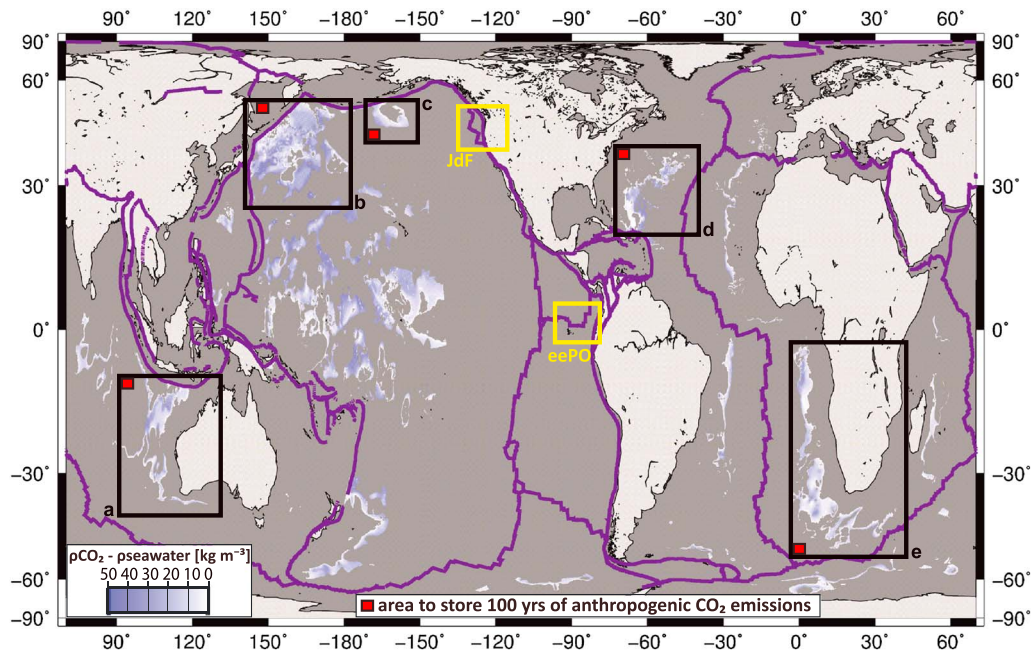


Figure 4. An equal area map showing locations for stable geological sequestration of CO₂. Shading shows the difference in density between CO₂ and seawater in areas where the sediment thickness is between 200 and 700 m and the CO₂ is denser than seawater. Five potential reservoirs (insets a–e) have been identified. The red box indicates the area required to store 100 yrs of current anthropogenic emissions of CO₂, assuming a pillow lava thickness of 300 m and 10% porosity [Carlson and Herrick, 1990; Jarrard *et al.*, 2003; Johnson and Pruis, 2003]. Yellow boxes show regions in Figure 5.

Table 1. Properties of the Five Potential Reservoirs^a

Code	Location	Area [$\times 10^6$ km ²]	Pore Volume [$\times 10^4$ km ³]	ρ_{CO_2} [kg/m ³]	CO ₂ [Gt]	$\Delta\rho$ [kg/m ³]	Age [Myr]	Sed. Thickness [m]	Distance [km]
a	Indian Ocean	1.47	4.42	1066	47,162	18	85	335	1500
b	NW Pacific	3.97	11.9	1073	127,870	24	100	310	1300
c	S-Aleutians	0.43	1.30	1063	13,791	15	60	275	950
d	Bermuda	1.15	3.45	1066	36,780	17	80	320	1500
e	SE Atlantic	2.22	6.66	1062	70,701	14	85	290	1700

^a $\Delta\rho$, in situ excess density of CO₂ over seawater; age from [Müller et al., 2008]; sediment thickness from the NOAA database [Divins, 2003]. Distance of the reservoir from land is taken from the nearest stationary source of CO₂ according to the IEA GHG database [International Energy Agency Greenhouse Gas R&D Programme, 2002].

(~2500 m of water) and temperature between 0 and 30°C (Figure 2). The density of CO₂ decreases dramatically with decreasing pressure and increasing temperature, compared to a near constant density for seawater. The combination of high pressure and low temperature requires old ocean crust with relatively thin sediments. Note that within this p - T window liquid CO₂ is the thermodynamically stable phase, with densities between 1040 and 1125 kg/m³ compared with 140 kg/m³ (at 100°C, 8 MPa) to 1045 kg/m³ (at 30°C, 60 MPa) for supercritical CO₂.

[13] Suitable reservoirs where physical (sediment between 200 and 700 m) and gravitational ($\rho_{\text{CO}_2} > \rho_{\text{seawater}}$) trapping can be combined are shown on the global map (Figure 4); the global map produced using the HSCM is available in the supporting information “Figure S4”. Selected potential reservoirs are in the Indian Ocean between Indonesia and Australia (inset a); in the northwest Pacific Ocean near the east coast of Japan and Russia (inset b), and south of the Aleutian Islands (inset c); and in the Atlantic Ocean near Bermuda (inset d) and close to South Africa (inset e) (Table 1).

[14] We have identified these sites based on the positive $\Delta\rho$ between CO₂ and seawater, the oceanic crustal age, the sediment thickness, and the distance to major industrial CO₂ sources [International Energy Agency Greenhouse Gas R&D Programme, 2002] (Table 1). Other areas also have suitable conditions for carbon dioxide trapping, but we have not yet explored these options due to their smaller sizes and lower $\Delta\rho$, although some are closer to land. We have computed the potential storage volume for each target (Table 1), assuming 300 m as reasonable thickness of permeable pillow lavas for old crust. Given an average porosity of 10% [Carlson and Herrick, 1990; Johnson and Pruis, 2003], even for old oceanic crust (e.g., ODP Hole 801C [Jarrard et al., 2003]), we determine the potential pore volume. The storage capacity in each area is between ~13,800 and 127,800 Gt of CO₂. At the current global annual anthropogenic flux of ~35 Gt of CO₂ per year [Le Quere et al., 2009], even the smallest identified reservoir (inset c), could provide sufficient carbon dioxide sequestration capacity for several centuries (Figure 4).

[15] Contrary to previous suggestions [Goldberg et al., 2008; Slagle and Goldberg, 2011], sites on the Juan de Fuca Plate and in the eastern equatorial Pacific Ocean are unsuitable for gravitational trapping of carbon dioxide (Figure 5) because thick sediment covering young oceanic crust results in high temperatures at the sediment-basement interface, that renders CO₂ less dense than seawater.

[16] Our evaluation based on global data sets shows that CCS using subsea basalts as the storage medium has

considerable potential. However, regional investigations are needed to determine local sediment properties, thicknesses, continuity, and seafloor thermal gradients. Drilling to facilitate detailed lithological, physical, thermal, and hydrological characterization of the sediment overburden and target basalt formations is essential.

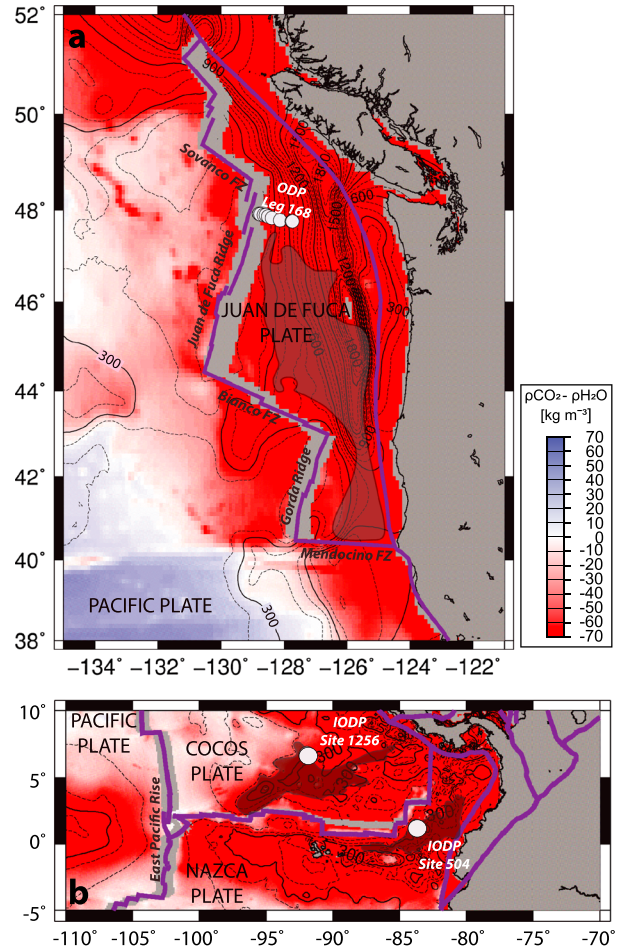


Figure 5. Map of density difference $\Delta\rho$ between CO₂ and seawater at (a) Juan de Fuca Plate (with $K_s = 2$ W/m/K) and (b) at eastern equatorial Pacific Ocean (with $K_s = 1$ W/m/K). Sediment thicknesses are shown with black contour lines. The dark shadows show the previously suggested regions for deep-sea basalt CO₂ sequestration [Goldberg et al., 2008; Slagle and Goldberg, 2011].

4. Conclusions

[17] Offshore basalt formations have been previously suggested as sites for geological carbon dioxide sequestration. We have used global data compilations to calculate the density of CO₂ in the pressure-temperature regime at the top of the basement throughout the world's oceans, and identified regions where CO₂ is denser than seawater. Previously suggested young sites on the eastern flank of the JdF Ridge and in the eastern equatorial Pacific (Sites 504, 1256) are not suitable for storing CO₂ because it is gravitationally unstable ($\Delta\rho_{\text{CO}_2 - \text{seawater}} = 0$ to $< -70 \text{ kg/m}^3$). However, we identify five large regions of old seabed where gravitational stability of stored CO₂ coincides with physical trapping by 200–700 m thickness of sediments. Using conservative assumptions about the porosity available, the smallest of these regions can store several centuries of anthropogenic CO₂ emissions.

[18] **Acknowledgments.** This research used data provided by the Integrated Ocean Drilling Program (IODP), the Ocean Drilling Program (ODP), and the Deep-Sea Drilling Project (DSDP). Funding for this research was provided by the University of Southampton Vice Chancellor's Scholarship to C. M., and NERC grant NE/I006311/1 to D.A.H.T. We thank Juerg M. Matter, Nick Woodman, and Martin R. Palmer for their discussions that assisted in the development of these ideas. We thank the Editor Peter Strutton, Robert Harris and a second anonymous reviewer for their constructive comments that have improved the paper.

[19] The Editor thanks Robert Harris and an anonymous reviewer for their assistance in evaluating this paper.

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