1 Objective estimates of mantle ^3He in the ocean and
2 implications for constraining the deep ocean circulation

3 Mark Holzer

4 *Department of Applied Mathematics, School of Mathematics and Statistics, University of*
5 *New South Wales, NSW 2052, Australia.*

6 *Also at Department of Applied Physics and Applied Mathematics, Columbia University,*
7 *New York, NY, USA*

8 Timothy DeVries

9 *Department of Geography / Earth Research Institute, University of California, Santa*
10 *Barbara, CA 93106-4060, USA*

11 Daniele Bianchi

12 *Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles,*
13 *CA 90095-1565, USA*

14 Robert Newton, Peter Schlosser*, Gisela Winckler

15 *Lamont-Doherty Earth Observatory, Columbia University, Palisades, NY 10964, USA.*

16 **Also at Department of Earth and Environmental Sciences and Department of Earth and*
17 *Environmental Engineering, Columbia University, New York, NY 10025, USA*

18 **Abstract**

Hydrothermal vents along the ocean's tectonic ridge systems inject superheated water and large amounts of dissolved metals that impact the deep ocean circulation and the oceanic cycling of trace metals. The hydrothermal fluid contains dissolved mantle helium that is enriched in ^3He relative to the atmosphere, providing an isotopic tracer of the ocean's deep circulation and a marker of hydrothermal sources. This work investigates the potential for the $^3\text{He}/^4\text{He}$ isotope ratio to constrain the ocean's mantle ^3He source and to provide constraints on the ocean's deep circulation. We use an ensemble of 11 data-assimilated steady-state ocean circulation models and a mantle helium source based on geographically varying sea-floor spreading rates. The global source distribution is partitioned into 6 regions, and the vertical profile and source amplitude of each region are varied independently to determine the optimal ^3He source distribution that minimizes the mismatch between modeled and observed $\delta^3\text{He}$. In this way, we are able to

Preprint submitted to Elsevier

September 1, 2016

fit the observed $\delta^3\text{He}$ distribution to within a relative error of $\sim 15\%$, with a global ^3He source that ranges from 640 to 850 mol yr $^{-1}$, depending on circulation. The fit captures the vertical and interbasin gradients of the $\delta^3\text{He}$ distribution very well and reproduces its jet-sheared saddle point in the deep equatorial Pacific. This demonstrates that the data-assimilated models have much greater fidelity to the deep ocean circulation than other coarse-resolution ocean models. Nonetheless, the modelled $\delta^3\text{He}$ distributions still display some systematic biases, especially in the deep North Pacific where $\delta^3\text{He}$ is overpredicted by our models, and in the southeastern tropical Pacific, where observed westward-spreading $\delta^3\text{He}$ plumes are not well captured. Sources inferred by the data-assimilated transport with and without isopycnally aligned eddy diffusivity differ widely in the Southern Ocean, in spite of the ability to match the observed distributions of CFCs and radiocarbon for either eddy parameterization.

19 *Keywords:* mantle helium-3, ocean circulation, inverse modelling

20 1. Introduction

21 Measurements of helium isotopes in the ocean provide important infor-
 22 mation on the early history of the earth, on the geochemical mantle-to-ocean
 23 fluxes of elements such as iron, that are correlated with mantle helium fluxes,
 24 and on the state of the current ocean circulation. Mantle ^3He is thought to
 25 be primordial in origin, having been trapped in the solid earth during the
 26 formation of the solar system. By contrast, the much more abundant ^4He
 27 is continually produced in the earth's interior via the uranium and thorium
 28 radioactive decay series. Mantle helium, enriched in ^3He relative to ^4He com-
 29 pared to the atmosphere, dissolves in hydrothermal fluid that is injected into
 30 the ocean interior at the mid-ocean ridges, where upwelling magma drives
 31 the spreading of tectonic plates and the formation of new seafloor. Mantle
 32 helium is therefore a unique oceanic tracer that has sources in the ocean
 33 interior, i.e., far from the ocean surface where most other transport trac-
 34 ers such as radiocarbon, tritium, and long-lived anthropogenic gases (e.g.,
 35 CFCs, SF_6) enter the ocean. ^3He is particularly promising for providing con-
 36 straints on the abyssal circulation and, indirectly, on the pathways and rates
 37 of interior-to-surface transport.

38 The value of ^3He as a constraint on the ocean's deep circulation and
 39 on the ventilation of abyssal water masses has been widely recognized (e.g.,
 40 *Farley et al.*, 1995; *Lupton*, 1998; *Dutay et al.*, 2004; *Garabato et al.*, 2007;
 41 *Bianchi et al.*, 2010; *Gnanadesikan et al.*, 2015). In fact, the simulation
 42 of ^3He for specified sources has been one of the components of the Ocean

Carbon Model Intercomparison Project (OCMIP). Intercomparing six coarse-resolution ocean models, *Dutay et al.* (2004) found that the fidelity of the simulated $\delta^3\text{He}$ distribution depends sensitively on the parameterization of eddy transport and that all models failed to capture the observed structure of the ^3He plumes in the deep ocean. More recently, *Bianchi et al.* (2010) used a suite of circulation models that are able to produce realistic radiocarbon distributions to constrain the global mantle ^3He source. *Bianchi et al.* (2010) found that the original box-model estimate of $1100 \pm 300 \text{ mol yr}^{-1}$ (*Clarke et al.*, 1969; *Craig et al.*, 1975) is likely too high and they estimate a ^3He source of $527 \pm 102 \text{ mol yr}^{-1}$.

Here our focus is on (i) constraining the sources of ^3He using data-assimilated global circulations, and on (ii) investigating how well these circulations capture the observed $\delta^3\text{He}$ distribution. Objective (i) is similar to that of *Bianchi et al.* (2010), but with three key differences: First, we optimize the mantle ^3He source by objectively minimizing the mismatch between the modeled and observed three-dimensional $\delta^3\text{He}$ distributions, and not by matching only the global non-atmospheric ^3He inventory as in *Bianchi et al.* (2010). Second, we use a more flexible source parameterization by allowing the ^3He source strengths and injection depths to vary regionally. Third, we use circulations that were not merely tuned, but rigorously data-constrained by assimilating both tracer and dynamical information into an ocean circulation inverse model (OCIM, *DeVries and Primeau*, 2011; *Primeau et al.*, 2013; *DeVries*, 2014). In the OCIM, the three-dimensional ocean circulation is objectively optimized using an adjoint approach with potential temperature, salinity, radiocarbon, and CFC-11 as constraints. This guarantees a good fit between the modeled and observed distributions of these transport tracers, thereby minimizing errors in the simulated ocean circulation. Objective (ii) is motivated by the fact that the transient tracers used in the data assimilation have surface sources, with only radiocarbon having a long enough timescale to provide constraints on the abyssal circulation. However, both the surface-to-interior and interior-to-surface transports are characterized by very broad transit-time distributions (e.g., *Primeau*, 2005; *Primeau and Holzer*, 2006) so that constraining transport accurately requires tracers with as many different timescales as possible. In the upper ocean, both CFCs and radiocarbon, with their widely separated timescales, constrain transport reasonably well but in the deep ocean the constraints come dominantly from radiocarbon with its single very long timescale. It is therefore important to investigate the potential of ^3He as an additional tracer constraint on the deep ocean circulation.

82 2. Model

83 2.1. Circulations

84 We use 11 data-assimilated global, steady ocean circulations. These cir-
85 culations have a $2^\circ \times 2^\circ$ horizontal resolution with 24 depth levels, ranging
86 from a thickness of 36 m at the surface, to ~ 300 m near 2000 m depth, to
87 ~ 600 m in the abyssal ocean. All these circulations were obtained using
88 CFCs and radiocarbon as constraints, in addition to potential temperature,
89 salinity, sea-surface height and surface heat and fresh-water fluxes. One of
90 the circulations additionally used phosphate as a constraint (*Primeau et al.*,
91 2013). This circulation’s transport operator has horizontal and vertical eddy
92 diffusivities of $10^3 \text{ m}^2 \text{ s}^{-1}$ and $10^{-5} \text{ m}^2 \text{ s}^{-1}$, respectively, and we will for
93 convenience refer to it as the “C-grid circulation” because of its numerical
94 grid. All other circulations have transport operators with a rotated diffusiv-
95 ity tensor (*DeVries*, 2014). *DeVries* (2014) generated an ensemble of these
96 circulations by varying the relative weights of the constraints in the data as-
97 simulation and by varying the isopycnal and diapycnal diffusivities. Because
98 these circulations are formulated on a numerical B grid, we will refer to them
99 as “B-grid circulations”.

100 2.2. Helium model

101 For each circulation, the advective-eddy-diffusive transport operator is
102 organized into a matrix $\mathbf{T}$. Organizing the gridded helium concentration
103 field into a corresponding vector χ , the steady-state helium equation is given
104 by

$$105 \quad \mathbf{T} \chi = \mathbf{K}(\chi_{\text{sat}} - \chi) + S \quad , \quad (1)$$

106 where $\mathbf{K} = \text{diag}(K)$, with air-sea exchange coefficients K that are non-zero
107 only in the surface layer. In (1), S is the mantle source, and χ_{sat} is the
108 helium concentration for solubility equilibrium with the atmosphere. We
109 solve two independent versions of this equation, one for ^3He and one for
110 ^4He (concentrations $^3\chi$ and $^4\chi$, respectively). The gas-exchange velocities,
111 Schmidt numbers, and sea-ice masks that enter $\mathbf{K}$ are all taken from the
112 $1^\circ \times 1^\circ$ OCMIP fields and interpolated onto the model’s $\sim 2^\circ \times 2^\circ$ grid.

113 2.3. Source model

114 Following *Farley et al.* (1995), the OCMIP protocol assumes the ^3He
115 sources to be located at the crests of mid-ocean ridges with a local magnitude
116 proportional to the sea-floor spreading rate based on the tectonic model of
117 *DeMets et al.* (1990). The OCMIP protocol provides the three-dimensional
118 location of the sources on a $1^\circ \times 1^\circ$ grid, with the injection depth of each source
119 300 m above the real ocean’s ridge axis. The 300-m offset is applied globally

120 and intended to approximate the depth where hot hydrothermal fluid has
 121 cooled enough to become neutrally buoyant. Off-ridge helium sources (e.g.,
 122 from sediments) are not modelled, effectively assigning such sources to the
 123 mid-ridge hydrothermal injections.

124 To allow for regional variations in the source strength of the overall
 125 spreading pattern, we partition the OCMIP source S into N regional parts
 126 S_Ω , where $\Omega = 1 \dots N$ labels the region. Because of the coarse vertical reso-
 127 lution of the circulations used, and the somewhat arbitrary globally uniform
 128 300 m vertical offset, we allow some flexibility in the depth of injection as
 129 follows: If the OCMIP injection depth falls in the grid box of level k with
 130 horizontal indices i, j , we construct the source field as a linear combination of
 131 3 weighted stacks of sources, one corresponding to the interpolated OCMIP
 132 field, one shifted down uniformly over the region by one grid box, and one
 133 similarly shifted up by one grid box. More precisely,

$$134 \quad S(i, j, k) = \sum_{\Omega=1}^N \sum_{k'=-1}^{+1} \sigma_{\Omega, k'} S_\Omega(i, j, k + k') \quad , \quad (2)$$

135 where the $3N$ coefficients $\sigma_{\Omega, k'}$ are optimized as described below. Even with
 136 our coarse vertical resolution, the helium injection sites typically fall well
 137 within the ocean interior. The upward shifted source pattern in the sum of
 138 (2) falls outside the ocean domain at only one grid point, and the downward-
 139 shifted source pattern falls outside the domain for only 20% of the grid points
 140 containing ^3He sources. Source points that fall outside the ocean domain are
 141 discarded.

142 The ^4He and ^3He sources are assumed to be proportional so that

$$143 \quad {}^4S = {}^3S/R_m \quad , \quad (3)$$

144 with the mantle isotope ratio $R_m = 8R_a$, where $R_a = 1.384 \times 10^{-6}$ is the
 145 reference $^3\text{He}/^4\text{He}$ isotope ratio of the atmosphere. The ratio R_m/R_a is as-
 146 sumed to be globally uniform although there is evidence that R_m/R_a varies
 147 regionally between 7 and 11 (*Graham et al.*, 1996; *Kurz et al.*, 2005), with
 148 even higher values reported for atypical basalts (*Lupton et al.*, 2012). While
 149 we have insufficient information to vary R_m/R_a geographically in our model,
 150 we varied the globally uniform value of R_m/R_a from 6 to 11. The optimized
 151 fit to the observed $\delta^3\text{He}$ distribution is insensitive to the precise value of
 152 R_m/R_a : Reducing R_m/R_a from 8 to 6 reduces the global RMS relative error
 153 by only 0.1% and increases the optimal global ^3He source by 6%, while in-
 154 creasing R_m/R_a from 8 to 11 increases the global RMS relative error by only
 155 0.1% and decreases the optimal global ^3He source by 4%. The insensitivity

of the mantle ^3He source to the precise value of R_m/R_a can also be derived analytically from the definition of $\delta^3\text{He}$ and equation (3), which imply that in order to keep $\delta^3\text{He}$ close to the observations, the fractional change in the mantle source is approximately given by the fractional change in R_m/R_a multiplied by $(1 - R_m/R_a)^{-1}$. We therefore adopted the ratio $R_m/R_a = 8$ of the OCMIP protocol, which is deemed to be characteristic of ridge basalts and hydrothermal vents (*Lupton and Craig, 1975; Hilton and Porcelli, 2014*).

Equation (1) is solved using standard matrix methods for the ^3He and ^4He concentrations from which the relative isotope ratio, in units of percent, is calculated as

$$\delta^3\text{He} = 100 \left(\frac{1}{R_a} \frac{{}^3\chi}{{}^4\chi} - 1 \right) . \quad (4)$$

The source parameters $\sigma_{\Omega,k'}$ are determined by minimizing a cost function for the mismatch between modelled and observed $\delta^3\text{He}$ subject to the constraints that $\sigma_{\Omega,k'} \geq 0$. The cost function was constructed as follows: The weight $w(\mathbf{r})$ for grid point $\mathbf{r}$ is given by $w(\mathbf{r}) = \mu(\mathbf{r})V(\mathbf{r})$, where $V(\mathbf{r})$ is the fractional ocean volume of the grid box at $\mathbf{r}$ and $\mu(\mathbf{r})$ is a mask that is unity for points below 1000 m where the mean age (available for steady flow by direct matrix inversion) is older than 100 years, and $\mu(\mathbf{r})$ is zero otherwise. The purpose of $\mu(\mathbf{r})$ is to minimize the influence of tritogenic ^3He . The local model-observation mismatch is $x(\mathbf{r}) = \delta^3\text{He}_{\text{mod}}(S) - \delta^3\text{He}_{\text{obs}}$, where the model's $\delta^3\text{He}$ depends on the source S whose parameters are to be optimized. Organizing the grid values of x and w into column vectors $\mathbf{x}$ and $\mathbf{w}$, the cost function that is minimized is given by

$$E = \mathbf{x}^T \mathbf{W} \mathbf{x} , \quad (5)$$

where $\mathbf{W} = \mathbf{diag}(\mathbf{w})$ is a diagonal matrix with the weights $\mathbf{w}$ along the diagonal.

For the observed $\delta^3\text{He}_{\text{obs}}$, we used the $1^\circ \times 1^\circ$ gridded field of *Bianchi et al.* (2010) for convenience and because this field has been corrected to account for proper inter-calibration of the various underlying sets of measurements. We acknowledge, however, that the objective analysis that extrapolates the original bottle data to the grid introduces significant smoothing and hence its own biases, but we deem the gridded data adequate for our purposes here. (The gridded observations were interpolated to our numerical grids for use in the cost function.)

3. Results

We optimized the source parameters independently for each of the 11 data-assimilated circulations. When we do not present individual results

for each circulation, we present the mean over the ensemble of 10 B-grid circulations. (We did not include the single C-grid circulation in the mean because it lacks the more realistic rotated diffusivity tensor and would have only minor influence on the mean.)

3.1. Optimized sources

We use the six regions shown in Figure 1a, with the following OCMIP-source-weighted mean ^3He injection depths: The Atlantic (ATL, 2.4 km), Southern Ocean (SO, 2.5 km), Indian Ocean (IND, 2.2 km), Western Pacific (WP, 1.5 km), south-eastern Pacific (SEP, 2.6 km), and north-eastern Pacific (NEP, 2.4 km). We find that partitioning the source regions more finely than this is problematic. For example, if the WP region is split into a northern and southern part, the source of the northern part optimizes to zero, while the source of the southern part, which is dominated by the Lau Backarc Basin near Fiji, optimizes to unrealistically high values of nearly a third of the global source. We also find that if the Juan DeFuca ridge system is assigned its own region, its source similarly optimizes to zero. A zero source for the northern WP or for the Juan DeFuca system is unphysical, given that ^3He plumes are observed to emanate from the ridge systems of these regions. Such artifacts resulting from a finer source partitioning are due to a combination of circulation biases and inaccuracies in the source specification. Specifically, we find that the WP source is optimally higher than its OCMIP value, which suggests missing sources. But when the southern part of the WP patch is allowed to vary freely, the southern source is increased yet further as this mimics the strong westward propagating helium plume from the EPR that is under-represented in our circulations. To avoid artifacts, we considered not partitioning the ridge system of the Pacific basin, but we find that then $\delta^3\text{He}$ in the entire western Pacific is underestimated (likely because of missing sources), while the eastern North Pacific retains a high bias in $\delta^3\text{He}$ that we see in the optimal solutions for all source partitionings and circulations considered. (We will compare optimal solutions and observations in detail below.) We therefore use the six patches defined in Figure 1a as a reasonable compromise between a robust source distribution and flexibility to adjust the sources of the OCMIP specification.

Figure 1b shows the optimized ^3He source per unit area with a logarithmic color scale. The dominance of the Pacific sources is evident, with the largest local sources being associated with the ridge systems of the Southwest Pacific near Fiji (including the Lau Backarc Basin), the Southeast Pacific along the East Pacific Rise (EPR), and the Pacific sector of the Southern Ocean.

Figure 2 plots the integrated source strengths of the tectonic ridge patches for each circulation and compares them to the corresponding OCMIP values.

232 There are key differences between the OCMIP sources and those inferred for
 233 the B-grid circulations, as well as between the B- and C-grid cases. Except
 234 for the Western Pacific (WP), the OCMIP source strengths exceed the B-grid
 235 estimates for all regions. The global C-grid source strength is 660 mol yr^{-1} ,
 236 while the B-grid source strengths range from 760 to 850 mol yr^{-1} , with mean
 237 and standard deviation of $800 \pm 25 \text{ mol yr}^{-1}$ compared to 1000 mol yr^{-1} for
 238 the OCMIP source. For the WP region, however, the B-grid sources are on
 239 average more than double the OCMIP sources.

240 To quantify the uncertainty associated with partitioning the Pacific sources
 241 into three independent regions, we also optimized the case of a single Pacific
 242 region (distributed within the Pacific according to the OCMIP). While this
 243 results in a Pacific source that is reduced by 13–35% with a pronounced low
 244 bias in the western South Pacific, the effect on the global source strength
 245 is more modest: The C-grid global source is 640 mol yr^{-1} , while the B-grid
 246 global source ranges from 720 to 780 mol yr^{-1} ($750 \pm 17 \text{ mol yr}^{-1}$), with 2–
 247 10% reductions compared to the corresponding optimal source partitioned as
 248 in Figure 1b.

249 The difference between the B- and C-grid cases is most pronounced for
 250 the Southern Ocean. This is a region of significant baroclinicity with strongly
 251 sloping isopycnals, and the difference between the circulations is likely dom-
 252 inated by their different eddy-diffusion parameterizations: the B-grid trans-
 253 port has a diffusivity tensor that is aligned with observed isopycnals (*Redi*,
 254 1982), while the C-grid transport uses fixed horizontal and vertical diffu-
 255 sivities. Because the B-grid diffusivity tensor is aligned with the sloping
 256 isopycnals of the Southern Ocean, the Southern Ocean is ventilated more
 257 vigorously, requiring a Southern Ocean mantle helium source that is ~ 100
 258 mol yr^{-1} larger in order to match $\delta^3\text{He}$ observations in this region (Figure
 259 2).

260 Figure 3 shows the effect of allowing flexibility in the injection depth
 261 by plotting the horizontally integrated sources per unit depth of each basin
 262 for both the OCMIP and optimized sources. The vertical profiles of the
 263 optimized sources all have their deep maximum $\sim 300 \text{ m}$ (one grid box) higher
 264 than the OCMIP sources. The shallow sources in the Pacific are shifted up
 265 by one grid box, and because the shallower box is also thinner, the source
 266 per unit depth develops a more pronounced spike. These results suggest
 267 that the coarse vertical resolution of the deep ocean typical of all circulation
 268 models is a potential impediment for the accurate modelling of ^3He and that
 269 the globally uniform 300-m vertical offset for ^3He injection assumed in the
 270 OCMIP protocol may not be realistic. Allowing flexibility in the injection
 271 depth improved the volume-weighted $\delta^3\text{He}$ RMS error from 21% to 15% of
 272 the global mean.

273 3.2. Fidelity to observed $\delta^3\text{He}$

274 We now compare the ensemble-mean $\delta^3\text{He}$ field from the optimized sources
 275 with the gridded observations. Figure 4 shows the volume-weighted joint
 276 probability density function (pdf) of the modeled and observed $\delta^3\text{He}$ fields,
 277 calculated using a kernel-density estimator. (The joint pdf is simply the con-
 278 tinuum limit of a volume-weighted histogram of the model-versus-observations
 279 scatter plot.) The model’s $\delta^3\text{He}$ is generally within 15% (relative error) of the
 280 observed values, but there are some systematic deviations from a perfect 1:1
 281 fit. The pdf can roughly be divided into three clusters as indicated on Figure
 282 4. For the North Pacific cluster, the models over-estimate the observed $\delta^3\text{He}$
 283 by as much as 4% (absolute $\delta^3\text{He}$ units). For the Southern Ocean cluster, the
 284 model systematically underestimates the observations by up to 2% (absolute
 285 $\delta^3\text{He}$ units). We note that some of the deep water in the Southern Ocean
 286 has a mean age less than 100 years, so that it is possible that the influence
 287 of tritiogenic ^3He there has not been completely masked out.

288 The vertical structure of the model fits to the data is summarized in the
 289 large-scale horizontally averaged $\delta^3\text{He}$ vertical profiles shown in Figure 5.
 290 Grid points where the mean water age is younger than 100 years or where
 291 the gridded data has missing values have been excluded from the averages to
 292 make the comparison as quantitative as possible and consistent with Figure
 293 4 that was masked in the same manner. Overall, Figure 5 shows an excellent
 294 fit of the modeled $\delta^3\text{He}$ to the observations. This demonstrates the utility of
 295 assimilating ventilation and water-mass tracers (here temperature, salinity,
 296 CFCs, and radiocarbon) to constrain the deep ocean circulation. These pro-
 297 files can be compared, for example, to the $\delta^3\text{He}$ distribution in the GFDL
 298 ESM2Mc model (*Gnanadesikan et al.*, 2015, Fig. 4d). The ESM2Mc model
 299 has a similar resolution as our model, but is not data-assimilated, and shows
 300 large discrepancies between modeled and observed global-mean $\delta^3\text{He}$ profiles
 301 (on the order of 4-5% in absolute $\delta^3\text{He}$ units).

302 Despite the overall excellent fit to observed $\delta^3\text{He}$, the regions of system-
 303 atic deviation between model and observations identified in Figure 4 also
 304 manifest in the basin-mean profiles. All of the circulations overpredict $\delta^3\text{He}$
 305 in the North Pacific between ~ 2.5 and 1 km depth. The abyssal North Pa-
 306 cific has the correct vertical gradients but is slightly underpredicted. The
 307 underprediction in the Southern Ocean is manifest in the South Pacific pro-
 308 file, which falls outside the ensemble spread above ~ 1800 m. The $\delta^3\text{He}$ in the
 309 Indian Ocean below ~ 3000 m is slightly underpredicted, while in the South
 310 Atlantic $\delta^3\text{He}$ is fit very well throughout the water column. The prominent
 311 discrepancy in the North Atlantic at small $\delta^3\text{He}$ values is due to tritiogenic
 312 ^3He , which is not completely eliminated by masking out water with a mean

age younger than 100 years because waters with older mean age can still contain a substantial fraction of water younger than 50 years.

Figure 6 compares the zonally averaged ensemble-mean $\delta^3\text{He}$ fields for each basin to the corresponding fields of the gridded observations. The ensemble mean of the optimized model solutions captures the primary (and much of the secondary) features of the observations quite well, but the over-prediction in the North Pacific is evident at mid depths (~ 2000 m). This could indicate that the deep North Pacific is underventilated in the OCIM, although further research is needed to separate the influence of circulation and the effects of possibly inaccurate representation of the ^3He source patterns. We also note that tritiogenic ^3He contributions to the observed $\delta^3\text{He}$ values could cause our mantle-source estimates to be too large, and in steady state the resulting excess $\delta^3\text{He}$ in the model would preferentially reside in the deep North Pacific, causing a high bias.

Figure 7 compares the modelled and observed geographical distribution of $\delta^3\text{He}$ at 2141 m depth. The overall patterns match very well, demonstrating that the large-scale abyssal circulation is well captured by the data-assimilated model. In particular, our estimates display a local saddle point near the equator in the vicinity of the EPR (Figure 7) that can also be seen in the observed $\delta^3\text{He}$ field. This structure is consistent with geostrophic shear in the deep equatorial circulation (*Reid, 1997*) that has an eastward “jet” along the equator with westward flow to the north and south. This feature of the $\delta^3\text{He}$ field is not captured by coarse-resolution forward ocean models that have been used to simulate $\delta^3\text{He}$ (*Dutay et al., 2004; Bianchi et al., 2010; Gnanadesikan et al., 2015*).

The previously discussed high bias in modeled $\delta^3\text{He}$ in the North Pacific is seen to be located primarily in the western North Pacific, while the low bias near Antarctica in the Southern Ocean is primarily in the Atlantic and Indian sectors. The Northwest Pacific high bias could be an artifact of the source distribution being incorrect within our WP patch, for which the optimization infers mantle ^3He fluxes much larger than the original OCMIP source (Figure 2). Much of this difference is presumably due to OCMIP missing sources in the Lau Backarc Basin, which accounts for about half of the WP sources. The Lau Backarc Basin has only recently been observed to be hydrothermally very active (*Beaulieu et al., 2015; Lupton et al., 2012*), and because of its particularly complex tectonic structure (e.g., *Zellmer and Taylor, 2001*), the model of *DeMets et al. (1990)* used to reconstruct the OCMIP helium sources likely underestimated spreading rates in this region. *Schlitzer (2016)* also found that the source of the Lau Backarc Basin needed to be increased to match the $\delta^3\text{He}$ observations there. However, because our WP patch includes ridge systems in both the South and North Pacific, the

354 optimized mantle ^3He sources are increased in both basins. As mentioned
 355 in Section 3.1, separating the WP patch into a northern and southern part
 356 results in a zero northern WP source and an unrealistically large southern
 357 WP source, without alleviating the high bias in the western North Pacific.
 358 Not allowing the WP to vary independently results in a large low bias in
 359 the western South Pacific, suggesting missing sources. Optimizing three in-
 360 dependently varying Pacific regions allowed us to improve the model in the
 361 Southwest Pacific, but may overestimate $\delta^3\text{He}$ in the Northwest Pacific.

362 While our model does a better job of capturing the structure of $\delta^3\text{He}$
 363 in the deep tropical Pacific than previous efforts, the ^3He plume emanating
 364 from the southern EPR has been observed to spread primarily westward
 365 (*Lupton and Craig, 1981; Resing et al., 2015*). This feature is captured by the
 366 gridded $\delta^3\text{He}$ data used here, but our optimal model estimate has too much
 367 ^3He to the east, and too little to the west, of the southern EPR, suggesting
 368 circulation deficiencies in this region. Possibilities could be a recirculation
 369 north and then south to the east of the rise, or inaccuracies in Southern
 370 Ocean ventilation considering that the density surfaces on which much of the
 371 ^3He plume lies outcrop in the Southern Ocean.

372 3.3. Ensemble mean outgassing and inventory

373 The only sink of mantle ^3He is outgassing to the atmosphere, which we
 374 calculate using our data-constrained helium model. We distinguish mantle
 375 helium from atmospheric helium as follows: All helium injected by mantle
 376 sources is labeled as mantle helium of concentration χ_m , but mantle helium
 377 loses its identity as soon as it outgases to the atmosphere. In other words,
 378 all helium that enters the ocean through air-sea exchange is labeled as at-
 379 mospheric helium with concentration χ_a , regardless whether it ultimately
 380 originated in the earth’s mantle or is of other origin. Thus, for each isotope
 381 ($\chi = ^3\chi$ or $^4\chi$) we write $\chi = \chi_m + \chi_a$, where

$$382 \quad \mathbf{T} \chi_m = - \mathbf{K} \chi_m + S \quad , \quad (6)$$

$$383 \quad \mathbf{T} \chi_a = \mathbf{K} (\chi_{\text{sat}} - \chi_a) \quad . \quad (7)$$

384 Note that (6) and (7) sum to equation (1) as they must.

385 Figure 8 shows the column inventory of mantle ^3He in the ocean ($^3\chi_m$
 386 vertically integrated) for both the ensemble mean inverse-model estimate and
 387 for the observation-based estimate of *Bianchi et al. (2010)*. Grid points above
 388 1000 m depth or where the ideal mean age is less than 100 years have been
 389 masked out before integrating to minimize the influence of tritogenic ^3He .
 390 The patterns of the column inventories are shaped by the circulation, the
 391 mantle helium source, and the bottom topography. The inverse-model and

392 observational estimates agree well in the Atlantic, Indian Ocean, and South
 393 Pacific in both pattern and magnitude. The bulk of the mantle ^3He can be
 394 seen to reside in the North Pacific for both model and observations. However,
 395 the model estimate is roughly 10% higher in the subpolar North Pacific and
 396 10% lower in the tropical North Pacific than in the observational estimate.
 397 This is consistent with the model’s overpredicted $\delta^3\text{He}$ in the North Pacific,
 398 although the uncertainty of the “observed” mantle ^3He column inventory is
 399 probably larger than the model-data misfit, given that the global observation-
 400 based ^3He inventory has an uncertainty of 15% (*Bianchi et al.*, 2010). In the
 401 Southern Ocean and South Atlantic, the inverse model has roughly 10–20%
 402 lower column burdens than the observational estimate. To the extent that
 403 the likely substantial uncertainties in the observational column burden can
 404 be ignored, this suggests that either our estimated Southern Ocean source
 405 is too small or that Southern Ocean ventilation is too vigorous in the data-
 406 assimilated circulations. The B-grid ensemble-mean ^3He inventory is 414 ± 6
 407 kmol (the C-grid value is 411 kmol), well within the estimated observational
 408 range of 390 ± 60 kmol. This is re-assuring, given that we minimized the local
 409 misfit in $\delta^3\text{He}$ and not, like *Bianchi et al.* (2010), the mismatch between
 410 global inventories. The bulk ^3He residence time (inventory/global source)
 411 implied by our estimates is ~ 500 years, which is shorter than the ~ 700 year
 412 residence time implied by the estimate of *Bianchi et al.* (2010) as expected
 413 given our larger global source.

414 We note that the ^3He column inventory has a striking resemblance to
 415 the Deep-North-Pacific (DNP) pattern of the ocean’s surface-to-surface path
 416 density (*Holzer and Primeau*, 2006, 2008). The DNP pattern is the pattern of
 417 the concentration of water whose surface-to-surface transit time approaches
 418 the longest timescales in the ocean, that is, water that was last ventilated
 419 order 1000 years ago and that will not be ventilated again for order 1000
 420 years. The deep North Pacific is therefore where mantle helium will accu-
 421 mulate most. Because the North Pacific does not have the strongest ^3He
 422 sources, this suggests that the highest concentrations of mantle ^3He are the
 423 result of lateral transport into the deep North Pacific.

424 Figure 9 shows the ocean-to-atmosphere one-way flux $\mathbf{K}^3\chi_m$ with which
 425 mantle ^3He outgases from the ocean, computed from the ensemble mean
 426 source and the control circulation that was perturbed to form the ensemble.
 427 Note that this flux is plotted with a logarithmic color scale to accentuate its
 428 pattern. Mantle ^3He outgases most strongly in the Southern Ocean and in the
 429 North Pacific, indicating that mantle helium is a good tracer for constrain-
 430 ing the deep-to-surface pathways and transport to these important surface
 431 regions. We find that 69% of the mantle ^3He outgases in the Southern Ocean
 432 (south of 40°S), 18% outgases in the extratropical North Pacific, and 8%

433 outgases in the tropical Pacific.

434 4. Discussion and Conclusion

435 We have used an ensemble of data-assimilated ocean circulations and a
436 simple representation of the mantle helium sources whose parameters were
437 determined through objective optimization. Our inversions for the global
438 ^3He mantle source range from 640 to 850 mol ^3He yr $^{-1}$. Our ^3He source
439 estimates fall between the higher estimates of 1100 ± 300 mol yr $^{-1}$ by *Clarke*
440 *et al.* (1969) and *Craig et al.* (1975), the 1000 mol yr $^{-1}$ estimates of *Farley*
441 *et al.* (1995) and *Cartigny et al.* (2008), and the lower estimates of 267–
442 534 mol yr $^{-1}$ by *Jean-Baptiste* (1992) based on box models, the 422 ± 181
443 mol yr $^{-1}$ estimate by *Saal et al.* (2002) based on mid-ocean-ridge basalt
444 geochemistry, the 527 ± 102 mol yr $^{-1}$ estimate by *Bianchi et al.* (2010) already
445 described, and a very recent estimate by *Schlitzer*¹ (2016) of 450 ± 50 mol
446 yr $^{-1}$. *Schlitzer*’s approach was methodologically very similar to ours: he
447 used the Alfred Wegner Institute Adjoint Tracer Model [AATM, *Schlitzer*
448 (2007)], fitted to hydrographic data including radiocarbon and CFCs but
449 not helium, to constrain the global amplitude of the OCMIP mantle helium
450 source by minimizing the quadratic mismatch with available $\delta^3\text{He}$ data in
451 the Pacific below 1500 m depth. These mantle ^3He source estimates from
452 the literature are also collected in Table 1, together with the estimates of
453 this work. The large range of the estimates in Table 1 shows that the true
454 uncertainty in our knowledge of the mantle helium sources is larger than the
455 ad-hoc methodological uncertainties of the individual studies (e.g., $\pm 2.5\%$
456 variation in $\delta^3\text{He}$ mismatch for *Schlitzer* (2016); ensemble spread for this
457 study). Even more importantly, the differences among the various estimates
458 underscore the importance of both the realism of the circulation and the
459 specification of the mantle sources.

460 The spatial distribution of sources is critically important to the inferred
461 mantle ^3He flux. Specifically, if we only optimize the global amplitude of the
462 OCMIP source pattern, as was done by *Bianchi et al.* (2010) and *Schlitzer*
463 (2016), we obtain a ^3He source of 676 ± 15 mol yr $^{-1}$ (620 mol yr $^{-1}$ for the
464 C-grid circulation) with an RMS $\delta^3\text{He}$ misfit of 3.0% $\delta^3\text{He}$ units (21% of the
465 mean). This is almost within the range estimated by *Bianchi et al.* (2010),
466 but still higher than that of *Schlitzer* (2016), with differences that must be
467 ascribed to differences in circulation and possibly spatial resolution. When
468 the OCMIP source is partitioned into 6 regions whose source strengths are

¹We became aware of this work after our manuscript was submitted.

469 optimized (keeping the original OCMIP injection depths), our B-grid source
 470 estimate increases to 751 ± 17 mol yr⁻¹, and the RMS misfit is reduced
 471 to 2.6% $\delta^3\text{He}$ units (19% of the mean). This increased source results from
 472 a regional redistribution, which roughly triples the source in the western
 473 Pacific (WP) at the expense of the eastern-Pacific and Indian Ocean sources.
 474 This redistribution allows ^3He to outgas to the atmosphere at a higher rate
 475 because the WP source is the least deep with a 1.5-km mean depth. To
 476 maintain $\delta^3\text{He}$ at the observational levels, a larger source is necessary. When
 477 we additionally allow flexibility in the vertical injection depths, the optimized
 478 sources move slightly upward in the water column (Figure 3), which further
 479 increases the outgassing rate and results in a yet larger source of 800 ± 25
 480 mol yr⁻¹ for our B-grid circulations. These systematic adjustments to the
 481 mantle helium sources do lead to an improved $\delta^3\text{He}$ fit with an RMS error of
 482 2.1% $\delta^3\text{He}$ units (15% of the mean).

483 Southern Ocean ventilation, particularly as driven by eddy diffusion along
 484 sloping isopycnals, is another critically important factor in the inference of
 485 mantle helium sources. The optimized Southern Ocean ^3He source of the C-
 486 grid model without a rotated diffusivity tensor is ~ 100 mol yr⁻¹ smaller than
 487 that of our B-grid model, which does have a rotated diffusivity tensor. Fur-
 488 thermore, the B-grid circulation with the largest isopycnal diffusivity (2000
 489 m² s⁻¹) results in the largest Southern Ocean mantle ^3He source (Figure 2).
 490 This shows that $\delta^3\text{He}$ provides an important constraint on the parameteriza-
 491 tion of eddy diffusion, particularly in the Southern Ocean. The importance
 492 of Southern Ocean $\delta^3\text{He}$ as a constraint on the mantle ^3He sources can also
 493 be seen by comparing the $\delta^3\text{He}$ field produced by our optimal sources to
 494 that of *Schlitzer* (2016). At 1650 m depth, our B-grid circulation underesti-
 495 mates $\delta^3\text{He}$ by $\sim 2.5\%$ ($\delta^3\text{He}$ units), possibly indicating too vigorous Southern
 496 Ocean ventilation. In the same region, the model of *Schlitzer* (2016) *over-*
 497 *estimates* Southern Ocean $\delta^3\text{He}$ by $\sim 5\%$, perhaps indicating too sluggish
 498 Southern Ocean ventilation. These differences in the Southern Ocean, where
 499 most deep waters upwell, likely account for much of the ~ 300 mol yr⁻¹ differ-
 500 ence in the global mantle ^3He source between our model and that of *Schlitzer*
 501 (2016), and point to the utility of $\delta^3\text{He}$ as a constraint on Southern Ocean
 502 ventilation rates.

503 The OCMIP helium sources and our optimized sources have qualitatively
 504 similar distributions of the regional source strength, but there are important
 505 quantitative differences even when the OCMIP sources are scaled to the
 506 optimized global mean source. In particular, the OCMIP sources of the NE
 507 Pacific and Indian Ocean are ~ 3 times larger than the optimized sources, and
 508 the OCMIP western Pacific sources are ~ 3 times smaller than the optimized
 509 sources. On one hand, these differences are likely due to incorrect OCMIP

source strengths (e.g., missing sources in the Lau Backarc Basin), or possibly due to an invalid hypothesis that the source is uniformly proportional to spreading rate with the same factor across the ridge system. On the other hand, the differences likely also reflect systematic biases of the circulations or imperfections in the assimilated data such as over-smoothed gridded $\delta^3\text{He}$ or inclusion of observations influenced by tritogenic ^3He . A combination of all issues is probably at play.

The optimized mantle ^3He sources were higher in the water column than the original OCMIP sources. This is consistent with the findings of *Gnanadesikan et al.* (2015) who simulated $\delta^3\text{He}$ in an ensemble of models using the OCMIP injection depths and found that all models produced a $\delta^3\text{He}$ profile with a maximum ~ 500 m deeper than observed. This suggests that hydrothermal plumes rise higher than 300 m above the vent site, and that the assumption of a globally uniform vertical offset of the injection depth assumed in the OCMIP protocol may need to be relaxed.

While the North Pacific does not have the strongest ^3He sources, it is a holding pen for old waters with the ocean's longest interior-to-surface mean transit times. The pattern of the vertically integrated ^3He closely resembles the Deep-North-Pacific pattern of the path density for millennial residence times (*Holzer and Primeau*, 2006, 2008). This suggests that the highest concentrations of mantle ^3He are the result of lateral transport into the deep North Pacific where it takes on the order of a millennium for the advective-diffusive ocean circulation to bring the mantle helium to the surface where it can outgas to the atmosphere.

Our analysis suggests that ^3He has strong potential to complement the constraints provided by radiocarbon on the ocean's deep circulation. The data-assimilated circulations used here are constrained by CFCs and radiocarbon (in addition to temperature and salinity), and provide a good match the observed $\delta^3\text{He}$ after optimization of the mantle helium sources. Comparison with the $\delta^3\text{He}$ observations demonstrated that our circulations have greater fidelity to the ocean's deep circulation than is the case for other models. Specifically, the structure of $\delta^3\text{He}$ in the deep tropical Pacific is captured much better than in previous studies, underlining the utility of mantle helium as a tracer of the deep circulation. Nevertheless, biases in the modeled $\delta^3\text{He}$ distributions remain. In particular, the mid-depth North Pacific contains too much mantle helium, and the helium plume emanating from the southern EPR is too strong east of the rise, and too weak west of the rise, compared to observations. These findings warrant future objective assimilations of the $\delta^3\text{He}$ observations to constrain the features of the deep circulation that appear to be not well constrained by the tracers already assimilated.

The features of the $\delta^3\text{He}$ distribution in the vicinity of the EPR that are

not captured by the tracers already assimilated may be due to an anomalous circulation driven by geothermal heating associated with the hydrothermal vents that inject mantle helium (*Stommel, 1982; Adcroft et al., 2001*). Such a circulation can be argued to produce vertically stacked counter-rotating horizontal gyres (*Stommel, 1982*) that are unlikely to strongly imprint on radiocarbon. This circulation is, however, important to the distribution of mantle helium (*Dutay et al., 2010*), and associated elements with hydrothermal sources such as iron and other trace metals (*Resing et al., 2015; Fitzsimmons et al., 2014; Sander and Koschinsky, 2011*). Assimilating $\delta^3\text{He}$ into the OCIM would allow the model to capture the transport due to the geothermally driven circulation associated with hydrothermal vents. Because the OCIM adjusts its circulation to match the imposed tracer constraints, this can be done without modelling a geothermal heat source.

The inferred ^3He sources of the Southern Ocean were particularly sensitive to how eddy diffusion is parameterized, with $\sim 30\%$ difference in source strength between the C-grid circulation using a Cartesian diffusivity tensor and the B-grid circulations employing a rotated diffusivity tensor. Both the B- and C-grid circulations have globally uniform eddy-diffusion coefficients. The addition of an eddy-induced bolus velocity as in the GM scheme (*Gent and McWilliams, 1990*) is not necessary because of the OCIM's adjustable velocity field, and because the OCIM imposes mixing along observed isopycnals diagnosed from climatological temperature and salinity. However, if the background diffusivities in the OCIM are overestimated, our inferred mantle ^3He sources may be biased high. Both observations and models indicate that the isopycnal eddy diffusivity in the Southern Ocean is highly variable spatially (*Griesel et al., 2015*) and in many regions significantly lower than the $1000 \text{ m}^2 \text{ s}^{-1}$ used in most versions of the OCIM. Therefore, future data assimilations may benefit from optimizing a locally varying diffusivity field, and our results suggest that $\delta^3\text{He}$ would provide an important constraint.

Our findings here will inform several additional aspects of future research on using $\delta^3\text{He}$ to constrain the ocean's deep circulation. It will be necessary to use a source parameterization that is more detailed and flexible than the OCMIP source to reduce biases due to errors in the sources. When jointly optimizing the circulation and source parameters, it should ultimately be possible to estimate an optimal mantle helium source at every model grid point above the tectonic ridge system. The sensitivity of the inferred mantle ^3He sources to the helium injection depth also suggests that increased vertical resolution would be beneficial. We plan to explore the degree to which using $\delta^3\text{He}$ as a constraint will improve the abyssal circulation of the OCIM in a future data-assimilation study.

591 **5. Acknowledgments**

592 We acknowledge funding from ARC grant DP120100674 (MH) and NASA
593 grant NNX16A122G (TD). We thank François Primeau for discussions and
594 for providing the data-assimilated C-grid transport operator. The gridded
595 $\delta^3\text{He}$ observations used here are available at the following web address:
596 www.eps.mcgill.ca/~dbianchi/Sites/personal_page/Oceanic_Helium.html

597 Adcroft, A., J. R. Scott, and J. Marotzke, Impact of Geothermal Heating on
598 the Global Ocean Circulation, *Geophys. Res. Lett.*, *28*, 1735–1738, 2001.

599 Beaulieu, S. E., E. T. Baker, and C. R. German, Where are the undiscovered
600 hydrothermal vents on oceanic spreading ridges?, *Deep-Sea Research II*,
601 *121*, 202–212, 2015.

602 Bianchi, D., J. L. Sarmiento, A. Gnanadesikan, R. M. Key, P. Schlosser, and
603 R. Newton, Low helium flux from the mantle inferred from simulations of
604 oceanic helium isotope data, *Earth Planet. Sci. Lett.*, *297*, 379–386, 2010.

605 Cartigny, P., F. Pineau, C. Aubaud, and M. Javoy, Towards a consistent
606 mantle carbon flux estimate: Insights from volatile systematics ($\text{H}_2\text{O}/\text{Ce}$,
607 δD , CO_2/Nb) in the North Atlantic mantle (14°N and 34°N), *Earth Planet.*
608 *Sci. Lett.*, *265*, 672–685, 2008.

609 Clarke, W., M. Beg, and H. Craig, Excess ^3He in the sea: evidence for
610 terrestrial primordial helium, *Earth Planet. Sci. Lett.*, *6*, 213–220, 1969.

611 Craig, H., W. Clarke, and M. Beg, Excess ^3He in deep water on the East
612 Pacific Rise, *Earth Planet. Sci. Lett.*, *26*, 125–132, 1975.

613 DeMets, C., R. Gordon, D. Argus, and S. Stein, Current plate motions,
614 *Geophys. J. Int.*, *101*, 425–478, 1990.

615 DeVries, T., The oceanic anthropogenic CO_2 sink: Storage, air-sea fluxes,
616 and transports over the industrial era, *Global Biogeochem. Cycles*, *28*,
617 631–647, 2014.

618 DeVries, T., and F. Primeau, Dynamically and observationally constrained
619 estimates of water-mass distributions and ages in the global ocean, *J. Phys.*
620 *Oceanogr.*, *41*, 2381–2401, 2011.

621 Dutay, J.-C., P. Jean-Baptiste, J.-M. Campin, A. Ishida, E. Maier-Reimer,
622 R. Matear, A. Mouchet, I. Totterdell, Y. Yamanaka, K. Rodgers,
623 G. Madec, and J. Orr, Evaluation of OCMIP-2 ocean models’ deep cir-
624 culation with mantle helium-3, *J. Mar. Syst.*, *48*, 15–36, 2004.

625 Dutay, J.-C., J. Emile-Geay, D. Iudicone, P. Jean-Baptiste, G. Madec, and
626 C. Carouge, Helium isotopic constraints on simulated ocean circulations:
627 implications for abyssal theories, *Environ. Fluid Mech.*, *10*, 257–273, 2010.

628 Farley, K., E. Maier-Reimer, P. Schlosser, and W. Broecker, Constraints on
629 mantle He-3 fluxes and deep-sea circulation from an ocean general cir-
630 culation model, *J. Geophys. Res.*, *100*, 3829–3839, 1995.

- 631 Fitzsimmons, J. N., E. A. Boyle, and W. J. Jenkins, Distal transport of
632 dissolved hydrothermal iron in the deep South Pacific Ocean, *Proc. Natl.*
633 *Acad. Sci. USA*, *111*, 16,654–16,661, 2014.
- 634 Garabato, A. C. N., D. P. Stevens, A. J. Watson, and W. Roether, Short-
635 circuiting of the overturning circulation in the Antarctic Circumpolar Cur-
636 rent, *Nature*, *447*, 194–197, 2007.
- 637 Gent, P. R., and J. C. McWilliams, Isopycnal mixing in ocean circulation
638 models, *J. Phys. Oceanogr.*, *20*, 150–155, 1990.
- 639 Gnanadesikan, A., M.-A. Pradal, and R. Abernathey, Exploring the isopycnal
640 mixing and helium–heat paradoxes in a suite of earth system models, *Ocean*
641 *Sci.*, *11*, 591–605, 2015.
- 642 Graham, D., P. Castillo, J. E. Lupton, and R. Batiza, Correlated He and Sr
643 isotope ratios in South Atlantic near-ridge seamounts and implications for
644 mantle dynamics, *Earth Planet. Sci. Lett.*, *144*, 491–503, 1996.
- 645 Griesel, A., C. Edena, N. Koopmanna, and E. Yulaeva, Comparing isopyc-
646 nal eddy diffusivities in the Southern Ocean with predictions from linear
647 theory, *Ocean Modelling*, *94*, 33–45, 2015.
- 648 Hilton, D., and D. Porcelli, Noble gases as mantle tracers, in *Treatise in*
649 *Geochemistry*, edited by T. K. K. and H. H. D., vol. 3, 2 ed., pp. 327–353,
650 Elsevier Science, Oxford, 2014.
- 651 Holzer, M., and F. W. Primeau, The diffusive ocean conveyor, *Geophys. Res.*
652 *Lett.*, *33*, 2006.
- 653 Holzer, M., and F. W. Primeau, The path-density distribution of oceanic
654 surface-to-surface transport, *J. Geophys. Res.*, *113*, 2008.
- 655 Jean-Baptiste, P., Helium-3 Distribution in the Deep World Ocean, in *Iso-*
656 *topes of Noble Gases as Tracers in Environmental Studies*, edited by
657 K. Rozanski, p. 219–240, International Atomic Energy Agency, Vienna,
658 1992.
- 659 Kurz, M. D., M. Moreira, J. Curtice, D. E. Lott III, J. J. Mahoney, and
660 J. M. Sinton, Correlated helium, neon, and melt production on the super-
661 fast spreading East Pacific Rise near 17°S, *Earth Planet. Sci. Lett.*, *232*,
662 125–142, 2005.
- 663 Lupton, J., Hydrothermal helium plumes in the Pacific Ocean, *J. Geophys.*
664 *Res.*, *103*, 15,853–15,868, 1998.

- 665 Lupton, J., and H. Craig, Excess He-3 in oceanic basalts: evidence for ter-
666 restrial primordial helium, *Earth Planet. Sci. Lett.*, *26*, 133–139, 1975.
- 667 Lupton, J. E., and H. Craig, A Major Helium-3 Source at 15°S on the East
668 Pacific Rise, *Science*, *214*, 13–18, 1981.
- 669 Lupton, J. E., R. J. Arculus, J. Resing, G. J. Massoth, R. R. Greene, L. J.
670 Evans, and N. Buck, Hydrothermal activity in the Northwest Lau Backarc
671 Basin: Evidence from water column measurements, *Geochemistry, Geo-*
672 *physics, Geosystems*, *13*, n/a–n/a, 2012.
- 673 Primeau, F. W., Characterizing transport between the surface mixed layer
674 and the ocean interior with a forward and adjoint global ocean transport
675 model, *J. Phys. Oceanogr.*, *35*, 545–564, 2005.
- 676 Primeau, F. W., and M. Holzer, The ocean’s memory of the atmosphere:
677 Residence-time and ventilation-rate distributions of water masses, *J. Phys.*
678 *Oceanogr.*, *36*, 1439–1456, 2006.
- 679 Primeau, F. W., M. Holzer, and T. DeVries, Southern Ocean nutrient trap-
680 ping and the efficiency of the biological pump, *J. Geophys. Res.*, *118*,
681 2547–2564, 2013.
- 682 Redi, M. H., Oceanic isopycnal mixing by coordinate rotation, *J. Phys.*
683 *Oceanogr.*, *12*, 1154–1158, 1982.
- 684 Reid, J. L., On the total geostrophic circulation of the pacific ocean: flow
685 patterns, tracers, and transports, *Prog. Oceanog.*, *39*, 263–352, 1997.
- 686 Resing, J. A., P. N. Sedwick, C. R. German, W. J. Jenkins, J. W. Moffett,
687 B. M. Sohst, and A. Tagliabue, Basin-scale transport of hydrothermal
688 dissolved metals across the South Pacific Ocean, *Nature*, *523*, 200–203,
689 2015.
- 690 Saal, A. E., E. H. Hauri, C. H. Langmuir, and M. R. Perfit, Isotopic compo-
691 sitions of the elements: Technical report, *Nature*, *419*, 451–455, 2002.
- 692 Sander, S. G., and A. Koschinsky, Metal flux from hydrothermal vents in-
693 creased by organic complexation, *Nature Geosci.*, *4*, 145–150, 2011.
- 694 Schlitzer, R., Assimilation of Radiocarbon and Chlorofluorocarbon Data to
695 Constrain Deep and Bottom Water Transports in the World Ocean, *J.*
696 *Phys. Oceanogr.*, *37*, 259–276, 2007.

- 697 Schlitzer, R., Quantifying He fluxes from the mantle using multi-tracer data
698 assimilation, *Phil. Trans. R. Soc. A. (in press)*, ?, ?, 2016.
- 699 Stommel, H., Is the South Pacific helium-3 plume dynamically active, *Earth*
700 *Planet. Sci. Lett.*, *61*, 63–67, 1982.
- 701 Zellmer, K. E., and B. Taylor, A three-plate kinematic model for Lau Basin
702 opening, *Geochemistry, Geophysics, Geosystems*, *2*, n/a–n/a, 2001, 1020.

Figure 1: (a) Definition of the 6 regions of the global tectonic ridge system. The mantle ^3He source strength for each region was optimized. (b) The B-grid ensemble mean optimized source of mantle ^3He . Note the logarithmic color scale. The horizontal structure of the source within each region is based on ridge spreading rates taken from the OCMIP protocol.

Figure 2: Optimized mantle ^3He source strengths for the 6 regions considered and for the global (GLB) total as estimated from 10 data-assimilated B-grid and one C-grid circulation. The points with error bars show the mean and standard deviation of the B-grid source strengths for each region. For comparison, we also show the regional source strengths of the OCMIP source (red crosses) and of the OCMIP source scaled to the B-grid ensemble global mean (red squares) so that the redistribution of the optimized sources relative to the OCMIP source can be more easily discerned. To give an indication of the uncertainty in the global source due to the regional partitioning, the results for the case of an unpartitioned Pacific source (4 regions globally) are plotted in open symbols in the GLB panel.

Figure 3: Optimized horizontally integrated mantle ^3He source per unit depth in blue compared to the corresponding OCMIP field in red for each basin and for the global (GLB) ocean.

Figure 4: Joint volume-weighted distribution function for the modelled and observed $\delta^3\text{He}$. For the model, the B-grid ensemble-mean $\delta^3\text{He}$ was used. The red dashed lines indicate $\pm 15\%$ deviations from the 1:1 line. Regions of the pdf dominated by the Atlantic (ATL), Southern Ocean (SO), and North Pacific (N PAC) are circled.

Figure 5: Vertical profiles of $\delta^3\text{He}$ horizontally averaged over the North and South Pacific (PAC), Indian Ocean (IND), North and South Atlantic (ATL), and globally (GLB). The observations are plotted in red, the B-grid ensemble mean is a blue line, and the blue dots are the values for the individual ensemble members. For both model and observations, points where gridded observations are missing and where the mean age is younger than 100 years have been masked out before the averages were computed.

Figure 6: Zonal averages of modelled and observed $\delta^3\text{He}$. The thick white line indicates the boundary of the regions masked out when matching model to observations (waters with an ideal mean age younger than 100 years and above 1000 m depth).

Figure 7: Ensemble-mean B-grid modelled (top) and observed (bottom) $\delta^3\text{He}$ at a depth of 2141 m.

Figure 8: (a) Ensemble-mean B-grid vertically integrated mantle ^3He concentration (the column burden per unit area). (b) Column burden computed from the observation-based estimate of mantle ^3He by *Bianchi et al.* (2010). For both the model and observational estimates, the part of the ocean above 1000 m depth and where the mean water age is less than 100 years below 1000 m depth has been masked out before the integrals were computed.

Figure 9: The ocean-to-atmosphere flux with which mantle ^3He first enters the atmosphere. Note the logarithmic color scale.

Table 1: Previous and current estimates of the mantle ^3He source

study	(year)	method	^3He source [mol/year]
<i>Craig et al.</i>	(1975)	box model of the deep-ocean $\delta^3\text{He}$ distribution	1070 $\pm$ 270
<i>Jean-Baptiste</i>	(1992)	multi-basin box model of the deep ocean	267–534
<i>Farley et al.</i>	(1995)	simulation of $\delta^3\text{He}$ with an ocean general circulation model	$\sim$ 1000
<i>Saal et al.</i>	(2002)	geochemical estimate from mid-ocean ridge basalt volatile content	422 $\pm$ 181
<i>Cartigny et al.</i>	(2008)	geochemical estimate as above	$\sim$ 1000
<i>Bianchi et al.</i>	(2010)	multi-model inversions of the global mantle ^3He inventory	527 $\pm$ 102
<i>Schlitzer</i>	(2016)	single-parameter optimization (OCMIP source strength) of the Pacific $\delta^3\text{He}$ distribution with the AWI Adjoint Tracer Model	450 $\pm$ 50
this study	(2016)	multi-parameter optimization of the global $\delta^3\text{He}$ distribution with multiple ocean circulation inverse models	640–850