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# Sensitivity of sequestration efficiency to mixing processes in the global ocean

B.K. Mignone<sup>a,\*</sup>, J.L. Sarmiento<sup>b</sup>, R.D. Slater<sup>b</sup>, A. Gnanadesikan<sup>b,c</sup>

<sup>a</sup> Department of Geosciences, Princeton University, Princeton, NJ 08544, USA <sup>b</sup> Program in Atmospheric and Oceanic Sciences, Forrestal Campus, Princeton University, Princeton, NJ 08544, USA <sup>c</sup> Geophysical Fluid Dynamics Laboratory, NOAA, Princeton, NJ 08542, USA

### Abstract

A number of large-scale sequestration strategies have been considered to help mitigate rising levels of atmospheric carbon dioxide (CO<sub>2</sub>). Here, we use an ocean general circulation model (OGCM) to evaluate the efficiency of one such strategy currently receiving much attention, the direct injection of liquid  $CO_2$ into selected regions of the abyssal ocean. We find that currents typically transport the injected plumes quite far before they are able to return to the surface and release CO<sub>2</sub> through air-sea gas exchange. When injected at sufficient depth (well within or below the main thermocline), most of the injected  $CO_2$ outgasses in high latitudes (mainly in the Southern Ocean) where vertical exchange is most favored. Virtually all OGCMs that have performed similar simulations confirm these global patterns, but regional differences are significant, leading efficiency estimates to vary widely among models even when identical protocols are followed. In this paper, we make a first attempt at reconciling some of these differences by performing a sensitivity analysis in one OGCM, the Princeton Modular Ocean Model. Using techniques we have developed to maintain both the modeled density structure and the absolute magnitude of the overturning circulation while varying important mixing parameters, we estimate the sensitivity of sequestration efficiency to the magnitude of vertical exchange within the low-latitude pycnocline. Combining these model results with available tracer data permits us to narrow the range of model behavior, which in turn places important constraints on sequestration efficiency.

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<sup>\*</sup> Corresponding author. Tel.: +1-609-258-1314; fax: +1-609-258-2850. *E-mail address:* bmignone@princeton.edu (B.K. Mignone).

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### 1. Introduction

Recently, a number of so-called "geo-engineering" strategies have been considered to help mitigate the rapid and potentially dangerous accumulation of carbon dioxide (CO<sub>2</sub>) in Earth's atmosphere [1,2]. Among the most notable of these strategies is the direct injection and subsequent storage of CO<sub>2</sub> in selected regions of the abyssal ocean [3]. Although the oceans have served as the largest natural sink of CO<sub>2</sub>, absorbing roughly a third of the total release by human activities during the industrial era [4], the rate at which the oceans currently absorb CO<sub>2</sub> is not sufficient to offset the large input from fossil-fuel emissions and deforestation. However, by venting CO<sub>2</sub> captured from power plant flue gases directly into the deep ocean and thereby "short-circuiting" the sluggish exchange of carbon between the major natural interfaces (i.e. atmosphere/upper-ocean and upper-ocean/deep-ocean), direct injection may provide an effective, albeit partial, solution to the greenhouse problem.

Before ocean sequestration can be considered a viable mitigation strategy, however, the nearfield environmental impacts of injection must be evaluated, and injected  $CO_2$  must be shown to remain sequestered from the atmosphere long enough to justify the associated environmental and technological costs. In-situ experimental studies are beginning to supply critical information about the local biotic response to injection (e.g., [5,6]), while the question of retention, which is the primary focus of this paper, has been explored previously using simple one-dimensional ocean models [7,8] and more recently with three-dimensional ocean general circulation models (OGCMs) [9–13]. Typically, these studies evaluate the response to injection in terms of a sequestration "efficiency" [10] that quantifies the ability of the ocean to retain injected  $CO_2$  over time. Unfortunately, there is little consensus at present about the overall viability of direct injection, in part because the magnitude of potential environmental impacts is uncertain, and in part because even the most sophisticated studies employing many OGCMs do not yet yield consistent efficiency estimates even when identical protocols are followed [10]. The most recent of these studies was performed as part of the second phase of the Ocean Carbon-Cycle Model Intercomparison Project (OCMIP-2), initiated by the International Geosphere-Biosphere Program (IGBP) through the Global Analysis, Interpretation and Modeling (GIAM) Task Force. The injection component of OCMIP-2 pooled the resources of eight international carbon-cycle modeling groups, a large subset of which were supported by the EC Global Ocean Storage of Anthropogenic Carbon (GOSAC) project [9].

We speculate in this paper that much of the inter-model variability observed in such studies can be traced to different representations of certain subgrid-scale processes that ultimately determine the extent to which high latitude processes ventilate the ocean interior in the models. The actual sensitivity to such processes is rather difficult to pinpoint, however, because each model differs in multiple respects from every other model, making it impossible to attribute observed differences to a single parameter or set of parameters. Single model sensitivity testing is equally perilous because altering mixing parameters in a model that has already been "tuned" typically leads to model output fields that match well-constrained tracer fields less well than the original and are thus considered poor representations of the real ocean. Here, we report results from a new single model sensitivity analysis in which we rely on a previously documented scheme [14,15] to vary important mixing parameters, such as the rate of diapycnal diffusion in the lowlatitude pycnocline, while preserving the large-scale density structure and overturning circulation in the global ocean. This allows us to probe the sensitivity of sequestration efficiency to oceanic vertical exchange far more effectively than previous studies. We conclude by comparing model results to selected tracer observations, provide early results from a more extensive model study currently underway and speculate on which model version is more likely to produce credible efficiency estimates.

### 2. Modeling protocol and results from previous work

In this study, we use the Princeton/GFDL Modular Ocean Model (MOM), version 3 [16]. MOM-3 is a primitive equation model [17] with 24 vertical levels and increased resolution near the surface. (The topmost level is 25 m thick, while the bottommost is 450 m thick.) The horizontal grid has a nominal resolution of  $4^{\circ}$  and is identical to the grid of the GFDL coupled climate model used for previous carbon cycle studies (e.g., [18]). Lateral mixing is oriented along isopycnal surfaces using the discretization developed by Griffies et al. [19] and includes the socalled "Gent-McWilliams" (GM) parameterization [20-22] of the advective effects of mesoscale eddies. In the version used for OCMIP-2 (henceforth PRINCE-LL), the coefficient of diapycnal diffusion (Kv) is prescribed as a function of depth using the inverse tangent functional form of Bryan and Lewis [23]. Making use of results from recent tracer release experiments in the open ocean pycnocline [24,25], we set Kv to  $0.15 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup> near the surface and increase it at depth (for both physical [26] and numerical [23] reasons) to a maximum value of  $1.3 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup>. The coefficient for lateral (i.e. along-isopycnal) diffusion is fixed throughout at  $1 \times 10^3$  m<sup>2</sup> s<sup>-1</sup> based on the same tracer release experiments. At the surface, heat and fresh water fluxes are applied using the climatology of da Silva et al. [27], but because these flux estimates are rather uncertain, surface temperature and salinity are also restored to the climatology of Levitus and Boyer [28]. Surface wind stresses are given by Hellerman and Rosenstein [29].

In performing simulations, we followed the OCMIP-2 injection protocol [30], which was developed in conjunction with the EC-funded GOSAC project. Briefly this stipulated the following: (1) The model was run out long enough to achieve pre-industrial conditions (atmospheric  $pCO_2$  stable at 278 ppm) and then forced to follow the observed historical trend from 1765 to 2000. During the period 2000–2500, the model tracked the IPCC S650 scenario, which stabilizes atmospheric  $pCO_2$  at 650 ppm in the year 2200. (2) During the period 2000–2100, 0.1 PgC per year was injected (as DIC) into each of seven ocean injection sites around the world (adjacent to New York, San Francisco, Rio de Janeiro, Bay of Biscay, Bombay, Jakarta and Tokyo). (3) Three separate model runs were performed, one with shallow injections at 800 m, another with intermediate injections at 1500 m and a third with deep injections at 3000 m. Separate tracers were used to track the individual DIC plumes and their subsequent escape into the atmosphere.

Further following the OCMIP-2 protocol, we characterized the ability of an ocean site to retain injected  $CO_2$  in terms of a sequestration efficiency. Very simply, this is the fraction of injected  $CO_2$  at a particular site that remains in the ocean after some period of time since the start of injection. In all of our model runs, injection begins in year 2000, so that a "100-year efficiency" measures fractional retention at the end of the injection period (year 2100), a "500-year efficiency" measures retention in year 2500, and so on. Global efficiencies are defined equivalently as the fraction of the *total* amount of injected  $CO_2$  remaining in the ocean after a

given period of time. It is important to note that estimates of sequestration efficiency depend strongly on how the atmospheric boundary conditions are specified in the model. For example, Herzog et al. [31] have shown that calculated efficiencies will be considerably higher in simulations in which a "free" atmosphere is employed (i.e. in which carbon emissions are prescribed and concentrations are calculated) than in simulations in which a fixed atmosphere is employed (in which concentrations are prescribed and allowable emissions are back-calculated). In the former case, injected  $CO_2$  that leaks from the ocean at one point in time may re-enter at a later time, thus markedly increasing net ocean retention, while in the latter case, escape is unidirectional, effectively preventing any re-absorption of lost CO<sub>2</sub>. Because a large fraction of anthropogenic CO<sub>2</sub> will ultimately wind up in the ocean regardless of where it is initially released, Herzog et al. conclude that efficiencies calculated under a fixed atmosphere are more appropriate gauges of sequestration effectiveness, since such calculations do not credit the ocean for uptake that would occur even in the absence of injection. Moreover, while absolute efficiency estimates will vary with the definition and type of simulation, relative differences between models are robust to these choices. For these reasons, and to maintain continuity with past studies, we have chosen to retain the OCMIP-2 protocol and definitions in this paper.

To a large extent, the efficiency of injection is determined by ocean transport processes. Once a parcel of water saturated with injected  $CO_2$  returns to the surface, it participates in air-sea gas exchange, and injected  $CO_2$  will be lost to the atmosphere at a rate that depends on the magnitude of the overall ocean-atmosphere  $pCO_2$  disequilibrium. Thus, how far injected  $CO_2$  can be transported by the ocean before returning to the surface is a critical factor in determining efficiency. As expected, our simulations confirm that deeper injections, in which injected carbon can be advected a great distance before resurfacing, are far more efficient than shallow injections. For example, in the 800 m injection our model predicts that 24% of the globally injected  $CO_2$  remains in the ocean at the end of the 500-year simulation. By contrast, when injections are performed at the same set of sites but at 3000 m, 93% of the injected  $CO_2$  remains in the ocean after 500 years, and at 1500 m, intermediate efficiencies are found (53% globally). In all cases, the main regions of outgassing, and hence the main pathways of escape for injected  $CO_2$ are areas in the Southern Ocean and in the north Atlantic basin, with deeper injections preferentially outgassing in high southern latitudes.

It is rather instructive to compare the PRINCE results with the entire set of results from OCMIP-2. In the latter study, shallow injection runs yielded efficiencies between 14% and 38%, intermediate runs produced efficiencies between 28% and 57% and the deepest injections yielded efficiencies between 48% and 93% [10]. The results show the same general dependence on injection depth and location that the PRINCE runs exhibit, but inter-model variability is quite large within OCMIP-2 because the models differ greatly from one another. For example, lateral mixing is horizontal in some models but oriented along isopycnals (with GM) in others, vertical diffusion is prescribed in some and given by a Richardson number-dependent scheme in others and sea-ice dynamics are explicitly included in some but omitted in others [10]. These and other parameters, taken together, determine the large-scale circulation features of the models, their ventilation characteristics and the degree to which they reproduce the general water mass structure of the global ocean. For example, ventilation of the deep Southern Ocean by Antarctic Bottom Water (AABW) is realistic only in circulation models coupled to sea-ice component models. While comparison with tracer data (e.g., <sup>14</sup>C and CFCs) has suggested that the

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OCMIP-2 models do provide strong upper and lower bounds on efficiency, these studies have been less successful at illuminating the underlying mechanisms at work.

## 3. Sensitivity analysis and discussion

We have suggested that one of the most important parameters for determining sequestration efficiency (and hence for understanding the origin of the large inter-model variability) is the rate of vertical exchange within the low-latitude pycnocline, parameterized in our model by the coefficient of diapycnal diffusion, Kv. The true value of Kv, however, is the subject of much debate among physical oceanographers. Recent observations tend to support low-values (on the order of  $0.1 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup>), at least in the open-ocean pycnocline [25,26], and similar results have been found using microstructure measurements [32,33]. However, theoretical considerations have led others to speculate that these field measurements consistently underestimate the true value of Kv and have suggested that more turbulent mixing near lateral and bottom boundaries might be necessary to ensure that the integral balances hold [34]. Some evidence for increased boundary mixing has come out of the Santa Cruz basin, where diapycnal mixing rates were observed to be on the order of  $1 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup>, an order of magnitude stronger than values observed in the open-ocean pycnocline [35,36].

Because Kv is not well known and is likely an important driver of sequestration efficiency, we have performed a sensitivity analysis, varying Kv across a range of possible values. Increasing Kv alone in our model produces a temperature field in the upper ocean that is far too warm (or equivalently, a pycnocline which is too deep). However, by varying both the lateral diffusivity (Ai) and the vertical diffusivity (Kv) together, it is possible to maintain the shape of the pycnocline (and hence the basic temperature structure) as well as the absolute magnitude of the Northern Hemisphere overturning [14,15]. Thus, it is possible to construct two model versions that describe certain zonally integrated features of the global ocean equally well that nevertheless have very different local circulation patterns. A "high-mixing" model, one in which Kv is large (and in which Southern Hemisphere winds are weak), will predominantly balance the Northern Hemisphere sinking flux by a diffusive return flow within the low-latitude pycnocline, while a "low-mixing" model (small Kv and strong Southern Hemisphere winds) will return a higher proportion of the sinking flux in high southern latitudes via Ekman upwelling.

Above we described the "low-mixing" model, PRINCE-LL, which employs the canonical experimental values for vertical and lateral diffusivities ( $Kv = 0.15 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$  within the upper ocean and Ai =  $1 \times 10^3 \text{ m}^2 \text{ s}^{-1}$ ). We now introduce a complimentary "high-mixing" model, PRINCE-HH, in which the coefficient of diapycnal diffusion is increased by a factor of four in the upper ocean ( $Kv = 0.6 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$ ) to test the effect of higher vertical mixing. The lateral diffusion coefficient is doubled (Ai =  $2 \times 10^3 \text{ m}^2 \text{ s}^{-1}$ ) in this model to offset changes in pycnocline depth and overturning circulation. Finally, we introduce two other versions of the model, PRINCE-LL-HSMIX and the so-called PRINCE-2 model. The former is exactly the low mixing version discussed above except that Kv is increased throughout the water column to  $1.0 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$  south of 55° S (with a linear transition between 45° S and 55° S) in order to test ad hoc the effects of increased mixing within the Southern Ocean. PRINCE-2, on the other hand, was developed systematically to capture other important features of the real

ocean, and in particular, to more accurately reproduce the observed natural <sup>14</sup>C distribution in deep Pacific waters. This model has an intermediate upper ocean diapycnal diffusivity  $(Kv = 0.3 \times 10^{-4} \text{ m}^2 \text{ s}^{-1})$  throughout, a lateral diffusivity of  $1 \times 10^3 \text{ m}^2 \text{ s}^{-1}$  (identical to PRINCE-LL), enhanced southern mixing as in PRINCE-LL-HSMIX and a more shallow inflection point for Kv in the low and middle latitudes (2000 m rather than 2500 m). Additionally, it was shown that biases in the boundary conditions around the Antarctic continent prohibited the formation of Weddell Sea Bottom Water and Ross Sea Bottom Water. To remedy this problem, we increased the surface salinity at a handful of gridpoints corresponding to certain data-sparse regions that are thought to be biased toward lower salinities, thus ensuring that these two water masses realistically outcrop at the surface (see Table 1 for a more comprehensive comparison of model versions).

The results from these four different model versions confirm that sequestration efficiency is indeed highly sensitive to the parameterization of vertical exchange in our model. Fig. 1 (panels A–C) shows the global efficiency at each injection depth for each of the four model versions described above. PRINCE-LL, the model with the smallest diapycnal diffusion coefficient, has the highest efficiency in each case (24%, 53% and 93% for 800, 1500 and 3000 m injections, respectively, in year 2500). Conversely, PRINCE-HH, the model with the largest diffusion coefficient, returns the lowest 500-year efficiencies in all cases (11%, 29% and 77% for 800 m, 1500 m

Table 1

Model	Pycnocline Kv $(10^{-4} \text{ m}^2 \text{ s}^{-1})$	Ai $(m^2 s^{-1})$	Other differences	500-year efficiency (800 m)	500-year efficiency (1500 m)	500-year efficiency (3000 m)
PRINCE-LL	0.15	1000		24.1%	53.0%	92.6%
PRINCE-HH	0.60	2000		11.2%	29.2%	76.5%
PRINCE-LL-	0.15	1000	1	21.3%	47.9%	87.6%
HSMIX						
PRINCE-2	0.30	1000	1, 2, 3	17.4%	42.0%	79.9%
PRINCE-3	0.15	1000	1, 2, 3, 4, 5, 6	19.9%	43.0%	70.1%
OCMIP-LB	Varies	Varies	Many	14%	28%	48%
OCMIP-UB	Varies	Varies	Many	38%	57%	93%

A summary of the major differences between models in the Princeton suite and the resulting global 500-year efficiencies for injections at each depth

The upper and lower bounds of the OCMIP-2 study (OCMIP-UB and OCMIP-LB, respectively) are included for comparison. (Note that the OCMIP-2 model that provides a lower or upper limit at one injection depth does not necessarily also provide the same limit at another injection depth. For this reason, the last two rows of the table can be seen as an "envelope" of all OCMIP-2 runs. Note also that surface boundary conditions are held constant in all OCMIP-2 runs, so that the variability must result from internal circulation differences. Results for these models is taken from Orr et al. [10].) The "other differences" column lists differences between a given model and PRINCE-LL according to the following key: (1) Kv is increased to  $1.0 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup> south of 55° S, with a linear transition between 45° S and 55° S. (2) Inflection point for Kv in low and mid-latitudes reduced from 2500 to 2000 m. (3) Surface salinity in four grid points around Antarctic continent forced to deep values during winter months, with a restoring time constant of 1 day. (4) Wind stresses are given by the ECMWF climatology [41] rather than by Hellerman and Rosenstein [29]. (5) Drake passage narrowed by one grid point (four grid boxes rather than three). (6) Kv between top two model levels everywhere increased to  $50 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup>.



Fig. 1. Panels A–C show global efficiencies for the suite of Princeton models at 800 m, 1500 and 3000 m. The global efficiency is defined as the total mass of  $CO_2$  remaining in the ocean at time *t* divided by the total mass of  $CO_2$  injected by time *t*. Panels D–F show the total loss of injected  $CO_2$  per latitude band, calculated by zonally integrating the air-to-sea flux of injected  $CO_2$  over the 500 year simulation period.

and 3000 m injections, respectively). As seen in the figure, PRINCE-2 and PRINCE-LL-HSMIX yield intermediate efficiencies at all depths after 500 years.

To shed more light on these differences, it is necessary to examine the main pathways by which injected  $CO_2$  escapes to the atmosphere. Fig. 1 (panels D–F) shows the total loss by latitude band, expressed as the zonally integrated cumulative air-to-sea flux of injected  $CO_2$  for each injection depth and model version. In the 800 m and 1500 m injections, the effect of

changing Kv is most apparent in low latitudes. Here, as the diapycnal diffusion coefficient is increased, a larger fraction of the injected  $CO_2$  escapes by upwelling through the main thermocline. At 3000 m, the largest fraction of injected  $CO_2$  outgasses via the Southern Ocean in all models, since this is the region in which most deep water is actively ventilated. As in OCMIP-2, the suite of Princeton models also displays the largest variability in the Southern Ocean. In the region south of 40° S, for example, PRINCE-HH, PRINCE-LL-HSMIX and PRINCE-2 have almost twice the loss of PRINCE-LL when  $CO_2$  is injected at 3000 m. In PRINCE-HH, this is mostly due to increases in convection within the Southern Ocean, while in the others, it is due to increases in the upward diffusive flux as well as to enhanced convection.

In order to determine which model version yields more credible efficiency estimates, we must rely upon available observational constraints. While it is clearly not possible in this study to directly compare model results to observations (there are no naturally occurring tracers with identical interior source functions), we can use other tracers to validate the models indirectly. A good body of evidence now suggests that the ocean is currently in a "low-mixing regime". For example, as discussed above, direct tracer and microstructure measurements indicate that Kv is small in the open ocean pychocline [25,26,32,33]. Furthermore, data-based new production estimates favor models with relatively little low-latitude upwelling [14], and CFC observations, which show only weak penetration into newly formed deep waters, also favor models with modest vertical exchange [37]. While these considerations seem to validate PRINCE-LL (at least over other models in the Princeton suite), a closer look at CFC sections in the Southern Ocean reveals that the ventilation of deep waters in PRINCE-LL is in fact too weak, a concern further highlighted by the inability of PRINCE-LL to accurately reproduce the deep Pacific natural <sup>14</sup>C distribution [9]. This inconsistency illustrates a frequently encountered paradox in global ocean modeling, namely, that accurately reproducing on a global scale both the distributions of transient anthropogenic tracers such as CFCs and the distribution of steady-state tracers with long equilibration timescales such as natural <sup>14</sup>C seemingly requires a model with contradictory ventilation characteristics. In a step toward resolving this quandary, Broecker [38] suggested that ocean ventilation rates may have changed over time in response to climatic fluctuations, with more vigorous circulation during the Little Ice Age (~1350–1880 AD) and a recent slowdown beginning sometime during the late 19th or early 20th century. According to this picture, simultaneous measurements of natural <sup>14</sup>C and CFCs should not be expected to yield similar estimates of ventilation rates, since the former distribution reflects contributions from a gradually slowing ocean, whose effective ventilation rate is given by an average over several centuries, while the latter distribution provides a snapshot of the modern ocean, whose ventilation is presumably much more sluggish. If this hypothesis proves correct, then we will be further justified in assuming that the ocean is currently in a low mixing state, though PRINCE-LL should still be viewed as supplying an upper bound rather than a central estimate for sequestration efficiency because of the overly sluggish mixing in the Southern Ocean.

Before turning to solutions that require significant ocean reorganization, however, we should first determine whether it is possible in principle to capture the full range of observed behavior within a single model, and if so, whether the models described above may simply be deficient in some way. As discussed, PRINCE-LL and PRINCE-HH represent two extreme views of the large-scale ocean circulation, with the former describing a "low-mixing" ocean in which the primary return flow of light water (i.e. the dense to light conversion) takes place in high latitudes, and the latter representing a "high-mixing" ocean in which light water returns primarily in low latitudes. Despite convincing empirical evidence that diapycnal mixing rates are small, comparison of CFC and natural <sup>14</sup>C observations clearly indicates that PRINCE-LL is simply too sluggish to adequately ventilate the deep ocean in high latitudes. Conversely, the same analysis shows that PRINCE-HH *over*estimates CFC inventories and barely falls within the error limits of measured natural <sup>14</sup>C inventories. These well-known shortcomings have spawned newer model versions (only some of which are described here) that attempt to capture the best features of each and that do so to varying degrees of success [39].

While the models presented in this paper rely upon fundamentally different circulations and thus implicitly incorporate a variety of ventilation mechanisms, the output of the model suite, when expressed in terms of basin-averaged inventories, can be crudely viewed as a linear combination of the output from each of the two end-member models, roughly tracing out a line in <sup>14</sup>C-CFC space that connects PRINCE-LL and PRINCE-HH. The data, which fall in between these extremes and slightly off the trend line toward lower CFC values, appear to most strongly validate PRINCE-2. However, this simple comparison ignores other important theoretical and empirical considerations, such as the fact that the rate of diapycnal diffusion in PRINCE-2 is still quite high (twice the empirically derived values of Ledwell et al. [25]). Ultimately, creation of a model that exhibits both realistic ventilation and empirically consistent vertical exchange may require finding the proper combination of surface boundary conditions and internal model physics. While the importance of internal physics cannot be ignored, there is indeed ample evidence that surface boundary conditions play a significant role in controlling circulation, and hence ventilation, with some studies suggesting, for example, that the strength of the global overturning circulation is tightly coupled to the magnitude of the Southern Hemisphere wind stress field [40]. To briefly explore the sensitivity of sequestration efficiency to surface boundary conditions, we consider one final model version, called PRINCE-3, in which the PRINCE-LL mixing coefficients are used but in which the Hellerman and Rosenstein [29] wind stress forcing is replaced by more recent, and presumably more accurate data from the ECMWF climatology [41]. Table 1 provides results from this simulation and summarizes all of the model results in this paper as well as the key differences between model versions. We see that PRINCE-3 yields efficiencies similar to PRINCE-2, except that the deepest injection (at 3000 m) is noticeably less efficient than all other models in our suite, including PRINCE-HH. A quick comparison with observations shows that PRINCE-3 overestimates CFCs and slightly underestimates C-14, but in both cases the model output is shifted in the right direction relative to PRINCE-LL, suggesting that higher winds may indeed induce greater ventilation without the need for unrealistically large diapycnal diffusivities. The increased ventilation, however, leads to noticeably decreased efficiency, with the gains in retention from reduced mixing being more than offset by losses from more rapid ventilation. This suggests that future studies aimed at evaluating the effectiveness of ocean injection should examine the sensitivity of efficiency to surface boundary conditions in addition to the dependence on internal model physics.

## 4. Conclusions

In this paper we have explored the sensitivity of sequestration efficiency to the rate of diapycnal diffusion in the low-latitude pycnocline, a parameter that is thought to drive a large fraction of the observed variability between OGCMs (including that seen within OCMIP-2) through its effect on ocean ventilation. We have shown that, in general, efficiency decreases with increased vertical exchange, as intuitive arguments might suggest. Although our model suite was specifically designed to examine variations in mixing without distorting other basic features of the large-scale ocean structure, a basic comparison with observations suggests that none of the models in the original Princeton suite (or in OCMIP-2) provides an entirely satisfactory description of real ocean behavior, thus casting some uncertainty onto efficiency estimates. Our canonical high-mixing model, PRINCE-HH, requires mixing parameters that are unrealistically large, while the standard low-mixing model, PRINCE-LL, generally fails to provide sufficient deep ocean ventilation. Furthermore, preliminary results from a more extensive model study indicate that changes in the Southern Hemisphere wind stress field may allow both adequate ventilation and realistic mixing to be simultaneously incorporated into a single model. Although the "highwind" model ostensibly yields the lowest efficiency estimates in the Princeton suite, it is possible that the ventilation in this model is in fact too strong, suggesting that these results should be considered a lower bound on efficiency. Ultimately, if PRINCE-LL and PRINCE-3 do indeed bracket real ocean behavior as the foregoing analysis suggests, then one may reasonably conclude that ocean injection will remain between 70% and 93% efficient after 500 years. Of course, none of the studies presented here explicitly accounts for changes in ocean carbon storage that may result from the sorts of climate changes anticipated in coming decades and centuries. It is clear that the largest gains in our future predictive ability will come from a deeper understanding of the mechanisms of ocean ventilation and Southern Ocean processes and from improved models that can accurately capture these processes and their response to human perturbation.

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