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## Brine-induced stratification

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# Impact of brine-induced stratification on the glacial carbon cycle

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## Abstract

During the cold period of the Last Glacial Maximum (LGM, about 21 000 years ago) atmospheric CO<sub>2</sub> was around 190 ppm (Monnin et al., 2001), much lower than the pre-industrial concentration of 280 ppm. The causes of this substantial drop remain partially unresolved, despite intense research. Understanding the origin of reduced atmospheric CO<sub>2</sub> during glacial times is crucial to comprehend the evolution of the different carbon reservoirs within the Earth system (atmosphere, terrestrial biosphere and ocean). In this context, the ocean is believed to play a major role as it can store large amounts of carbon (Sigman and Boyle, 2000), especially in the abyss, which is a carbon reservoir that is thought to have expanded during glacial times. To create this larger reservoir, one possible mechanism is to produce very dense glacial waters, thereby stratifying the deep ocean and reducing the carbon exchange between the deep and surface ocean (Paillard and Parrenin, 2004). The existence of such very dense waters has been inferred in the LGM deep Atlantic from sediment pore water salinity (Adkins et al., 2002). Based on these observations, we study the impact of a brine mechanism on the glacial carbon cycle. This mechanism relies on the formation and rapid sinking of brines, very salty water released during sea ice formation, which brings salty dense water down to the bottom of the ocean. It provides two major features: a direct link from the surface to the deep ocean along with an efficient way of setting a strong stratification. We show with the CLIMBER-2 coupled carbon-climate model (Petoukhov et al., 2000) that such a brine mechanism can account for a significant decrease in atmospheric CO<sub>2</sub> and contribute to the glacial-interglacial change. This mechanism can be amplified by low vertical diffusion resulting from the brine-induced stratification. The results obtained substantially improve the modeled glacial distribution of oceanic  $\delta^{13}\text{C}$  as well as the deep ocean salinity in line with reconstructions from sediment cores (Curry and Oppo, 2005; Adkins et al., 2002), suggesting that such a mechanism could have played an important role during glacial times.

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# 1 Introduction

Proxy data suggest that the climate of the Last Glacial Maximum (LGM, about 21 000 years ago) was very cold ( $-2$  to  $-6^{\circ}\text{C}$  in the Southern Ocean, Kucera et al., 2005) with huge Northern Hemisphere ice sheets (Peltier, 2004). The associated carbon cycle was characterized by low atmospheric  $\text{CO}_2$  concentrations of  $\sim 190$  ppm (Monnin et al., 2001) and very negative deep ocean  $\delta^{13}\text{C}$ . The latter is defined as:

$$\delta^{13}\text{C} = \left( \frac{R}{R_{\text{ref}}} - 1 \right) \cdot 1000$$

$$\text{with } R = \frac{{}^{13}\text{C}}{{}^{12}\text{C}}$$

$R_{\text{ref}}$  is the PDB (Peedee belemnite) carbon isotope standard, which corresponds approximately to average limestone (Craig, 1957).

The Atlantic  $\delta^{13}\text{C}$  reached low values down to  $-0.8\text{‰}$  in the deep ocean participating in a higher surface to deep oceanic gradient (Duplessy et al., 1998 and Curry and Oppo, 2005). The mean surface ( $-2000$  m to  $0$  m) to deep ( $-5000$  m to  $-3000$  m) gradient in the Atlantic, (noted  $\Delta\delta^{13}\text{C}$ ), was about  $1.4\text{‰}$  in the Atlantic, compared to only  $0.6\text{‰}$  during the pre-industrial.

LGM climate can be explained by the extended ice sheets in connection with a different orbital configuration (Berger, 1978) and lower  $\text{CO}_2$  (Jahn et al., 2005). To explain the crucial question of the low glacial  $\text{CO}_2$  numerous hypotheses have been proposed. Some of them imply changes of physical mechanisms such as modifications of sea ice coverage (Stephens and Keeling, 2000) or winds (Toggweiler et al., 2006). Many have focused on enhancing or making more efficient the marine biology, for example through higher C/P ratio (Broecker and Peng, 1982), iron fertilization (Martin, 1990), a shift of dominant plankton species (Archer and Maier-Reimer, 1994), larger nutrients availability (Matsumoto et al., 2002), or modified rain ratio (Brovkin et al., 2007). Other studies have involved coral reef fluctuations (Berger, 1982; Opdyke and Walker, 1992),

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or oceanic chemistry with carbonate compensation (Broecker and Peng, 1987). But most only have a small impact on  $\text{CO}_2$  compared to the glacial-interglacial change, or would require unrealistic modifications to become dominant (Archer et al., 2000, 2003; Kohfeld et al., 2005; Menviel et al., 2008). Moreover, it has remained especially difficult to simulate simultaneously the very negative  $\delta^{13}\text{C}$  in the deep ocean inferred from marine sediment cores. Additionally, the strong link between Antarctic temperatures and atmospheric  $\text{CO}_2$  variations (Lüthi et al., 2008) suggests that mechanisms closely tied to Southern Ocean surface processes are likely to be dominant in controlling atmospheric  $\text{CO}_2$  variability.

The general consensus is that the ocean is at the core of the solution regarding LGM  $\text{CO}_2$ . It is the biggest carbon reservoir active on the short time scale studied (a few thousand years) and the only one that could increase during glacial times, since the other two reservoirs, i.e., the atmosphere (Monnin et al., 2001) and terrestrial biosphere (Bird et al., 1994; Crowley, 1995), were both reduced. Observations indicate that the deep glacial ocean was much saltier and colder than today (Adkins et al., 2002), thus more stratified in the abyss. Such a deep stratification has important impacts on the ocean's circulation and carbon cycle, as pointed out by climate models of different complexities (Toggweiler, 1999; Paillard and Parrenin, 2004; Köhler et al., 2005; Bouttes et al., 2009; Tagliabue et al., 2009). In particular, deep stratification is required to reconcile  $\delta^{13}\text{C}$  (Bouttes et al., 2009; Tagliabue et al., 2009). However, only box models could significantly reduce atmospheric  $\text{CO}_2$  when prescribing a reduced Southern ventilation (Toggweiler, 1999; Paillard and Parrenin, 2004; Köhler et al., 2005; Watson and Naveira-Garabato, 2005). With models of higher complexity (Bouttes et al., 2009; Tagliabue et al., 2009) the induced  $\text{CO}_2$  drop remained too small which highlights the need of another mechanism that would still maintain the benefits of deep stratification (deep negative  $\delta^{13}\text{C}$  values) while reducing  $\text{CO}_2$  more drastically.

Here we test the impact of the formation and rapid deep sinking of brines, i.e. very salty waters rejected during sea ice formation, on the glacial carbon cycle. The role of brines has been previously proposed as an explanation for the reduced Southern venti-

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lation (Paillard and Parrenin, 2004; Köhler et al., 2005; Watson and Naveira-Garabato, 2005), but they were not explicitly simulated. Instead, the proposed effect of reducing the ventilation was tested by imposing lower mixing in box models only. Here we directly test this mechanism with a more complex model which physically includes advection and diffusion in the ocean and thus is able to reproduce the dynamical effect of brines. Because they are enriched in salt, these “pockets of water” are very dense. In the modern Antarctic they are generally mixed with fresh water from ice shelf melting (Joughin and Padman, 2003). However, during the glacial periods the sea level progressively fell (down to about 120 m at the Last Glacial Maximum) and the northward Antarctic ice sheet extent resulted in a progressive reduction of the shelf slope (Ritz et al., 2001) where modern waters are mixed. Tidal dissipation, which is important in today’s mixing, would then seriously decrease. Accordingly, the brine signal, which is diluted today, would be more preserved at the LGM. Furthermore, the sea ice formation location would also shift northward and closer to the continental shelf break. The preserved brine-dense water, whose volume was increased because of enhanced sea ice formation, would arrive at the shelf break and then flow quickly down the slope (helped by thermobaricity and supercritical flux properties) (Foldvik et al., 2004). This mechanism (referred herein thereafter as the brine mechanism) provides two major features: a direct link from the surface to the deep ocean and an effective physical way of achieving a strong stratification. In this study we test its impact on the carbon cycle and compare the modeled results to atmospheric  $\text{CO}_2$ , oceanic  $\delta^{13}\text{C}$  and salinity data inferred from ice core and marine sediment cores.

## 2 Methods

### 2.1 CLIMBER–2 model

The brine mechanism has been implemented and tested in CLIMBER–2, an intermediate complexity climate model (Petoukhov et al., 2000) well suited for the long term

simulations we run. Indeed, the simulations are run for 20 000 years to ensure the equilibrium of the carbon cycle. Moreover we realize an ensemble of about 100 simulations to test the mechanism, which would be unfeasible with a general circulation model (GCM) in a reasonable amount of time. The intermediate complexity model, although simpler than a state of the art OGCM, includes the main known processes and mechanisms and computes the dynamics of the oceanic circulation contrary to box models. Furthermore, like other GCMs, CLIMBER-2 does not exhibit box model sensitivity to high-latitude sea ice or presumably stratification (Archer et al., 2003). Additionally, the model compares favourably with a state of the art OGCM and gives the same response in terms of carbon cycle when the circulation is arbitrarily modified (Tagliabue et al., 2009).

CLIMBER-2 has a coarse resolution of 10° in latitude by 51° in longitude in the atmosphere, and 21 depth levels by 2.5° latitude in the zonally averaged ocean, which is precise enough to take into account geographical changes, while allowing the model to be fast enough to run the long simulations considered. The model is composed of various modules simulating the ocean, the atmosphere, and the continental biosphere dynamics. No sediment model is included, the model thus does not take into account the carbonate compensation mechanism. CLIMBER-2 has already been used and evaluated in previous studies (Ganopolski et al., 2001; Brovkin et al., 2002a, b; Brovkin et al., 2007; Bouttes et al., 2009). As the model version used explicitly computes the evolution of the carbon cycle and carbon isotopes (such as  $\delta^{13}\text{C}$ ) in every reservoir, it allows us to compare the model output with data from sediment cores. In the glacial simulations three boundary conditions are simultaneously imposed: the ice sheets (Peltier, 2004), the solar insolation (Berger, 1978), and the atmospheric  $\text{CO}_2$  concentration for the radiative forcing (190 ppm, not used in the carbon cycle part of the model) (Monnin et al., 2001). To account for the glacial sea level fall of about 120 m, salinity and nutrient mean concentrations are increased by 3.3%.

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## 2.2 Implementation of brines in the CLIMBER-2 model

When sea ice is formed, a flux of ions, among which salt, is released into the ocean as sea ice is mostly composed of fresh water (Wakatsuchi and Ono, 1983; Rysgaard et al., 2007, 2009). Because the underlying water is then enriched in salt it becomes denser and can thus sink and transport salt to deeper waters. The rejection of salt by sea ice formation plays an important role in the formation of deep water, as it is the case for the Antarctic Bottom Water (AABW) which has been studied for a few decades (Foster and Carmack, 1976; Foster and Middleton, 1980; Whitworth and Nowlin, 1987; Foldvik et al., 2004; Nicholls et al., 2009). The brine mechanism has been mostly observed in details in the Northern Hemisphere oceans (Haarpaintner et al., 2001; Shcherbina et al., 2003; Skogseth et al., 2008, 2004), where measures are easier than in the Southern Ocean. For instance, measures in the Arctic fjords of the first rapid salt release during sea ice formation show that it constitutes around 82% of the total salt flux, the rest being more slowly released later. Moreover it has been observed that approximately 78% of the brine-enriched shelf water was released out of the fjord in the Norwegian Sea (Haarpaintner et al., 2001), i.e. around 62% of the salt flux rapidly released by sea ice formation. The impact of the brine formation on the concentration of other geochemical variables, especially dissolved inorganic carbon (DIC) has also been studied and it has been shown that DIC is rejected together with brine from growing sea ice (Rysgaard et al., 2007, 2009). Besides, numerical studies of this local mechanism show that the topography plays an important role in the transport of salt (Kikuchi et al., 1999).

Thus although today the brine formation around Antarctica does not seem to reach the bottom waters (Toggweiler and Samuels, 1995), the formation of brines is a mechanism that can be observed and studied in some places of the modern ocean and that strongly depends on local conditions. The inferred higher salinity in the glacial Southern Ocean seems to indicate that enhanced brine formation could well have taken place around Antarctica during glacial conditions, which should be tested.

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In order to assess the potential impact of brines on the carbon cycle we need to use a global coupled carbon-climate model. Yet the spatial resolution of such models, even state of the art GCMs, is too large to resolve the brine sink, requesting a parameterization. We thus develop a simple parameterization for this mechanism. The simplicity of the scheme used permits a first evaluation of such a mechanism and allows a separation of the different processes to understand the reasons of the changes observed.

In the standard version of CLIMBER-2 only the flux of salt ( $F_S$ ) was considered. It is usually a good approximation on the first order, yet the release of other ions and dissolved gases could be of importance for this study. Hence we have added the same process as the release of salt for the other ions and dissolved gases simulated in the model (DIC, ALK, nutrients, dissolved organic carbon, oxygen,  $^{13}\text{C}$ ). As a first approximation the surface oceanic cell is enriched in these geochemical variables in the same proportion as salt as it has been observed to be not very different on the first order (Rysgaard et al., 2007, 2009). The flux ( $F_X$ ) of any geochemical variable  $X$  is then:

$$F_X = \frac{F_S}{S_{\text{surface}}} \cdot X_{\text{surface}}$$

With  $F_S$  the flux of salt,  $S_{\text{surface}}$  the surface salinity, and  $X_{\text{surface}}$  the surface concentration of any geochemical variable. This process is active in all simulations, though it has a small impact on the standard pre-industrial and LGM simulations (atmospheric  $\text{CO}_2$  is changed by less than 5 ppm).

In the standard version of CLIMBER-2 when sea ice is formed all the salt is released in the corresponding surface oceanic cell, which is relatively large according to the coarse resolution of the model and thus dilutes the brine signal. In the brine simulations, a fixed fraction *frac* of this salt flux (and of the other ions flux) will form the brines (see Fig. 1). This fraction *frac* can be set to 0 when no brine is formed to 1 when all the salt is used in the brine mechanism. The very salty (thus dense) brine water sinks to the bottom of the ocean where it modifies the concentration of any geochemi-

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cal variable  $X$  (salinity, DIC, ALK, nutrients, dissolved organic carbon, oxygen and  $^{13}\text{C}$ ) as following:

$$V_{\text{bottom}} \frac{dX_{\text{bottom}}}{dt} = \text{frac} \cdot F_X \cdot \text{area}$$

With  $V_{\text{bottom}}$  the volume of the bottom cell,  $X_{\text{bottom}}$  the concentration of  $X$  at the bottom and  $\text{area}$  the area of the surface cell. At the surface the rest of the ion flux from sea ice formation not sinking with brines  $((1 - \text{frac}) \cdot F_X)$  is diluted in the surface cell. The geochemical variables are thus modified as following:

$$V_{\text{surface}} \frac{dX_{\text{surface}}}{dt} = (1 - \text{frac}) \cdot F_X \cdot \text{area}$$

Even if this brine mechanism is idealized, it reflects the impact of intense Antarctic sea ice formation during the LGM. As the glacial Antarctic ice sheet extends close to the limit of the continental shelf, sea ice formation enhanced by the strong katabatic winds happens close to the continental slope. The brines formed by the coeval salt rejection can then rapidly sink along the continental slope down to the deep ocean. Unlike convection, the brine mechanism directly transports this fraction of salt and other ions  $(\text{frac} \cdot F_X)$  to the deep ocean, and thus represents a direct link from the surface to the bottom of the ocean.

We present model results and compare them to the main carbon data available for the LGM, e.g. atmospheric  $\text{CO}_2$  concentration and  $\delta^{13}\text{C}$  distribution in the ocean, which constitute a major constraint with which to validate this mechanism. We also compare model results to the glacial deep ocean salinity.

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### 3 Results and discussion

#### 3.1 Standard simulation

The CLIMBER-2 version used here has a closed carbon cycle i.e. the total (fixed) amount of carbon is interactively distributed between the three reservoirs (atmosphere, terrestrial biosphere and ocean) (Brovkin et al., 2002a), and does not include carbonate compensation. From the pre-industrial value of 280 ppm, CO<sub>2</sub> rises to 296 ppm for the standard glacial simulation (LGM-std, Fig. 2a). This CO<sub>2</sub> rise is due to the prevailing effect of increased ocean salinity and terrestrial vegetation decline (of approximately 700 GtC) which increase CO<sub>2</sub>, despite increased nutrient concentration (linked to sea level drop) and colder sea surface temperatures which decrease CO<sub>2</sub>. This modeled 16 ppm increase adds to the observed 90 ppm drop between the Pre-industrial and the LGM and means the total required simulated LGM drawdown is 106 ppm ( $\Delta\text{CO}_2$ ). The simulated oceanic  $\delta^{13}\text{C}$  is also far from the data value, in particular the surface to bottom  $\delta^{13}\text{C}$  gradient ( $\Delta\delta^{13}\text{C}$ ) is not well represented, with a simulated value of around 0.5‰ compared to the data value of about 1.4‰ (Fig. 2b). Finally the simulated salinity in the deep Southern Ocean (Fig. 2c) lies well below the data value of around 37.1 psu. This mismatch between model results and data indicate the need for a mechanism which would increase the deep Southern salinity (the global mean increase of salinity due to sea level fall is already taken into account in all simulations) and lower the deep  $\delta^{13}\text{C}$  thus probably changing oceanic circulation. The brines seem to be a good candidate as they will bring saline water to the abyss and hence increase deep water salinity and modify the circulation. In the following we will assess the impact of the brines and compare with data to either validate or reject such a possibility.

#### 3.2 CO<sub>2</sub> drawdown induced by the brine mechanism

Brines can have a very large effect on atmospheric carbon in our simulations (Fig. 2a). Atmospheric CO<sub>2</sub> progressively decreases when the fraction of salt *frac* that sinks to

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the abyss increases. The  $frac=0$  simulation (no brine mechanism) corresponds to the standard LGM simulation (LGM-std), where  $CO_2$  is 296 ppm. The maximum effect of the brines is obtained for  $frac=1$ , i.e. when all the salt released by sea ice formation sinks to the bottom of the ocean with the brine mechanism. The latter gives a maximum  $CO_2$  drop of about 52 ppm ( $CO_2$  is 244 ppm).

To understand the reasons of the  $CO_2$  drawdown, we consider the effect of the transport of the geochemical variables separately by brines (Fig. 3). The geochemical variables considered are salinity, dissolved inorganic carbon (DIC) and alkalinity (ALK), nutrients, and dissolved organic carbon (DOC). They are transported by brines along with salt and their distribution is modified in two ways: directly by the brine transport, as well as indirectly by the brine-induced stratification due to the salt transport, which alters circulation.

First we consider only the transport of salt to the deep ocean; the other variables do not sink with the brines. The salt sink appears to be the main driver of the atmospheric  $CO_2$  drawdown as it accounts for most of the entire brine induced drop (Fig. 3). Indeed the increased salinity of the bottom waters (Fig. 2c) results in greater deep ocean density. Hence the deep stratification is greater and the oceanic circulation modified (Fig. 4). The thermohaline circulation is slowed and the Glacial North Atlantic Deep Water (GNADW) becomes shallower. This leads to an increased efficiency of trapping of  $CO_2$  released from the remineralization of organic matter, as the carbon enriched Antarctic Bottom Water (AABW) is less mixed with the surrounding waters and constitutes a greater volume of water. The deep carbon reservoir has thus expanded thanks to the density change induced by the salt transport only.

The transport of dissolved inorganic carbon (DIC) and alkalinity (ALK) by brines also plays a role as it further decreases  $CO_2$  (Fig. 3). The direct transport of DIC and ALK to the abyss helps building an increased deep oceanic carbon reservoir as carbon is brought to the deep ocean but can not escape because of the stratification set by the increased salinity.

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However, the transport of nutrients has an opposite effect, as their transport to the abyss decreases the surface concentration, thus the biological activity (Fig. 3). The biological pump being less efficient, less carbon is taken up. Yet this effect is smaller than the two previous ones. Finally the transport of DOC is negligible (less than 1 ppm).

Now that we have seen that the salt sink is the main driver of the CO<sub>2</sub> drop and that the DIC and ALK transport can also play a role, we investigate how they impact the atmospheric CO<sub>2</sub>. Because of the direct effect (transport from the surface to the bottom) and indirect effect (change of oceanic circulation because of the stratification) their distribution in the ocean (especially the repartition between surface and deep waters) is modified. The changed surface concentrations modify the oceanic dissolved CO<sub>2</sub> which ultimately drives atmospheric CO<sub>2</sub>.

Because of the salt transport by brines, salinity is increased in the deep ocean (Fig. 2c) and decreased in the surface. As the solubility of CO<sub>2</sub> increases when salinity decreases it results in a CO<sub>2</sub> drawdown. To quantify this impact we have run the geochemical module of CLIMBER-2 alone forced by the standard distribution of all variables (from the LGM-std simulation without brines) except salinity. For the latter we prescribe the distribution obtained in the simulation with brines for *frac*=0 to 1. This allows us to get the CO<sub>2</sub> change only due to the salinity distribution change. It appears that the modification of the salinity distribution generally accounts for around 1/4 to a 1/3rd of the total CO<sub>2</sub> decrease (Fig. 5).

Similarly, DIC and ALK are increased in the deep ocean and decreased in the surface (Fig. 6), both because of the salinity induced stratification (we have seen that this is the main process) and the direct transport of DIC and ALK by brines. To assess the impact of the change of DIC and ALK distribution we realize the same test as for salinity. With the geochemical module of CLIMBER-2 we evaluate the impact of the change of distribution of DIC and ALK alone for *frac*=0 to 1. It shows that the DIC and ALK distribution modification can generally explain between 1/2 and 2/3rd of the CO<sub>2</sub> decrease (Fig. 5). Indeed, as the brines sink and make the deep ocean saltier, increased vertical stratification results. Southern convection is greatly reduced and

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very little exchange exists from the deep ocean to the surface, creating a deep isolated water mass that is enriched in DIC and ALK (while the surface is depleted, Fig. 6). With respect to CO<sub>2</sub>, DIC and ALK changes have opposite effects (reduced surface DIC decreases CO<sub>2</sub>, reduced ALK increases CO<sub>2</sub>), but the DIC change prevails and results in a net CO<sub>2</sub> drawdown.

### 3.3 Impact of brines on oceanic $\delta^{13}\text{C}$

Carbon isotopes, especially  $\delta^{13}\text{C}$ , are crucial data used to constrain the model results.  $\delta^{13}\text{C}$  is usually used to track oceanic circulation, since their changes reflect the changes in ventilation of the different water masses (Duplessy et al., 1998 and Curry and Oppo, 2005). In the brine simulations, the modeled  $\Delta\delta^{13}\text{C}$  in the Atlantic Ocean is increased when the fraction of salt *frac* increases (Fig. 2b), which improves the results compared to proxy data. Several mechanisms can affect  $\Delta\delta^{13}\text{C}$ , as biology and circulation both impact the distribution of  $\delta^{13}\text{C}$ . Indeed, the vertical gradient of  $\delta^{13}\text{C}$  is primarily due the biological activity which preferentially incorporates  $\delta^{12}\text{C}$  during photosynthesis. This tends to deplete the surface waters of  $\delta^{12}\text{C}$  and hence increase  $\delta^{13}\text{C}$ . On the opposite, when organic matter is remineralized deeper in the ocean it releases  $\delta^{12}\text{C}$  thus decreases the deep  $\delta^{13}\text{C}$ . This process is then modulated by the oceanic circulation which transports and mixes water masses with different  $\delta^{13}\text{C}$  signatures and thus modifies the  $\delta^{13}\text{C}$  distribution. With the brine mechanism these two processes are involved as the salinity transport alters circulation, the DIC, ALK and DOC transport directly modifies the surface and deep  $\delta^{13}\text{C}$  values, and the nutrient transport impacts the biological activity.

To assess the separate role of the salinity, DIC and ALK, Nutrients and DOC transport on the increase of  $\Delta\delta^{13}\text{C}$  we analyze the same simulations as for CO<sub>2</sub>, when considering only the transport of one variable at a time (Fig. 3b). As for the CO<sub>2</sub> decrease, most of the  $\Delta\delta^{13}\text{C}$  increase is explained by the salinity transport. Because of the brine-induced stratification and resulting modified circulation (Fig. 4) the waters

are less mixed. Hence the low  $\delta^{13}\text{C}$  deep waters and high  $\delta^{13}\text{C}$  surface water tend to maintain their different values and the vertical gradient increases.

The DIC and ALK transport as well as nutrient transport have an opposite yet minor effect. The DIC and ALK transport brings high surface  $\delta^{13}\text{C}$  values down to the bottom and tends to lower the  $\Delta\delta^{13}\text{C}$ , but it has only a small impact. The transport of nutrients lowers the surface nutrient concentration which decreases the biological production. As the latter is responsible for the initial  $\Delta\delta^{13}\text{C}$  it also lowers  $\Delta\delta^{13}\text{C}$ . However this effect is negligible. Finally the effect of DOC transport is too small to be seen.

It appears that the salinity transport is clearly the main process driving the  $\Delta\delta^{13}\text{C}$  increase, while the other variables have only a small impact. The  $\Delta\delta^{13}\text{C}$  is thus mainly improved due to the change of circulation.

### 3.4 Amplifications by low vertical diffusion

In the previous simulations, only the impact of brines on convection and advection was taken into account via salinity changes. But it is highly probable that such changes would also reduce vertical diffusion. CLIMBER-2 prescribes vertical diffusion as a fixed vertical profile, whereas in reality it depends on the vertical density profile in the ocean. The brine induced stratification would therefore also change diffusion, as the more stratified the water becomes, the less vertical diffusion there would be (it requires more energy to mix well stratified water masses). To test this impact on  $\text{CO}_2$ , we conduct a series of simulations with the brine mechanism where three low vertical diffusion profiles (Kz1, Kz2 and Kz3) are prescribed (Fig. 7). The impact of such low diffusion profiles alone in CLIMBER-2 has been previously studied (Bouttes et al., 2009) which showed that it helps reconcile the deep glacial  $\delta^{13}\text{C}$  though with a small impact on atmospheric  $\text{CO}_2$  (10 ppm decrease only). Here we test these low diffusion profiles when the brine mechanism is activated in the model. We run a set of simulations with varying the *frac* value between 0 and 1 for the three diffusion profiles.

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The effect of low diffusion alone that was previously analyzed resulted in a very small impact on atmospheric CO<sub>2</sub>, which is the case for the *frac*=0 simulations. When combined with brines, the brine induced CO<sub>2</sub> decline is amplified and it leads to a further significant CO<sub>2</sub> decrease (Fig. 8a). It shows a clear amplification of the brine mechanism. Indeed, the low diffusion prevents even more the mixing between the carbon enriched bottom waters and the surface. Therefore it amplifies the trapping of carbon in the deep ocean and results in the supplementary simulated CO<sub>2</sub> drawdown.

The amplification also applies for  $\delta^{13}\text{C}$ , especially for  $\Delta\delta^{13}\text{C}$  which is increased (Fig. 8b). Indeed, when low diffusion is added, the deep  $\delta^{13}\text{C}$  becomes even more negative. The deep (below 2000 m) ocean  $\delta^{13}\text{C}$  distribution thus compares far better with the data than in our standard simulation (LGM-std) where the values were too high (Fig. 9). The surface-intermediate (above 2000 m) values are greater, resulting in the stronger surface to deep gradient. Hence  $\delta^{13}\text{C}$  strongly support the brine mechanism amplified by low diffusion. Yet even though the Antarctic Bottom Water (AABW) expands and the Glacial North Atlantic Deep Water (GNADW) shoals, the  $\delta^{13}\text{C}$  values are still not negative enough in the intermediate south. This suggests that either GNADW should be further reduced and confined to the northern Atlantic, or that another southern water mass exists at intermediate depths (e.g. a Glacial Antarctic Intermediate Water).

### 3.5 Glacial-interglacial carbon cycle changes

Although the brine mechanism has the potential to decrease significantly CO<sub>2</sub>, additional mechanisms are also very likely to be involved. In particular, within the already tested mechanisms, the addition of carbonate compensation is a likely candidate, since it would further decrease CO<sub>2</sub> by around 30 ppm (Broecker and Peng, 1987). Additionally, modifications to the biological pump (linked to dust reconstructions and phytoplankton stoichiometry) can provide an additional 10–25 ppm (Bopp et al., 2003; Tagliabue et al., 2009), as well as increasing carbon export in line with proxy reconstructions. Overall, a combination of low diffusion, carbonate compensation and enhanced

biological pump added to the brine mechanism could yield the full range of glacial to interglacial variability of CO<sub>2</sub>.

## 4 Conclusions

In conclusion, the brine mechanism can lead to a large CO<sub>2</sub> drop supported by the concomitant simulated changes in oceanic carbon isotope distribution, in agreement with proxy data. We have shown that this CO<sub>2</sub> decline is primarily due to the salinity transport although the DIC and ALK sink also play a role. Because of the salt transport to the deep ocean the oceanic circulation is modified and the deep ocean becomes stratified. The abyss is enriched in carbon which is trapped because of the reduced mixing with other water masses. This decrease can be amplified if the simultaneous impact on oceanic vertical diffusion is accounted for. The induced low diffusion would further reduce the mixing between the deep and upper ocean, hence decreasing atmospheric CO<sub>2</sub> and increasing the  $\Delta\delta^{13}\text{C}$  in line with data. We hypothesize that a combination of the already known carbonate compensation mechanism, iron fertilization, brine mechanism and low diffusion would be sufficient to reach the glacial value of 190 ppm. The brine mechanism provides two major features: a physical way of setting the needed glacial deep ocean stratification and a direct connection from surface to deep waters, which is crucial to create a very large deep ocean carbon reservoir. Beyond the understanding of past climate, this mechanism sheds new light on ocean dynamics. Brines have a crucial role in the formation of deep water and should therefore be better accounted for in climate models.

*Acknowledgements.* We thank Alessandro Tagliabue for comments and ideas, and Laurent Bopp for discussion. We also thank Bill Curry for providing the  $\delta^{13}\text{C}$  data and Victor Brovkin for his help on the CLIMBER-2 model.

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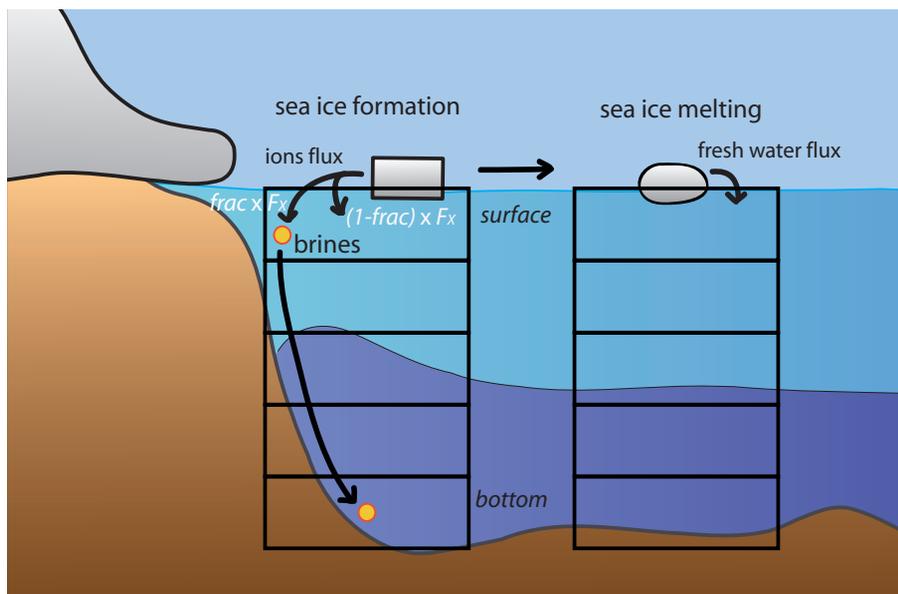
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**Fig. 1.** Brine sinking mechanism scheme. When sea ice is formed salt and other modeled ions (and dissolved gases), such as dissolved inorganic matter (DIC), alkalinity (ALK), dissolved organic carbon (DOC), nutrients, oxygen, and  $^{13}\text{C}$  are released into the surface ocean beneath as a flux ( $F_X$  which is the flux of variable  $X$ ). A fraction of this flux ( $\text{frac} \cdot F_X$ ) is dedicated to the brine sinking mechanism and is directly transported to the deep ocean instead of being diluted in the surface cell as the rest of the flux ( $(1 - \text{frac}) \cdot F_X$ ). This mechanism both creates a link from the surface to the deep ocean and sets an efficient deep ocean stratification.

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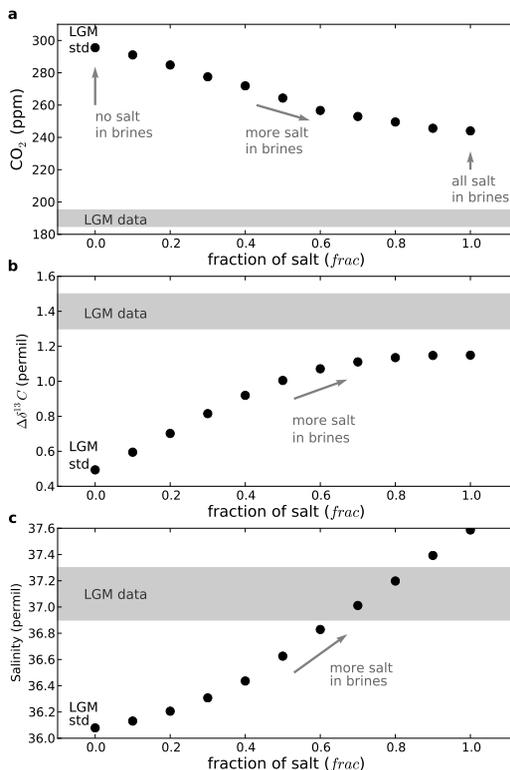
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**Fig. 2.** (a) Atmospheric CO<sub>2</sub> concentration, (b) Δδ<sup>13</sup>C (mean surface (–2000 m to 0 m) to deep (–5000 m to –3000 m) gradient in the Atlantic) and (c) salinity as a function of the fraction of salt released by sea ice formation used for the brine mechanism (fraction of salt *frac*, 0 ≤ (*frac*) ≤ 1). *frac*=0 corresponds to the standard simulations without any brine mechanism (all the salt and ions are diluted in the surface cell where sea ice is formed). When *frac* = 1 all the released salt and other ions sink with the brine mechanism down to the abyss.

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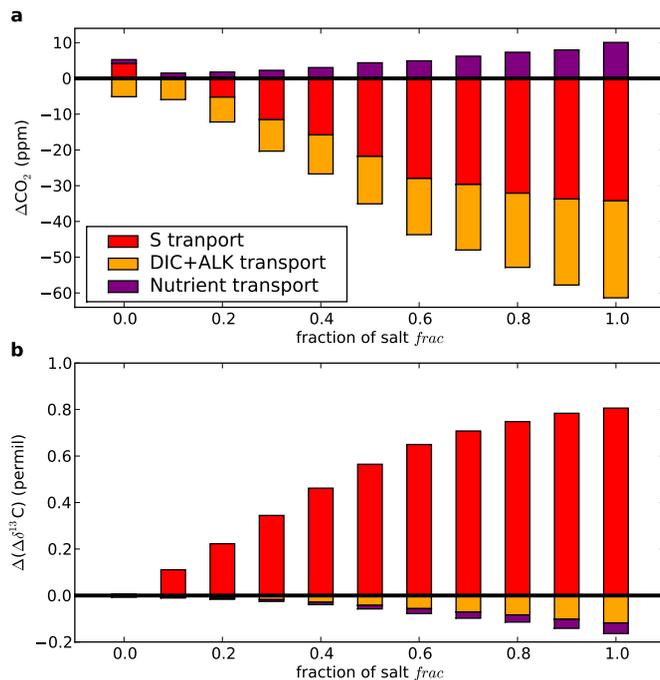
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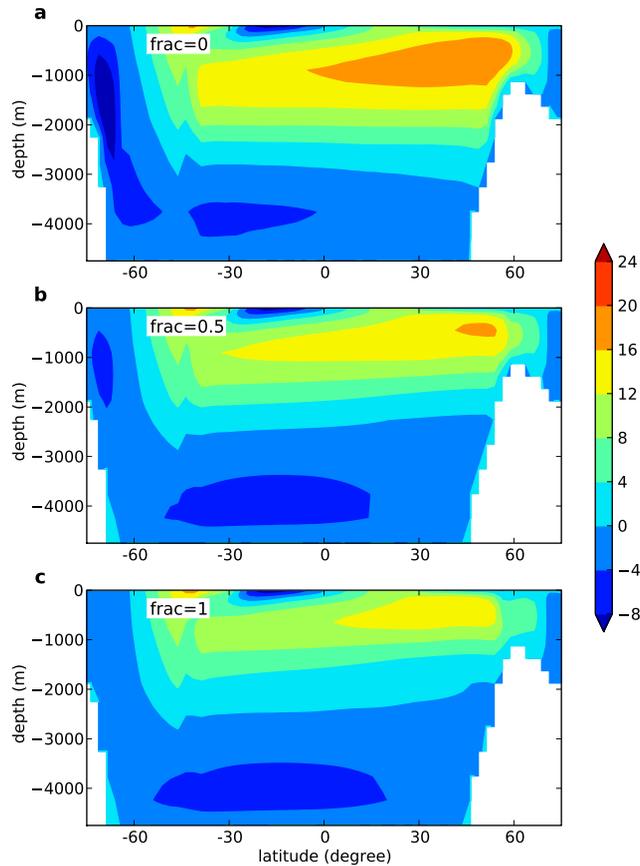


**Fig. 3.** (a)  $\text{CO}_2$  change ( $\Delta\text{CO}_2$ ) and (b)  $\Delta\delta^{13}\text{C}$  change ( $\Delta(\Delta\delta^{13}\text{C})$ ) due to the separate transport of salt, dissolved inorganic carbon (DIC) and alkalinity (ALK), nutrients, and dissolved organic carbon (DOC).  $\Delta\text{CO}_2$  is the difference between the modeled  $\text{CO}_2$  and the standard modeled  $\text{CO}_2$  (LGM std,  $\text{CO}_2=296$  ppm).  $\Delta(\Delta\delta^{13}\text{C})$  is the difference between the modeled  $\Delta\delta^{13}\text{C}$  and the standard modeled  $\Delta\delta^{13}\text{C}$  (LGM std).

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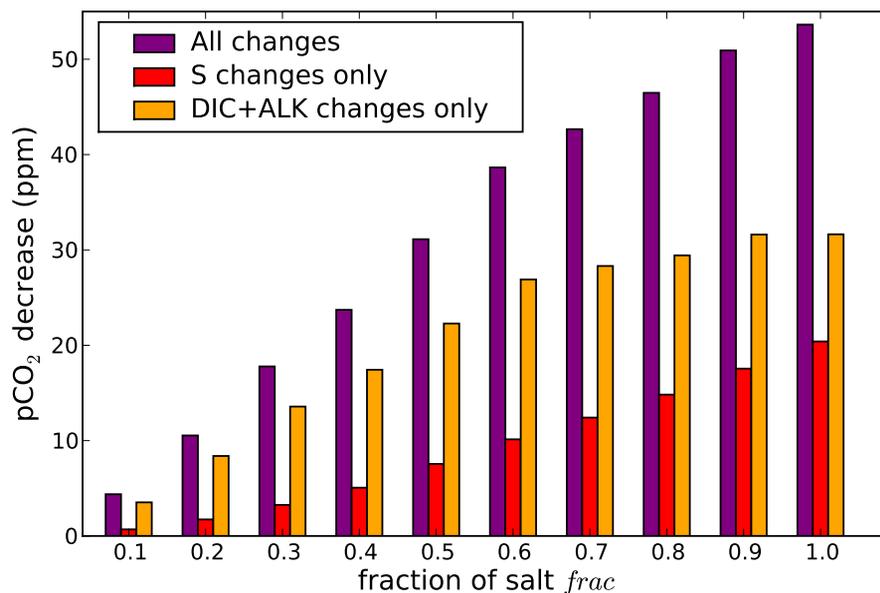

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**Fig. 4.** Simulated thermohaline circulation in the Atlantic for **(a)** the standard simulation (LGM-std) with  $frac=0$  (no impact of brines), **(b)**  $frac=0.5$  (medium impact of brines), **(c)**  $frac=1$  (maximum impact of brines).

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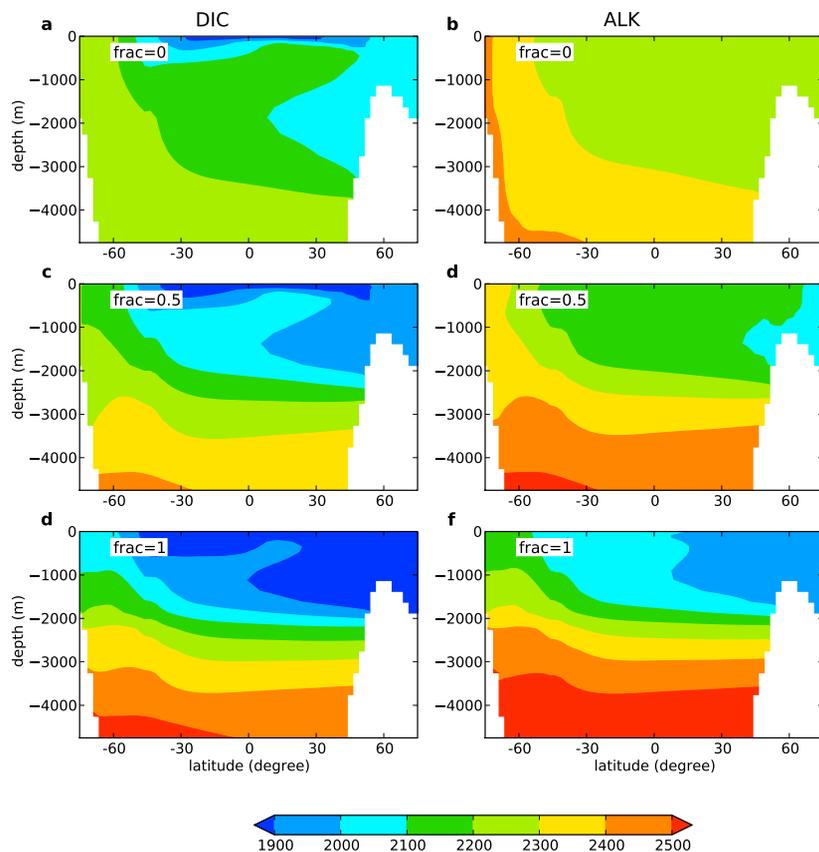


**Fig. 5.** Oceanic surface  $p\text{CO}_2$  decrease due to the brine mechanism as a function of the fraction of salt rejected by sea ice formation used for the brine mechanism (fraction of salt  $frac$ ,  $0 \leq frac \leq 1$ ). “All Changes” (purple) corresponds to the drawdown due to the change of distribution of all oceanic variables. The “S changes only” (red) is the contribution of the modification in the salinity (S) distribution only to the  $\text{CO}_2$  drawdown (the distribution of the other variables is the one from the standard simulation LGM-std with  $frac=0$ ). Similarly, the “DIC + ALK changes only” (orange) corresponds to the contribution of the modification of dissolved inorganic carbon (DIC) and alkalinity (ALK) only.

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**Fig. 6.** (a), (c), (d) Dissolved inorganic carbon (DIC) and (b), (d), (f) alkalinity (ALK) distribution in the Atlantic for (a), (b) the standard simulation (LGM-std) with  $frac=0$  (no impact of brines), (c), (d)  $frac=0.5$  (medium impact of brines), (e), (f)  $frac=1$  (maximum impact of brines).

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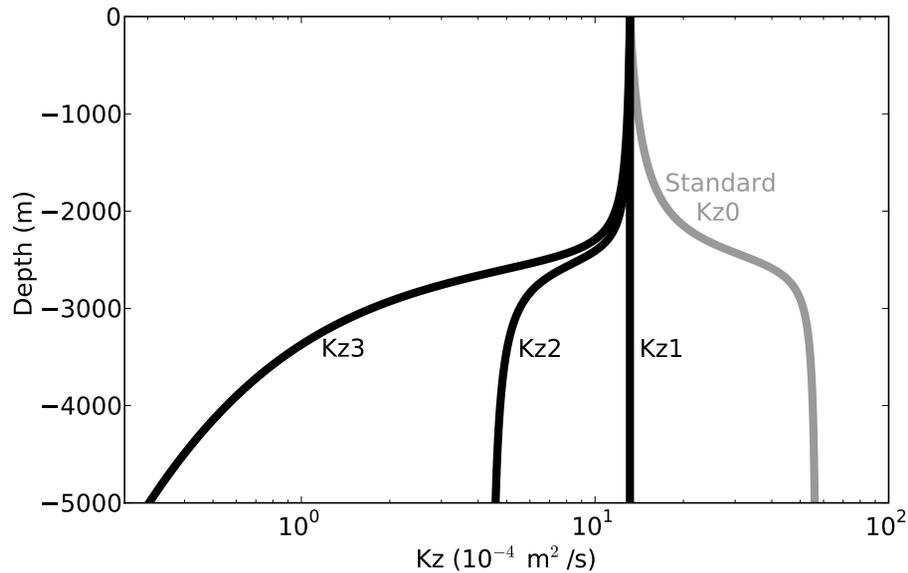
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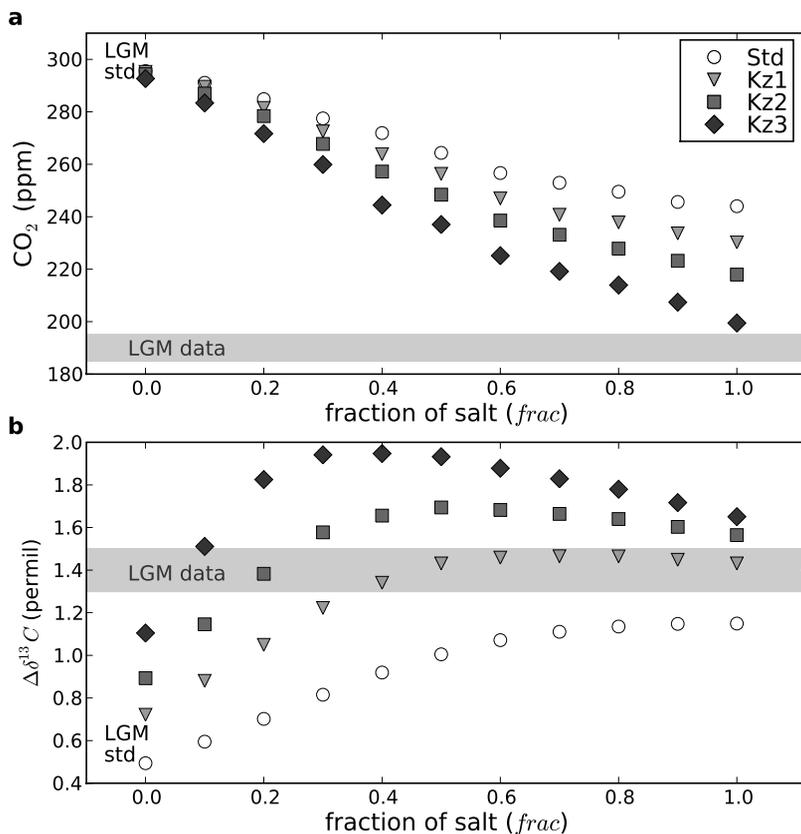
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**Fig. 7.** Vertical diffusion coefficient profiles imposed in the simulations.  $K_{z0}$  is the standard profile,  $K_{z1}$ , 2 and 3 are the low diffusion profiles used to test the impact of deep low diffusion.

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**Fig. 8.** (a) Atmospheric  $CO_2$  concentration and (b)  $\Delta\delta^{13}C$  as a function of the fraction of salt released by sea ice formation used for the brine mechanism (fraction of salt  $frac$ ,  $0 \leq frac \leq 1$ ) for different vertical diffusion profiles. Kz0 (grey) is the standard profile, Kz1, 2 and 3 are the tested low diffusion profiles (black).

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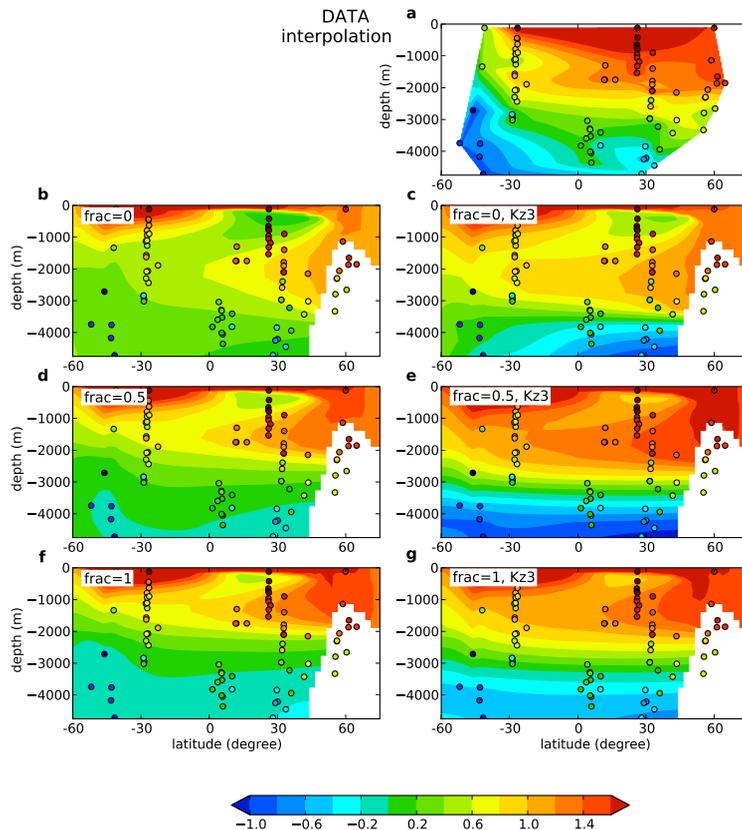
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## Brine-induced stratification

N. Bouttes et al.



**Fig. 9.**  $\delta^{13}\text{C}$  (‰) distribution in the Atlantic. The scatter plots represent the LGM data (Curry and Oppo). **(a)** Interpolation of the data. **(b), (c), (d), (e), (f), (g)** Modelled  $\delta^{13}\text{C}$  distribution: **(b), (d), (f)** with the standard diffusion profile (Kz0); **(c), (e), (g)** with a low diffusion profile (Kz3). **(b), (c)**  $\text{frac}=0$  (no impact of brines), **(d), (e)**  $\text{frac}=0.5$  (medium impact of brines), **(f), (g)**  $\text{frac}=1$  (maximum impact of brines).

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