Regulation of atmospheric CO2 by sea iron solubility impacts during the Last Glacial Maximum and Holocene in an Earth system model

1 Introduction

Iron (Fe) stands as the fourth most abundant element in the Earth's crust. Despite its abundance, the concentrations of iron at the oceanic surface remain relatively low, ranging from 0.1 to 2 nM (nanomolar) (Nakabayashi et al., 2002; Tani et al., 2003). These concentrations have a remarkable impact on the delicate balance of the marine ecosystem. Phytoplankton production, a cornerstone of oceanic food webs and a significant driver of global carbon cycling, can be significantly constrained due to Fe's crucial enzymatic role (Martin, 1990; Jickells et al., 2005; Martínez-García et al., 2014). Notably influencing processes like photosynthesis, respiration, and nitrogen fixation (Falkowski et al., 1998; Morel and Price, 2003; Kustka et al., 2003), Fe becomes a limiting factor that reverberates through the marine food chain. This influence assumes heightened significance in specific regions, such as the Southern Oceans, the Pacific and North Atlantic, and the Eastern Equatorial Pacific, collectively known as high-nutrient low-chlorophyll concentration (HNLC) zones (Coale et al., 1996; Boyd et al., 2000, 2004, 2007).

Several pathways contribute to delivering this essential micronutrient to the ocean's surface. Hydrothermal vents, rivers, glaciers, icebergs, continental edges, and upwelling mechanisms all play roles in introducing iron into oceanic ecosystems (Ducklow et al., 2003; Tagliabue et al., 2016). Nevertheless, beyond continental margins, the importance of dust fluxes, sourced from arid and semi-arid desert regions, emerges as a fundamental factor (Tagliabue et al., 2017; Lambert et al., 2021). These dust fluxes, mobilized by wind forces and sometimes traveling substantial distances, settle onto the ocean surface through dry or wet deposition, thereby becoming a vital avenue for Fe input. The propensity for dust generation lies in regions characterized by low vegetation coverage and water deficits (Prospero and Lamb, 2003; Prospero et al., 2002; Jickells et al., 2005; Mahowald et al., 2005; Buseck and Adachi, 2008; Hand et al., 2003).

This relationship between dust and carbon balances was first illuminated by the work of Gran et al. (1931), followed by the research of Martin (1990) in the Southern Oceans. These studies revealed the pronounced influence of dust events on primary productivity. Since then, several works (Kohfeld et al., 2005; Jaccard et al., 2013; Petit et al., 1990; Steffensen, 1997; Lambert et al., 2008; Archer et al., 2000) postulate that the efficiency of the soft tissue biological pump during glacial periods could be attributed to the increased availability of aeolian iron, thus linking iron availability to CO2 levels. This mechanism, operating as a recurrent Earth system feedback, is believed to have exerted periodic influences on the carbon cycle over the past 800,000 years. This could potentially explain up to one-third of the observed natural variability in CO2 concentrations, ranging approximately between 180 ppmv and 280 ppmv during glacial and interglacial periods, respectively (Petit et al., 1990; Siegenthaler et al., 2005; Lüthi et al., 2008).

Approximately 3% of atmospheric dust consists of Fe (Marcotte et al., 2020), contributing around 14-16 Tg annually in the form of mineral-sourced dust aerosols (Jickells et al., 2005; Gao et al., 2003). Regrettably, only a fraction of this deposited Fe, ranging from 1 to 10%, is available to support phytoplankton growth (Journet et al., 2008; Jickells and Spokes, 2001; Archer and Johnson, 2000; Bopp et al., 2003). Fe can exist in two oxidation states – Fe(II) and Fe(III), as organic ferrous (Fe^{2+}) or organic ferric (Fe^{3+}), respectively. Among these, only the ferrous form is bioavailable, although it is less prevalent. The solubility of Fe, typically defined as the amount of metal that passes through a 0.2 or 0.4 µm filter, depends on factors encompassing deposited dust mineralogy, acidity, water pH, and other environmental variables (Luo et al., 2005; Sholkovitz et al., 2012; Marcotte et al., 2020). While Fe(III) dominates due to processes like oxidation, reduction, and photochemical interactions, it exhibits limited solubility (Wells et al., 1995; Byrne et al., 2000). However, it can eventually dissolve by diverse mechanisms, such as proton-induced Fe-O bond breakage (Cwiertny et al., 2008), photochemical reduction (Fu et al., 2010), and organic ligand complexation, which is the most prevalent. Organic ligands, arising biologically from water column organic matter, play a significant role in increasing soluble Fe concentrations (Fe2+), facilitating biological uptake, and extending the residence time of bioavailable Fe, thus mitigating precipitation and scavenging within the water column (Baker and Croot, 2010).

From the mid-1990s onward, efforts emerged to simulate CO2 fluctuations on a global scale using numerical models (Johnson et al., 1997). Diverse modeling approaches, spanning from box models to intricate General Circulation Models (GCMs), attempted to elucidate the complex causes underlying CO2 variability between glacial and interglacial periods. Yet, despite considerable advancement, the intricacies of this variability remained incompletely understood. Consequently, biogeochemical processes, intricately interwoven with ocean-atmosphere dynamics, have been integrated to offer additional insights into CO2 dynamics (Flato et al., 2014).

Our objective is to elucidate the impact of iron solubility on glacial-interglacial atmospheric CO2 balances over the past 800,000 years. Employing cGENIE, an intermediate complexity model emphasizing the carbon cycle, we conducted sensitivity simulations for pre-industrial and Last Glacial Maximum (LGM) periods. Leveraging a diverse array of simulations, models, empirical dust deposition data, and heterogeneous Fe solubility fields, our study hones in on regions highly sensitive to iron biogeochemistry, particularly the HNLC zones. Through this integrated approach, we aim to enhance our current understanding of this intricate interplay between iron solubility and atmospheric carbon capture.

2 Methodology

2.1 Model description

(van de Velde et al., 2021)

In this work we use a version of GENIE focused on the carbon cycle, cGENIE muffin, free version v0.9.5 (it is hosted in the cGENIE Github repository). cGENIE is a type of Earth System Model of Intermediate Complexity (EMIC), composed of different modules, each one representing different components of the Earth system. In this work, we use the 3D frictional–geostrophic ocean module, with 16 depth levels and 36 × 36 equal-area horizontal grid. In longitude, it has a compression of 10°, and in latitude it goes from 3.2°, at the Equator, to 19.2°, at high latitudes (Edwards and Marsh, 2005). We also use the biogeochemistry module (Ridgwell et al., 2007) and, an atmospheric component of the 2D energy–moisture balance model (EMBM) with prescribed climatological wind fields (Cao et al., 2009). The continental distribution and bathymetry are published in Ridgwell et al. (2007).

2.2 Experiment design

Both phytoplankton and zooplankton require specific concentrations of certain macro and micronutrients that promote their existence and development. For this work, this will be reflected as organic matter production. It will be estimated based on the fixed radio of Redfield (1934, 1963), where $P/C/O_2 = 1/106/- 138$. However, concerning Fe and phosphorus, the current version of cGENIE considers a collimitation scheme (Fe/C and P/C). This is due to the high variability in the demand for these nutrients, determined by the biogeochemistry of the environment, the availability of light, water temperature, and the growth stage of the species . In order to make a correct carbon inventory, tracers and preformed insert cite nutrients were used following the base configuration of Cao et al. (2009). In addition to a simplified ligands scheme and their processes similar to that portrayed by Parekh et al. (2004, 2006). Dissolved Fe input to the system, consumed during biological uptake, will be driven by various dust flux fields deposited in the surface ocean. The residence time of the particulate organic matter that forms and sinks from the surface layer will be directly related to the scavenging rate. Additional supplies of iron associated with sedimentary processes in the ocean, or any other type of source other than wind, will not be considered.

Regarding the forcing of the system, we worked with: 1) 6 pairs of dust fluxes fields from the LGM and Holocene period. One of them is empirical data called "Lambert" (Lambert et al., 2015), the rest are CMIP5 model simulations surface dust flux reconstructions called "Albani", "Takemura", "Ohgaito","MIROC-ESM" and "MRI-CGCM3" (Albani et al., 2014; Takemura et al., 2009; Ohgaito et al., 2018; Sueyoshi et al., 2013; Yukimoto et al., 2012) para mayor detalle ver Lambert et al. (2021); or more detail see Lambert et al. (2021); and since the solubility of iron is not well known both today and during the past, 2) different iron solubility fields were created for each of these six initial flux field pairs. These latest results from the application of a linear regression model, estimated from the relationship between the Holocene dust flux field of Mahowald et al. (2006) and the calibrated iron solubility field, developed by the marine biogeochemical cycling module of cGENIE, called BIOGEM (Ridgwell et al., 2007). Additionally, for each iron solubility reconstruction, we varied its values in only one of the 5 HNLC regions. This alteration is the product of the application of 4 factors that multiply the original values of the iron solubility fields. These factors are 1/2, 2/3, 2, and 3. Thus, for each of the 6 pairs of iron solubility reconstructions, 5 modified iron solubility reconstructions were produced (one for each of the 5 HNLC zones), by 4 scalar factors. Consequently, the model was forced with 240 different global patterns of iron solubility and six dust flow fields to estimate CO2 capture.

We initialized by performing 6 different pre-industrial equilibrium experiments (spin-up), in which the atmospheric CO2 concentration have been settled at 278 ppmv compared to the control (free CO2), using the cGENIE Earth System model, and runing for 10000 years. (to reach the equilibrium state). We forcing the experiments using Holocene dust and iron solubility fields from differents authors, with the goal of compared the ocean carbon uptake potential, even so, they have the same pCO2 in their spin-up have variable oceanic nutrients inventory and carbon distribution, which will affect the behavior of the biological pump. Starting from the end of each spin-up, we run global and regional sensitivity experiments (see figure **??**) for the Holocene and LGM data, and again we allow the model to run 10000 years. finally, we took the mean of the last 500 years of pCO2 estimates from each simulation (240 in total).

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3 Results

4 Discussion

5 Conclusion

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