Dust fluxes and iron fertilization in Holocene and Last Glacial Maximum climates

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Abstract
Mineral dust aerosols play a major role in present and past climates. To date, we rely on climate models for estimates of dust fluxes to calculate the impact of airborne micronutrients on biogeochemical cycles. Here we provide a new global dust flux data set for Holocene and Last Glacial Maximum (LGM) conditions based on observational data. A comparison with dust flux simulations highlights regional differences between observations and models. By forcing a biogeochemical model with our new data set and using this model’s results to guide a millennial-scale Earth System Model simulation, we calculate the impact of enhanced glacial oceanic iron deposition on the LGM-Holocene carbon cycle. On centennial timescales, the higher LGM dust deposition results in a weak reduction of <10 ppm in atmospheric CO2 due to enhanced efficiency of the biological pump. This is followed by a further ~10 ppm reduction over millennial timescales due to greater carbon burial and carbonate compensation.

1. Introduction

Mineral dust aerosols in the atmosphere affect the climate directly by absorbing and scattering incoming solar and outgoing infrared radiation [Tegen et al., 1996; Sokolik et al., 2001] and indirectly by acting as ice nuclei [Sassen et al., 2003; Lohmann and Feichter, 2005]. The deposition of dust on the Earth’s surface also provides the terrestrial biosphere and surface ocean with micronutrients, affecting biogeochemical cycles [Martin et al., 1991; Bristow et al., 2010; Mahowald, 2011]. Early efforts to simulate dust deposition under past climatic conditions were conducted using various global climate models [Mahowald et al., 1999; Werner et al., 2002; Tegen, 2003]. Within the Paleoclimate Modeling Intercomparison Project (PMIP) 2, dust simulations were performed using the CAM3-CCSM3 (Mahowald et al. [2006], hereafter CCSM) and SPRINTARS-MIROC3 (Takemura et al. [2009], hereafter MIROC) climate models. Recently, the PMIP 3 climate models MIROC-ESM [Watanabe et al., 2011] and MRI-CGCM3 (Yukimoto et al. [2012], hereafter MRI) also included dust in their paleoclimatic simulations. However, there are significant differences in dust load and spatial distribution among various dust simulations, even under modern conditions [Huneeus et al., 2011].

From an observational perspective, paleoclimatic archives provide us with a record of past variability of dust deposition at specific locations. An extensive amount of Holocene and Last Glacial Maximum (LGM) dust flux measurements were compiled in the DIRTMAP database [Kohfeld and Harrison, 2001; Maher et al., 2010]. These observations have varying analytical uncertainties, and the spatial distribution of the global data set is irregular. Globally, the LGM atmosphere is thought to have contained 3–4 times more dust than during the Holocene [Kohfeld and Harrison, 2001; Maher et al., 2010], which reflects the changes in the tropics, where most of the atmospheric dust is emitted. In remote regions far away from sources, LGM dust deposition has been found to be 2–3 times higher in the tropical and South Pacific [Winckler et al., 2008; Kohfeld et al., 2013; Lamy et al., 2014], ~5 times higher in the South Atlantic [Martinez-Garcia et al., 2009] and 20–30 times higher in polar regions [Steffensen, 1997; Lambert et al., 2008, 2013].

Dustborne iron fertilization of the nutrient-rich Southern Ocean (SO) waters and the subsequent sequestration of carbon in the ocean interior has been proposed as a driver of lower levels of atmospheric CO2 during glacial periods of the late Pleistocene [Martin et al., 1990]. In certain regions of the world’s oceans known as
“High Nutrient Low Chlorophyll” (HNLC) regions, macronutrients like nitrate and phosphate are abundant, but the lack of iron restricts oceanic primary productivity [Martin and Fitzwater, 1988]. It is argued that increased aeolian iron input during the LGM stimulated primary production, thus amplifying the actions of the biological and carbonate pumps that transport carbon down from the ocean surface [Martin, 1990; Martin et al., 1990; Hain et al., 2014]. Artificial iron fertilization experiments have stimulated phytoplankton blooms [e.g., Coale et al., 1996; Boyd et al., 2000, 2007], with more than half of the captured carbon potentially able to sink into the deep ocean [Smetacek et al., 2012]. However, it is unclear how much of the sinking carbon returns to the ocean surface layers [de Baar, 2005]. Studies using paleoclimatic records of high southern latitudes to link dust deposition to marine productivity estimate that increased Last Glacial Maximum (LGM) dust deposition may have contributed up to 40% of the Holocene-LGM difference of 80–100 ppm in the partial pressure of atmospheric CO$_2$ (pCO$_2$) [Röthlisberger et al., 2004; Martinez-Garcia et al., 2011; Lambert et al., 2012]. In contrast, many model studies that directly relate the ocean carbon cycles to fluctuations in dust deposition tend to assign a lower significance (<20%) to this phenomenon [Bopp et al., 2003; Ridgwell, 2003; Parekh et al., 2006; Tagliabue et al., 2009, 2014], although these models usually focus on biological productivity over centennial timescales and thereby neglect millennial-scale processes of carbon burial and carbonate compensation.

Here we present a new interpolation of the unevenly spaced Holocene and LGM dust flux measurements compiled in the DIRTMAP database [Kohfeld and Harrison, 2001; Maher et al., 2010] to two global grids. We compare our interpolated dust deposition fields with four climate model simulations of global dust flux to identify regions with large discrepancies. We use our interpolated dust fluxes to force a biogeochemical model and calculate the carbon export impact due to iron fertilization between Holocene and LGM climatic conditions. Together with a long Earth System Model simulation guided by results of the biogeochemical model, we estimate the centennial- and millennial-scale effects of carbon export changes on atmospheric pCO$_2$ levels.

2. Methods

The spatial distribution of dust flux measurements has greatly improved in recent times, with the third version of the DIRTMAP database including almost twice the number of sites compared to the previous version [Maher et al., 2010]. For this study we used 375 and 311 data points for average Holocene and LGM conditions, respectively, with additional data points not included in DIRTMAP 3 listed in Table S1 in the supporting information (SI). The data locations are sufficiently well distributed to perform a global interpolation on a T42 (128 longitude × 64 latitude) grid. At first order, the aerosol concentration in an air parcel decreases at an exponential rate with distance from the source [Hansson, 1994; Ruth et al., 2003; Fischer et al., 2007]. We extend this exponential relationship with distance to produce deposition fluxes, assuming in effect constant deposition velocities. This assumption is valid as we are considering multiyear average Holocene and LGM values and not seasonal variations and as dust flux is lognormally distributed on a large scale in all climate models (SI Figure S3). We do not account for the fact that the dry deposition velocity changes due to varying surface roughness; however, this is justified by the fact that in the majority of the world and particularly over land, wet deposition dominates [Mahowald et al., 2006; Takemura et al., 2009]. Large-scale precipitation and wind direction patterns result in directional variance that is around 4 times higher in the north-south direction than in the east-west direction. This anisotropy of the field results from the preferential zonal transport of dust by the main wind currents and was addressed in the directional semivariogram of the interpolation procedure (see SI). Thus, we interpolated the logarithmic values of the measured dust fluxes using a Kriging algorithm [Cressie, 1991] (Figure 1). For loess data that usually contain a significant fraction of large dust particles that do not travel far, we used the PM10 (particle diameter <10 μm) fraction, whereas total dust deposition measurements from ice cores and marine sediments were considered as representing long-range transport. This primary stage is similar to the method used previously [Lambert et al., 2013]. Here we additionally corrected the variogram for the difference in distance represented by 1° of longitude at low and high latitudes (see SI), thus, producing a global data set of yearly average dust fluxes for both Holocene and LGM conditions. The distribution of the original dust flux measurements, the interpolation, and the simulations of the four models we used are compared in SI Figure S3 and show the spatial lognormality of dust deposition. To force the biogeochemistry
model, a monthly component was added by applying the ensemble median of the monthly variations around the yearly mean from the four climate models at each grid cell.

The interpolated global dust flux grid was used to force the biogeochemical model NEMO-PISCES [Tagliabue et al., 2014]. Although the iron fraction of dust and the iron solubility are spatially and temporally variable [Schroth et al., 2009], we kept these values constant as in previous simulations [Tagliabue et al., 2014] because of the lack of available observations. PISCES is a relatively complex ocean biogeochemical model that considers two phytoplankton size classes, two zooplankton grazers, two particle size classes, and five nutrients (nitrate, phosphate, silicic acid, ammonium, and iron). The iron cycle in PISCES accounts for organic speciation, scavenging, and its variable biological uptake and considers iron sources from dust, rivers, sediments, and hydrothermal vents [Tagliabue et al., 2014]. The enhancement of nitrogen fixation through additional dust input in tropical oceans [Mills et al., 2004] is present in PISCES simulations. We conducted simulations of 500 years where the dust field was replaced by the LGM and Holocene fields produced in this study. By coupling a well-mixed atmospheric reservoir, we were able to determine the influence of dust variations on atmospheric pCO2 levels on centennial scales. Lastly, we conducted a 50,000 year simulation with the Danish Center for Earth System Science (DCESS) Earth System Model [Shaffer et al., 2008; Zickfeld et al., 2013] using the relative enhancement of high-latitude export production found in the shorter PISCES simulations. With its relatively sophisticated ocean sediment module, the DCESS model allows us to estimate the millennial-scale CO2 effects of iron fertilization from carbon burial and carbonate compensation.

3. Results and Discussion

On average, we find the interpolated global LGM dust flux to be about 4 times higher than during the Holocene (Figure 1, top row). This is in the upper range of previous estimates based on observational data [Kohfeld and Harrison, 2001; Maher et al., 2010] and slightly higher than model results [Werner et al., 2002; Mahowald et al., 2006; Takemura et al., 2009; Watanabe et al., 2011; Yukimoto et al., 2012], which may reflect the recognized underestimation of dust over land in models [Maher et al., 2010]. The uncertainty of

![Figure 1.](image-url)
the interpolation is indicated in Figure 1 (bottom row). By definition and within the uncertainty of the observations (which the models have to deal with as well for their validation), the interpolation is a more trustworthy estimate for dust fluxes than the models in the vicinity of measurement points. In Figure 1 (top row) the nonshaded areas correspond to data where the uncertainty is smaller than its global mean (i.e., the more trustworthy half), whereas the shaded regions correspond to uncertainties higher than the mean.

North and South American sources appear particularly more potent in the interpolation during the LGM than during the Holocene, something which is generally difficult to reproduce in models [Mahowald et al., 2006; Takemura et al., 2009; Watanabe et al., 2011; Yukimoto et al., 2012]. A large part of the North Atlantic LGM dust deposition appears to originate from sources in North America and North Africa (Figure 1). Although dust in Greenland ice cores has traditionally been considered to originate in Asia [Biscaye et al., 1997; Svensson et al., 2000], its isotopic signature remains ambiguous [Burton et al., 2007; Aleinikoff et al., 2008] and few LGM reference samples have been measured. In our interpolation, Siberian and Alaskan LGM dust sources were very strong and may have also been potential contributors to Greenland dust. In the Southern Hemisphere, extremely low Holocene dust fluxes are observed in the subtropical South Pacific gyre, a situation that persists today in one of the most oligotrophic and iron-limited regions of the world [Blaun et al., 2008]. This characteristic appears not to have been present during the LGM, though, possibly due to enhanced dust transport from Australasia, but maybe also westward from the South American continent. The southern part of the LGM dust “plume” west of South America (Figure 1) may be an interpolation artifact, as it goes against the prevailing westerly winds and there are few local measurements to constrain these values. In midlatitudes, however, aerosol incursions from coastal areas into these waters have been documented under modern conditions [Huneeus et al., 2006; Saïdé et al., 2012] and one could imagine a scenario where dust from strong LGM sources in northern Chile was picked up by coastal lows and transported far into the Pacific via the Pacific subtropical high. In addition, model results also show a large incursion of South American dust into the southeast Pacific, even in the southern part of South America during LGM conditions [Albani et al., 2014]. Although the effect of Australian and New Zealand sources are visible downwind of the source regions into the SW Pacific, the lack of terrestrial observational data from Australia results in a probable underestimation of dust fluxes over that continent in our data set. The uncertainty of the interpolation process (Figure 1, bottom row) is a good indicator of where the interpolation is robust and where dust flux measurements are sparse and new measurements are most needed to better constrain the dust fields. The sparsity of terrestrial data available for the Holocene is due to the fact that loess deposition is largely limited to glacial periods. New observations from peat bogs [Kylander et al., 2013] in Siberia, North America, and South America are expected to fill this gap. The regions with largest uncertainty during the LGM are now located in the Indian Ocean and the oceanic subtropical Pacific.

In Figure 2 we compare the spatial distribution of our dust flux interpolation with simulated output from the four climate models (CCSM, MIROC, MIROC-ESM, and MRI). To account for biases in the interpolation and the climate models, we have standardized each data set by removing the mean and dividing by the standard deviation of the logarithms of the dust fluxes (dust fluxes are spatially lognormally distributed, see SI Figure S3). In Holocene conditions these include large parts of the world’s oceans, as well as the East Asian dust source regions. During the LGM there is a high confidence level for all major dust source regions except for Australia. Based on these results we suggest that dust models generally overestimate LGM dust deposition over West Africa and underestimate it over North America. All but the CCSM also underestimate Siberian LGM sources. Both MIROC models underestimate LGM South American sources. Finally, there are large discrepancies between all model outputs and the interpolation in the HNLC regions of the oceans. The South Pacific dust deposition especially seems to be underestimated in all models over both periods (see also SI Figures S4 to S7).

To evaluate the initial impact of dust fluxes on the ocean carbon cycle, we forced the PISCES model with our interpolated dust deposition fields, providing estimates of oceanic carbon export for both Holocene and LGM conditions. During both periods, the main HNLC regions (as illustrated by high simulated surface nitrate concentrations, Figure 3a, left column) are the SO and the subarctic North Pacific. Carbon export during both periods is highest where there is a substantial supply of nutrients to surface waters (Figure 3a, right column). During the LGM, the additional iron input from aeolian dust increases surface nutrient consumption and reduces preformed nutrient concentrations in line with increased carbon export by the more efficient biological pump in iron-limited regions, i.e., those with excess nitrate, in the Holocene (Figure 3b). High rates
of LGM dust deposition in the North Atlantic also increased carbon export in this seasonally iron-limited region [Nielsdóttir et al., 2009]. The SO response is heterogeneous, with decreased nitrate and increased carbon export downwind of the South American and Australasian LGM sources, consistent with recent observations [Anderson et al., 2014; Lamy et al., 2014; Martínez-García et al., 2014]. However, there is little iron fertilization from LGM dust in the Antarctic sector of the SO, despite a large reservoir of available macronutrients. Our model results suggest that iron limitation in the Antarctic SO did not change markedly between Holocene and LGM scenarios, due to the preferential zonal transport of dust via the westerlies, which results in very low rates of iron deposition at high southerly latitudes under both Holocene and LGM conditions (see Figure 4 and SI Figure S8).

Previous studies have suggested that about half the observed Holocene-LGM difference of 80–100 ppm in the atmospheric pCO₂ concentrations were due to a stronger stratification of the Antarctic SO and associated carbon buildup in the deep ocean, isolating CO₂ from the atmosphere [Francois et al., 1997; Fischer et al., 2010; Hain et al., 2010; Sigman et al., 2010; Martínez-Botí et al., 2015]. Much of the remaining

Figure 2. Ratio of standardized global dust fluxes from various climate models with the standardized interpolated dust fluxes. The color scale shows powers of 10.
The difference has been attributed to a more efficient biological pump due to increased LGM iron fertilization of the subantarctic SO [Watson et al., 2000; Kohfeld et al., 2005; Jaccard et al., 2013], whereby the solubility pump drawdown may have been compensated to a large degree by a decreased terrestrial biosphere and increased ocean salinity [Sigman and Boyle, 2000]. However, in our 500 year PISCES simulation atmospheric pCO₂ was only reduced by <10 ppmv between the LGM and the Holocene, due to the enhanced dust-induced carbon sequestration via the soft tissue pump (although there is an additional long-term effect discussed below). The small change in atmospheric pCO₂, despite larger carbon export and overall greater absolute dust fluxes in most regions than in previous climate model simulations, suggests that enhanced global iron deposition only had a limited influence on atmospheric pCO₂ levels over centennial timescales. This is likely due to the role of other iron sources in the high-latitude SO [Tagliabue et al., 2014] and/or reduced CO₂ drawdown at low latitudes (see discussion of Figure 4b). Our results are similar to other ocean biogeochemical model simulations that estimate low atmospheric pCO₂ reductions (8–15 ppm) due to increased LGM dust loads [Bopp et al., 2003; Parekh et al., 2006; Tagliabue et al., 2009], although other box model studies still attribute a more substantial role (~35 ppm) to iron fertilization [Hain et al., 2010].

Figure 3. (a) left column: Holocene and LGM surface nitrate concentrations (mmol m⁻³) in color superposed with contour lines showing surface iron fluxes (kg m⁻² yr⁻¹) in powers of 10. (right column) Holocene and LGM annual carbon export (mol C m⁻² yr⁻¹) at 100 m depth. (b) Same as Figure 3a but for the LGM-Holocene difference.
Figure 4 shows dust fluxes integrated over 1 year for the interpolation and the four climate models' (CCSM, MIROC, MIROC-ESM, and MRI) ensemble median, as well as the simulated (PISCES) ocean carbon export fluxes by ocean basin. Under Holocene conditions, the climate model median is within a factor 2 of the interpolated dust fluxes in most regions (Figure 4a). However, the SO and the North Pacific dust deposition are strongly underestimated by the climate models, which is significant since these are the HNLC regions that are most likely to respond to dust variations. Under LGM conditions, the models tend to underestimate dust increases in most regions, notably the North Atlantic, which in our interpolation receives a very large additional dust input from North American sources. Generally, the models tend to perform better in low than in high latitudes [Lambert et al., 2013].

Stronger glacial sources in Australia and New Zealand resulted in increased LGM dust deposition in the South Pacific. But despite the larger size of the ocean basin, the South Pacific only received about half as much dust as the South Atlantic. This suggests that Australian and New Zealand sources produced a lower absolute amount of dust than South American sources.

Basin changes in the impact of dust iron on carbon export between the Holocene and the LGM depend on the role of iron limitation in each region and macronutrient transport by ocean currents. The main source of macronutrients for biological productivity in low latitudes is thought to be intermediate depth waters originating from the SO surface layers [Toggweiler et al., 1991; Sarmiento et al., 2004; Sigman et al., 2010]. Thus, any change in macronutrient concentrations of the subantarctic ocean will be conveyed to other ocean regions [Boyle, 1988; Palter et al., 2005; Tagliabue et al., 2008, 2014]. In PISCES, the subantarctic south Atlantic and south Pacific Oceans are iron limited and we find additional carbon export due to the greater LGM dust supply (Figure 4b). Conversely, non-iron-limited regions such as the tropical oceans show a reduction of carbon export. More efficient biological carbon sequestration in iron-limited subantarctic regions reduces nutrient concentrations in the SO surface waters. Then, as expected, these local nutrient deficits are transported at intermediate depths toward low latitudes, reducing carbon export and thereby CO₂ drawdown when these waters upwell into tropical nitrate-limited regions [Matsumoto et al., 2002;
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