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Dust transport enhanced land surface weatherability in a cooling world

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Abstract

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The weatherability of exposed silicate rocks drives the efficiency of climatic feedback on the geological carbon cycle through silicate weathering. However, the controls and evolution of land surface weatherability are not fully understood. Tectonically induced exposure of fresh silicates can induce a wide range of weatherability, depending on the maturity and lithology of the exhumed rocks. Here, we propose that aeolian dust has potentially been pivotal in sustaining land surface weatherability during global cooling. Our analysis of palaeoclimate simulations shows an additional transport of 1072 ± 69 Tg yr⁻¹ of dust to regions with precipitation of more than 400 mm yr⁻¹ during the Last Glacial Maximum compared to the pre-industrial period. As dust mainly contains fresh minerals with high surface areas, such dust

transport markedly increases land surface weatherability, yielding an additional atmospheric CO_2 consumption of 0.431 ± 0.030 Tmol yr⁻¹, which would offset the reduced silicate weathering induced by weaker climatic forcing. It is suggested that a dustier world could increase global land surface weatherability, leading to a more buffered carbon cycle that sustained low atmospheric CO_2 levels.

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Introduction

Land surface weatherability is the susceptibility of the land surface to chemical weathering (e.g., Kump and Arthur, 1997). Silicate weathering is a key negative feedback that regulates the long term global carbon cycle (e.g., Walker et al., 1981). Therefore, together with the amount of silicate rocks in the critical zone, the weatherability of those silicate rocks has acted as an important variable in silicate weathering and the associated atmospheric carbon dioxide consumption. Variation in land surface weatherability was proposed as the dominant mechanism regulating the long term evolution of atmospheric carbon dioxide levels in the Phanerozoic (e.g., Francois and Walker, 1992; Kump and Arthur, 1997; Caves et al., 2016; Caves Rugenstein et al., 2019). In particular, inverse modelling of marine proxies suggested that the Neogene cooling was driven by a rise in weatherability over time (Caves Rugenstein et al., 2019), which may have been caused by the uplift of the Himalaya-Tibetan Plateau (e.g., Raymo et al., 1988; Kump and Arthur, 1997) and in combination with the exposure of Mg-rich lithologies in East and Southeast Asia (Park et al., 2020; Yang et al., 2021). Nevertheless, all of these studies suggest the dominant control of denudation of the continents on silicate weatherability. However, the temporal evolution of denudation fluxes during the late Cenozoic on the Himalaya-Tibetan Plateau and globe remains controversial (*e.g.*, Métivier *et al.*, 1999; Clift, 2006; Herman *et al.*, 2013; Lenard *et al.*, 2020).

Reconstructing land surface weatherability is not easy because it has always been affected by different factors, e.g., tectonic activity, mountain building, surface lithology, glacier erosion, and vegetation cover (Francois and Walker, 1992). Unlike those proposed influencing factors of weatherability, here we propose that aeolian dust plays a role in sustaining high land surface weatherability in a cooling world. Aeolian dust is transported from arid sources to less arid areas, which can increase global land surface weatherability in two ways. First, the transport of less altered fine grained material to a more humid environment leads to a net increase in land surface weatherability. Second, aeolian dust provides nutrients to the vegetation (Ridgwell, 2002), which further increases silicate weathering rates. To test this idea, here we quantify the increase in dust deposition fluxes to the less arid continental regions and the associated atmospheric CO2 consumption caused by silicate weathering between the preindustrial (PI) period and the Last

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Glacial Maximum (LGM), because global dust cycles in the two periods are only well constrained by palaeo-dust record and climate modelling.

Results and Discussion

We conducted palaeoclimate simulations in our study (Figs. S-1 to S-3), where the preindustrial experiment (called PI-Exp) represents the warm condition of the last few kyr, and the LGM experiment (called LGM-Exp) corresponds to the cold condition ~21 kyr ago. To reveal the uncertainty of modelling data, we also collected another four modelling results from previous studies (Supplementary Information). The mean dust deposition flux over land and standard error of all five data sets are illustrated in Figure 1. We also summed the PI and LGM dust deposition fluxes from each model based on the mean annual precipitation (MAP) ranges (Fig. S-4). All the modelling results showed enhanced LGM dust deposition in terms of flux and spatial extent compared to PI conditions (Figs. 1, S-4). Dust deposition is high in the arid region around deserts in Central and East Asia, North Africa and Australia. However, the simulations clearly show that dust has also been transported to regions where the climate is wetter (Figs. S-1 to S-4). For both PI and LGM simulations, the amount of dust transported to regions with MAP >400 mm and >200 mm accounts for ~31 % and ~47 % of the global dust deposition flux in both simulations, respectively, despite the expansion of the arid regions during the LGM. Global land dust deposition has increased in almost all MAP zones during the LGM (Fig. 2a). The global net increases in aeolian dust (LGM-Exp minus PI-Exp) are 1621 ± 99 Tg yr⁻¹ and 1072 ± 69 Tg yr⁻¹ for regions with MAP >200 mm and >400 mm, respectively.

Dust promotes weatherability. The intensity of dust silicate weathering is strongly dependent on climatic conditions (*e.g.*, Goddéris *et al.*, 2010, 2013). In arid regions, the scarcity of precipitation and vegetation limits the weathering of silicate minerals, as seen by a low degree of Na depletion in silicates (Fig. S-5). In particular, a silicate Na₂O/Al₂O₃ ratio of 0.17 in arid regions

with MAP <200 mm is slightly less than, but close to, that of 0.20 for unweathered upper continental crust (Figs. S-5, S-6). However, when the dust settles in more humid zones, silicates will be more prone to weathering, as seen by the much lower Na₂O/Al₂O₃ ratio (Fig. S-7a). Such dust has even become the main provider of fresh silicates and nutrients to the biosphere in some extreme scenarios where the local supply of fresh rocks is insufficient. For example, on the Hawaiian basaltic bedrock, much of the soil inorganic element originates from China and central Asian deserts (*e.g.*, Kurtz *et al.*, 2001). In addition, the fining of dust during long distance transport produces material with a greater surface area that promotes weathering.

Dust exerts important biogeochemical controls on the downwind terrestrial ecosystems of humid regions (Ridgwell, 2002) because 1) its mineralogy and grain size strongly influence the water- and nutrient-holding properties of the soil, and 2) it is a source of nutrients for some ecosystems. For example, in Amazonia, the soils are already highly weathered and nutrient-depleted, and dust transported across the Atlantic supports the ecosystems (Swap *et al.*, 1992). Some soils on the Hawaiian Islands suggest an analogous situation (Chadwick *et al.*, 1999). Therefore, *via* the fertilisation of vegetation, dust settling in areas with such highly weathered soils facilitates the silicate weathering process.

Weathering of aeolian dust is also pronounced in broad, less humid regions. Rainfall composition provides a valuable approach for addressing dust weathering kinetics. Rainfall compositions in less humid North China show high ⁸⁷Sr/⁸⁶Sr and low Ca/Sr ratios (Rao *et al.*, 2017), corresponding to fast dissolution of some silicates. Dust silicate weathering can also be found in Himalayas with heavy monsoon rainfall. The precipitation composition in Bangladesh and Nepal (Galy and France-Lanord, 1999) can be well explained by the mixing of silicate with low Ca/Sr ratios and Himalayan carbonate and sea salts with high Ca/Sr ratios. This evidence suggests that part of the dust is rapidly weathered in the atmosphere or shortly after deposition, increasing the weatherability.



Figure 1 Mean land dust deposition fluxes (a, b) and their standard error (c, d) in experiments of preindustrial (PI) and Last Glacial Maximum (LGM) periods integrated from five climate model results (see Methods).



Figure 2 Increases in land dust deposition (a), efficiency (b) and flux (c) of atmospheric CO_2 consumption by silicate weathering in the LGM compared with the PI categorised by mean annual precipitation (MAP). Note that increases in CO_2 consumption efficiency (a measure of the amount of CO_2 consumed by silicate weathering *per* kilogram rock) and flux in each zone with MAP >200 mm are estimated from the differences in CO_2 consumption efficiency and flux between those zones and the arid region (MAP <200 mm). Error bars show one standard error of the mean.

In addition, dust weathering involves not only silicates but also carbonates (Goddéris *et al.*, 2010). Due to abundant carbonate in dust and its rapid weathering rates in Mississippi loess, dolomite weathering can contribute to as high as 90 % CO₂ consumption of overall dust weathering (Goddéris *et al.*, 2010, 2013). Carbonate weathering can consume CO₂ on time scales shorter than a million years, and dust transport thus enhances the overall continental weatherability through this short term CO₂ consumption mechanism as well.

Dust promotes atmospheric CO₂ consumption by silicate weathering. Global continental weatherability has been elevated by dust transport to humid regions in multiple ways, but only atmospheric CO₂ consumption by silicate weathering can be estimated because the biogeochemical impact of dust and climate modulated dust carbonate weathering rate are difficult to assess quantitatively. In the context of dust silicate weathering, with an increase in MAP, the degree of silicate weathering alternation has increased, as shown by the decreasing Na₂O/Al₂O₃ ratio and the increasing long term atmospheric CO₂ consumption efficiency (a measure of the amount of CO₂ consumed by silicate weathering per kilogram rock) (Fig. S-7). We use the dust silicate weathering and the associated CO₂ consumption efficiency in regions with MAP <200 mm as a base value to evaluate the increases in dust deposition flux and the associated atmospheric CO₂ consumption in each climate zone with MAP >200 mm (Supplementary Information, Fig. 2). The additional long term atmospheric CO₂ consumption flux by silicate weathering yielded 0.431 ± 0.030 Tmol yr⁻¹ and 0.503 ± 0.037 Tmol yr⁻¹ for regions with MAP >400 mm and MAP >200 mm during the LGM period, respectively. Such fluxes are significant and represent 15-19 % of modern day long term global continental silicate weathering (Gaillardet et al., 1999). On the other hand, the cooling by

4.4–6.8 °C from PI to LGM conditions used in our modelling would at least lower modern day silicate weathering by 23 to 35 %, depending on the type of lithology and parametric laws used (West, 2012; Li *et al.*, 2016). Our calculation shows that the increase in weatherability related to enhanced dust production and transport under the LGM is likely to be as important as the effect of global temperature decrease on atmospheric CO_2 consumption by silicate weathering, which may sustain less variable silicate weathering flux into the ocean at glacial-interglacial time-scales (*e.g.*, von Blanckenburg *et al.*, 2015).

Implications

Our study suggests that a dustier world in glacial periods comes with higher land surface weatherability during the late Neogene cooling, where higher land surface weatherability caused by aeolian dust can have an enhanced ability to buffer the carbon cycle fluctuation. A dusty world may have appeared in the Neogene, because modern major global dust source areas, e.g., North Africa and the broad Asian arid region, were formed during the Neogene (Guo et al., 2002; Zhang et al., 2014). Pre-Quaternary global dust deposition is difficult to constrain due to a paucity of robust boundary conditions (e.g., vegetation) and palaeo-dust reconstructions. The global dust emission of the mid-Pliocene (3.3–3.0 Ma, a warm period prior to Quaternary glacial cycles) was only half or even less of that in the PI period (Shi et al., 2011; Sagoo and Storelvmo, 2017), which may suggest less dust deposition in warming periods during the late Neogene on timescales of millions of years. This aeolian dust-driven high degree of weatherability in a cooling world may provide an independent explanation for the overall low levels of atmospheric CO₂ in the Neogene and similar cooling stages during the deep past, but it requires to be further examined by more robust palaeo-dust reconstruction and modelling.

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Additional Information

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Supplementary Information

The Supplementary Information includes:

- > Methods
- ➢ Figures S-1 to S-7
- Supplementary Information References

Methods

Dust model performed in this study

In this study, the climate model Community Earth System Model version 1.2.2 (CESM1.2.2; Hurrell *et al.*, 2013) is used, which was developed and is maintained by the National Center for Atmospheric Research. CESM1.2.2 can simulate the present climate well (Knutti *et al.*, 2013) and has also been widely used in palaeoclimate research (*e.g.*, Liu *et al.*, 2020; Zhang *et al.*, 2021). In this model, the atmospheric component, Community Atmosphere Model version 5 (CAM5), is run at a horizontal resolution of ~1.9° (latitude) $\times 2.5^{\circ}$ (longitude) with 30 levels in the vertical dimension, and each cell considers the emission, transport and deposition of dust (Ganopolski *et al.*, 2010). The land module, Community Land Model version 4.0 (CLM4.0), is run at the same horizontal resolution as CAM5.

Both palaeoclimate records and climate models suggest that the global mean surface temperature during the LGM was 4.4–6.8 °C lower than that in preindustrial times (Tierney *et al.*, 2020; Osman *et al.*, 2021) and that the land ice sheet was much greater during the LGM (Peltier *et al.*, 2015). These two points are considered in LGM-Exp. The orbital parameters and the levels of greenhouse gases (GHGs) during the LGM follow the Palaeoclimate Modelling Intercomparison Project phase 3 (PMIP3) protocol. The vegetation cover for the LGM-Exp corresponds to a modified PI-Exp vegetation cover according to the response of vegetation phenology to climate change based on the active CN biogeochemical cycle (Lawrence *et al.*, 2011).

In PI-Exp, the sea surface temperatures (SSTs) and sea ice fractions correspond to the default values of the CESM. LGM-Exp is run with the SSTs and sea ice fractions resulting from the equilibrium of a coupled run with the settings of Zhang *et al.* (2022). PI-Exp and LGM-Exp were run for 20 years, and only the data corresponding to the last 10 years are analysed and presented here.



On land, the annual mean precipitation from North Africa to inland Asia is less than 200 mm in PI-Exp, where the dust deposition fluxes are the greatest across the world (Fig. S-1a). This dust deposition pattern is also consistent with modern observations (Jickells *et al.*, 2005). As global cooling occurred during the LGM period, the land area with annual mean precipitation less than 200 mm expanded poleward in the Northern Hemisphere, and the global dust deposition fluxes increased obviously (Fig. S-1b). A drier and dustier world during the LGM is in accordance with previous simulations (*e.g.*, Lambert *et al.*, 2015, 2021) and geological reconstructions (*e.g.*, Lorius *et al.*, 1984; Maher *et al.*, 2010). We also modelled the near-surface winds in the two experiments (Figs. S-2, S-3), showing that the winds can transport dust out from arid regions (<200 mm and <400 mm MAP).

Collection of land dust deposition from other models

Our simulations produced just a single realisation of PI and LGM dust deposition fields. To estimate the uncertainty of the dust deposition fields, we also compiled paired PI and LGM dust deposition fluxes from four previously published studies (Takemura *et al.*, 2009; Yukimoto *et al.*, 2012; Albani *et al.*, 2014; Lambert *et al.*, 2015). Note that we chose a subset of available palaeoclimatic dust simulations. Because the observed modern global dust deposition is less than 3500 Tg yr^{-1} and there is a much larger LGM dust flux derived from palaeo-reconstructions (Maher *et al.*, 2010), those models with PI-derived global dust deposition >3500 Tg yr⁻¹ or with a dust deposition ratio of LGM/PI < 2 are not included in our compilation.

Calculation of atmospheric CO₂ consumption by silicate weathering

To quantitatively assess the atmospheric CO_2 consumption by dust silicate weathering, we established a transformation equation from the degree of silicate Na depletion to atmospheric CO_2 consumption. The degree of Na depletion is a useful proxy for tracing silicate weathering intensity because plagioclase is the most common silicate mineral prone to weathering in the upper continental crust. Na depletion in silicates expressed by Na₂O/Al₂O₃ or τ_{Na} in soil or surface sediments shows a clear dependence on MAP at continental or global scales (Fig. S-5), regardless of lithology, suggesting that it is useful to trace the degree of dust silicate depletion in each MAP zone globally.

First, we establish a transformation equation from the Na₂O/Al₂O₃ silicate of surface soil to long term atmospheric CO₂ consumption *per* kilogram rock (hereafter atmospheric CO₂ consumption efficiency, mol kg⁻¹). Here, the CO₂ consumption efficiency is calculated by the chemical difference between soil and parent rock following France-Lanord and Derry (1997):

$$CO_2$$
 consumption efficiency = $\Delta Ca + \Delta Mg + 0.10 \Delta K + 0.15 \Delta Na$,

where Δ shows the difference in base cations (Ca, Na, K, and Mg) between soil and parent rock. Note that the CO₂ consumption efficiency in this study reflects the atmospheric CO₂ consumption *per* kilogram rock on million-year timescales because only a fraction of the alkalinity is associated with alkalis (20 % for K⁺ and 30 % for Na⁺) (France-Lanord and Derry, 1997), thus providing a lower-limit estimate. Similar to Yang *et al.* (2021), we use loess and palaeosol samples in arid northern China and three average compositions of typical red soil datasets in humid southern China as well as the upper crust composition of central East China to yield a linear regression between Na₂O/Al₂O₃ and CO₂ consumption efficiency (Fig. S-6). These data were collected from a range of climatic settings and have an excellent linear distribution, suggesting a close link between the degree of Na depletion and the overall depletion of base cations in silicates.



Second, by using a recently reported dataset of surface soil Na₂O/Al₂O₃ ratios across China, we recalculated the soil Na₂O/Al₂O₃ ratio distribution in each MAP zone (Fig. S-7a). By using the transformation equation from Na₂O/Al₂O₃ to CO₂ consumption efficiency, we are able to estimate the CO₂ consumption efficiency in each MAP zone (Fig. S-7b). The increase in CO₂ consumption efficiency when dust is transported to less arid regions should correct CO₂ consumption efficiency in arid regions. Thus, the increase in CO₂ consumption efficiency in each less arid region is estimated to be the difference in CO₂ consumption efficiency between each region with MAP > 200 mm and the arid zone with MAP < 200 mm (Fig. 2b). Based on the increase in dust deposition fluxes in each MAP zone (Fig. 2a), the increase in CO₂ consumption flux by dust silicate weathering in each MAP zone (MAP > 200 mm) can be calculated (Fig. 2c).

Due to a lack of data, the dataset of surface soils in China is used to obtain a transformation equation for a global estimate, which inevitably yields bias. However, such a bias may be insignificant. First, the dataset includes 1996 surface soil composition data across variable lithology and climate settings (Guo *et al.*, 2022), thus reflecting a continental-scale Na₂O/Al₂O₃ distribution. Second, plagioclase is widely seen in granitic and basaltic rocks, which suggests that Na/Al can be widely used to trace silicate weathering in variable lithologies. Third, the similar mean composition of the upper continental crust between China and other commonly used estimates from different continents (Gao *et al.*, 1998) suggests that data from the Chinese continent are suitable for conducting a global estimate. More importantly, what we try to estimate is indeed the difference in the silicate Na₂O/Al₂O₃ ratio between the less arid region with MAP > 200 mm and the arid region with MAP < 200 m, which should be less influenced by the absolute value of Na₂O/Al₂O₃ in different types of parent rocks. For example, Yang *et al.* (2021) evaluated the lithology impact, and they used different Na₂O/Al₂O₃ ratios of upper continental crust ranging from 0.20 to 0.25 as parent rocks to produce a linear regression line between Na₂O/Al₂O₃ and CO₂ consumption efficiency in surface soil. They found that the estimated difference in CO₂ consumption efficiency value in each regression exhibits slightly larger variabilities.

Supplementary Figures



Figure S-1 Modelled land dust deposition (colour gradient, in g $m^{-2} yr^{-1}$) in experiments of (a) preindustrial (PI) and (b) Last Glacial Maximum (LGM) periods conducted from this study. Red and blue dots mark regions with mean annual precipitation (MAP) of <200 mm and 200–400 mm, respectively.





Figure S-2 Annual mean precipitation (shaded: orange, MAP < 200 mm; yellow, MAP = 200–400 mm) and four seasonal mean winds (vectors; units: $m s^{-1}$) near the surface of PI-Exp from this study. (a) DJF, (b) MAM, (c) JJA, (d) SON.



Figure S-3 Annual mean precipitation (shaded: orange, MAP < 200 mm; yellow, MAP = 200–400 mm) and four seasonal mean winds (vectors; units: $m s^{-1}$) near the surface of LGM-Exp from this study. (a) DJF, (b) MAM, (c) JJA, (d) SON.



Figure S-4 Mean land dust deposition in experiments of preindustrial (PI) and Last Glacial Maximum (LGM) periods. (a) Mean land dust deposition in each MAP range; (b) accumulation of land dust deposition from low to high MAP. Error bars in (a) mark one standard error of the mean, and statistics are compiled from five dust deposition datasets (see Methods).



Figure S-5 Correlation of the degree of silicate Na depletion in surface soils with MAP. (a) Mean Na₂O/Al₂O₃ ratio of bulk surface soil across China in each MAP zone (data from Guo *et al.*, 2022, MAP division follows Fig. 2). Error bars mark one standard error of the mean. (b) τ_{Na} in global soils with MAP (data from Brantley *et al.*, 2023). τ_{Na} shows differences in the Na/Zr ratio between soil and parent rock, and a more negative τ_{Na} indicates a higher degree of Na depletion.



Figure S-6 Correlation between Na₂O/Al₂O₃ and long-term CO₂ consumption efficiency for loess and palaeosol samples (yellow dots) in northern China and three average compositions of typical red soil (red dots) datasets in southern China. The upper crust composition of East China (black pentagram; Gao *et al.*, 1998) is chosen as the parent rock. All data are from Yang *et al.* (2021), and the calculation of CO₂ consumption efficiency can be found in Methods. These data form an excellent fit line with 95 % confidence (shading area), which thus yields a transfer equation from silicate Na₂O/Al₂O₃ to long term CO₂ consumption efficiency in Figure S-7.



Figure S-7 (a) Averages of Na_2O/Al_2O_3 in bulk surface soil across China categorised by mean annual precipitation (data are from Guo *et al.*, 2022) and (b) associated CO₂ consumption efficiency calculated by the equation in Figure S-6. Error bars mark one standard error of the mean.

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