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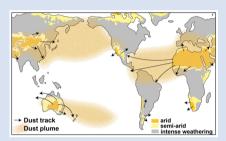
# Dust transport enhanced land surface weatherability in a cooling world

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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  $1072 \pm 69 \, \mathrm{Tg} \, \mathrm{yr}^{-1}$  of dust to regions with precipitation of more than 400 mm yr<sup>-1</sup> 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  $0.431 \pm 0.030$  Tmol yr<sup>-1</sup>, 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  $1621 \pm 99 \text{ Tg yr}^{-1}$ and  $1072 \pm 69$  Tg yr<sup>-1</sup> 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 Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> 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  $\rm Na_2O/Al_2O_3$  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 <sup>87</sup>Sr/<sup>86</sup>Sr 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.

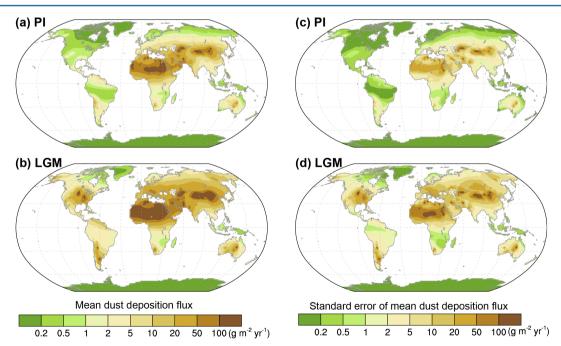


Figure 1 Mean land dust deposition fluxes (a, b) and their standard error (c, d) in experiments of preindustrial (PI) and Last Glacial Maximum (LGM) periods integrated from five climate model results (see Methods).



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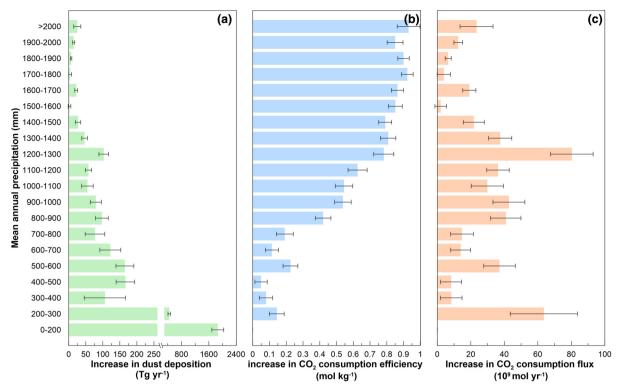


Figure 2 Increases in land dust deposition (a), efficiency (b) and flux (c) of atmospheric CO<sub>2</sub> consumption by silicate weathering in the LGM compared with the PI categorised by mean annual precipitation (MAP). Note that increases in CO<sub>2</sub> consumption efficiency (a measure of the amount of CO<sub>2</sub> consumed by silicate weathering *per* kilogram rock) and flux in each zone with MAP >200 mm are estimated from the differences in CO<sub>2</sub> 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 %  $\rm CO_2$  consumption of overall dust weathering (Goddéris *et al.*, 2010, 2013). Carbonate weathering can consume  $\rm CO_2$  on time scales shorter than a million years, and dust transport thus enhances the overall continental weatherability through this short term  $\rm CO_2$  consumption mechanism as well.

Dust promotes atmospheric CO<sub>2</sub> consumption by silicate weathering. Global continental weatherability has been elevated by dust transport to humid regions in multiple ways, but only atmospheric CO<sub>2</sub> 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 Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> ratio and the increasing long term atmospheric CO<sub>2</sub> consumption efficiency (a measure of the amount of CO<sub>2</sub> consumed by silicate weathering per kilogram rock) (Fig. S-7). We use the dust silicate weathering and the associated CO<sub>2</sub> consumption efficiency in regions with MAP <200 mm as a base value to evaluate the increases in dust deposition flux and the associated atmospheric CO<sub>2</sub> consumption in each climate zone with MAP >200 mm (Supplementary Information, Fig. 2). The additional long term atmospheric CO<sub>2</sub> consumption flux by silicate weathering yielded  $0.431 \pm 0.030$ Tmol  $yr^{-1}$  and  $0.503 \pm 0.037$  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<sub>2</sub> consumption by silicate weathering, which may sustain less variable silicate weathering flux into the ocean at glacial-interglacial timescales (*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 Storelymo, 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 CO<sub>2</sub> 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**

**Supplementary Information** accompanies this letter at https://www.geochemicalperspectivesletters.org/article2322.



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