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Aerosol-Climate Interactions During the Last Glacial Maximum

Samuel Albani¹ · Yves Balkanski¹ · Natalie Mahowald² · Gisela Winckler^{3,4} · Valter Maggi⁵ · Barbara Delmonte⁵

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Abstract

Purpose of Review Natural archives are imprinted with signs of the past variability of some aerosol species in connection to major climate changes. In certain cases, it is possible to use these paleo-observations as a quantitative tool for benchmarking climate model simulations. Where are we on the path to use observations and models in connection to define an envelope on aerosol feedback onto climate?

Recent Findings On glacial-interglacial time scales, the major advances in our understanding refer to mineral dust, in terms of quantifying its global mass budget, as well as in estimating its direct impacts on the atmospheric radiation budget and indirect impacts on the oceanic carbon cycle.

Summary Even in the case of dust, major uncertainties persist. More detailed observational studies and model intercomparison experiments such as in the Paleoclimate Modelling Intercomparison Project phase 4 will be critical in advancing the field. The inclusion of new processes such as cloud feedbacks and studies focusing on other aerosol species are also envisaged.

Keywords Aerosol · Mineral Dust · Last Glacial Maximum · Iron Fertilization · Radiative Forcing

Introduction

Aerosols are a key component of the climate system; yet their impacts on climate are still characterized by a high degree of uncertainty, because of the variety in physical and chemical composition, the complexity of their interactions, and the large spatial and temporal variability of emissions and dispersion [1, 2]. The temporal variability in aerosol emissions is imprinted

in both instrumental and paleoclimate records across a variety of time scales, including glacial-interglacial [3].

Aerosols directly impact the atmosphere radiation budget by the reflection and absorption of solar and terrestrial radiation. Depending on their size and chemical composition, specific aerosol species cause a net positive (black carbon) or negative (sulfates) direct radiative forcing, while other aerosol species have more complex net effects in different contexts [4]. Aerosols also affect climate by acting as cloud condensation nuclei (CCN) and ice nuclei (IN), thus affecting cloud lifetime and albedo [5, 6]. Absorbing aerosols can also impact cloud formation by heating, which can reduce relative humidity hence the liquid water path and/or cloud cover (semi-direct effect) [7, 8]. Finally, aerosols affect heterogeneous chemistry in the atmospheric environment [9]. Aerosol deposition to the surface can also impact climate. Snow albedo can be diminished by the presence of dust and especially black carbon (BC) [10, 11]. Last but not least, aerosols contribute elements such as phosphorus and iron to the terrestrial and marine biosphere, that can alter different global biogeochemical cycles and in turn the global carbon budget and climate [12].

Wind stress on the surface is tightly linked to the emissions of the most abundant natural primary aerosols. Mineral (desert) dust originates by eolian erosion from arid and semi-arid areas with low vegetation cover [13, 14], whereas

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inorganic sea salts (mostly NaCl) are emitted as sea sprays at the air–water interface within breaking waves via bubble bursting and by tearing of wave crests [15], along with organic components [16]. Seawater brine and frost flowers forming on the surface of sea ice are a potentially important source of fractionated sea salt species, as well as organic matter [17, 18]. Land vegetation is also an important source of primary aerosols, either as primary biological aerosol particles (PBAPs) such as viruses, bacteria, fungal spores, pollen, plant debris, and algae [19, 20] or in the form of black carbon and particulate organic matter emitted via biomass burning (vegetation fires) [21]. Volcanic eruptions also emit ash [22]. Secondary aerosols can form from gaseous precursors: wetland emissions of ammonia, biogenic emissions of volatile organic compounds (BVOC) from terrestrial vegetation and biomass burning [23], and sulfate compounds from volcanic eruptions (SO₂) and marine biota (dimethyl sulfide) [17]. Natural nitrate aerosols are formed from sources of NO_x such as biomass burning, biogenic soil emissions, lightning, and stratospheric injection [24].

There is sometimes confusion about aerosol naming conventions. Different categorizations are possible, depending on [1] source process [2] chemical characterization as an airborne species or [3] operational definition when measured from sediments for the paleoclimate record. Here, we choose to organize the discussion around the potential of accumulation and measurement in paleoclimate archives.

Changing climate conditions that affect winds, the hydrological cycle and vegetation will affect aerosol emissions [25]. This is of interest for future climate change, where non-fossil fuel aerosol emissions can change along with direct anthropogenic emissions [26, 27], and it is relevant for the attribution of changes in the framework of the Intergovernmental Panel on Climate Change (IPCC). This is also relevant for climate, because of aerosols' feedback on the climate system [2, 28], as well as for public health [29, 30]. Past climate changes will also have caused differences in aerosol emissions, as evidenced by natural archives [31]. In the context of climatic changes, simulations of the past climate constitute a test bed for global Earth System Models (ESM), including for aerosols such as dust [32].

Driven by changes in the amount and distribution of incoming solar radiation, and mediated by internal feedbacks in the climate system, the last 100,000 years long glacial period culminated ~21,000 years before present (21 ka BP) during the last glacial maximum (LGM). The LGM was characterized by a significant drop in temperatures of several degrees, more markedly at high latitudes, associated to a massive reduction in the concentration of greenhouse gases compared to pre-industrial values [33]. Extensive ice sheets covered North America and West Eurasia, and the sea level was 120 m below the present level, associated with a reorganization of atmospheric and oceanic circulation, the hydrological

cycle, and ecosystems [34]. As we describe later, there is ample evidence that natural aerosol emissions were profoundly impacted by such climatic changes, in particular by changes in vegetation, glacial erosion and sea ice cover, and winds. Therefore, the LGM is an ideal target to explore natural aerosols (and dust in particular) interactions with climate.

Climate archives such as ice cores, marine sediments, lakes, peat bogs, and soil and loess profiles (Table 1) contain most of the information available about past aerosols. To interpret a paleoclimate record, the aerosol (or proxy) deposition flux to the surface of the archive needs to be connected to atmospheric concentrations of aerosols, and we need to understand how much the record is representative of larger scale patterns on different temporal and spatial scales [57]. Prognostic aerosol models represent a good example of process-based modeling—the modeling of a variable as it could be retrieved from a climate proxy [58]; in order to (almost) directly compare models with observations, the variable of interest is deposition flux, which for a meaningful model should include both dry and wet deposition processes [59].

Most of the information we have is about desert dust; dust is insoluble and, while it undergoes aging, it can be conserved to some extent in several natural archives from close to the source areas (e.g., loess) to remote sinks into the ocean or polar ice sheets [60]. Ice cores also preserve information about sea salt, sulfur and nitrogen species deposited to the surface [33]. Ice core data on carbonaceous aerosols is more limited, although some complementary information can be gathered, in terms of paleofire proxies, from charcoal, which offers chances of constraining Earth system models [56]. However, ice cores are limited in spatial coverage.

Some observations (Table 1) refer to the aerosol directly (e.g., particle counter dust measurements) or semi-directly (e.g., ²³²Th for dust), and may be also used as a quantitative constraint of the aerosol mass in the past; other observations provide information about a process or a combination of processes, in which case we have a paleoclimate proxy in a traditional sense (e.g., ammonia for fires). In yet other cases both perspectives hold, e.g., desert dust is both a tracer and an agent of climate change. Besides determining the physical, chemical, and optical properties of aerosols, their mineralogical and/or elemental composition (including the isotope composition) may provide very useful insights onto geographical provenance and genesis processes (Table 1), as exemplified by dust radiogenic isotopes [33, 59].

Uncertainties from present day estimates of aerosols will propagate into the understanding of paleoclimate; for instance, modeling uncertainties in the parameterizations of cloud nucleation or the prescribed intrinsic optical properties of dust; or uncertainties in relating air–snow transfer of tracers. On the other hand, information from paleoclimate archives can also help to clarify some of these issues, and indeed provide a test for those models aiming to predict future climate change.

Table 1 Summary of the information on past aerosol from natural archives, relevant for the last glacial maximum

Type of aerosol	Natural archive	Measured property	Derived property	Comments and example reference
Mineral dust	Ice cores	Insoluble dust number and volume concentration (Coulter counter, laser optical counters)	Dust mass accumulation rates (MAR) and associated particle size distributions	The most direct measurement of dust, in “minimally disturbed” analytical conditions [35–37]
		Non sea salt calcium	Dust MAR	Proxy for dust, with uncertainties related to variable dust composition [38]
	Marine sediments	Radiogenic isotope composition (Nd, Sr, Pb, Hf); rare earth elements (REE); relative abundance of major elements; mineralogy; and magnetic properties	Provenance fingerprinting	Spatial variability and limited sampling in potential source areas can be a limiting factor [39–42]
		Dust extinction and/or scattering (optical methods)	Shape and size-resolved intrinsic optical properties	Only a couple of pilot studies [43]
Sea salts	Ice cores	Mass of residual sediment after selective removal of opal and carbonates (operationally defined fraction)	Dust MAR	Usually used in combination with age-model-derived mass accumulation rates; no focusing corrections [44]
		Geochemical lithogenic proxies (e.g., Al, Ti, ^{232}Th , ^4He) concentration in sediment	Dust MAR	Usually used in combination with age-model-derived mass accumulation rate or, in the case of ^{232}Th , a constant flux-based mass accumulation rate ^{230}Th -excess, ^3He), to correct for sediment focusing [45–47]
		Laser optical counters	Particle size distributions	Use of dispersants: size may not be fully representative of atmospheric deposition [48]
		Radiogenic isotope composition (Nd, Sr, Pb); REE; $^4\text{He}/\text{Th}$	Provenance fingerprinting	Spatial variability and limited sampling in potential source areas can be a limiting factor [49]
	Loess deposits	Radiocarbon or luminescence dating of soil horizons	Dust MAR	Reliability highly dependent on geomorphological settings and dating protocols [50, 51]
		Laser optical counters	Particle size distributions	Use of dispersants: size may not be fully representative of atmospheric deposition [52]
	Ice cores	Radiogenic isotope composition (Nd, Sr, Pb); REE	Provenance fingerprinting	Spatial variability and limited sampling in potential source areas can be a limiting factor [52]
		Major ions, e.g., Na^+	Sea salt from open water and sea ice	Uncertainty in disentangling the two sources and interpreting the signal [53]
	Ice cores	SO_4^{2-} and MSA	Sulfates; biogenic activity, volcanoes, and sea salt; MSA; biogenic emissions	Preservation issues for MSA. Very limited data, difficulties in interpreting the signal [54]
		NO_3^- and NH_4^+	Nitrate aerosol from lightning, fires, and biogenic emissions	Contamination and preservation issues. Very limited data, difficulties in interpreting the signal [54]
Nitrogen species	Ice cores		Biogenic emissions, fires	Contamination and preservation issues. Very limited data, difficulties in interpreting the signal [55]
Carbonaceous aerosols	Ice cores	Organic carbon, formate, levoglucosan NH_4^+	Biogenic emissions, fires	Contamination and preservation issues. Very limited data, difficulties in interpreting the signal [55]
	Lakes sediments, peats, and soil	Charcoal	Source location of paleo fires, type of burning regime	Geographical limitations in identifying the sources. Uncertainties in linking fires to type and amount of aerosol species [56]

Next, we will review the information arising from natural archives, and the state of the art in explaining the variability in LGM aerosol and understanding their feedback on climate, by targeting key scientific questions that have sparked research for the last decades or that are prone to become hot topics in the near future.

Climate Impacts on Aerosol Emissions

In this section, we review the information from paleoclimate archives concerning the major natural aerosol species documented for the LGM. For each aerosol species, we provide a brief description of the specific characteristics and the potential issues concerning the preservation of the depositional signal and estimates of the mass accumulation rates; we also describe the picture emerging from the specific paleoclimate records, as well as possible causes for observed changes in aerosol load/deposition rates.

Mineral Dust

Mineral dust is entrained in the atmosphere by wind erosion of sparsely vegetated soils or loose surface sediments [61]. It is composed mostly of silicates, along with carbonates and metal oxides, in the clay and fine silts dimensional range [62]. Aggregates have been observed [62], but their features are poorly understood, documented in the observational record, and represented in models [63, 64]. Dust composition, shape, and size combine to determine the intrinsic optical properties (mass extinction efficiency, single scattering albedo, asymmetry factor), which determine dust interaction with radiation [43, 65]. Pure mineral dust can be considered almost insoluble. This feature makes it a stable tracer that can be preserved in different environmental matrices, therefore potentially allowing the reconstruction of paleodust records from several natural archives (Table 1), including ice cores [35, 36], marine sediments [47, 66], loess/paleosol sequences [50, 52].

Extracting a paleodust record requires (1) preservation of the deposition signal, (2) the possibility to establish a chronology and (3) the possibility to isolate the eolian fraction, either operationally or geochemically, from the environmental matrix and other terrigenous components. In addition, it is necessary to verify that (4) the sediment accumulation rate is representative of the deposition flux to the surface [59].

Paleodust archives worldwide show a generalized two- to fivefold increase in dust deposition in the LGM compared to the Holocene [60], more marked at high latitudes [66–68]. Reconstructions from polar ice cores consistently show an order of magnitude variability in dust deposition flux between different climate states [35, 53]. These extreme variations are particularly evident in the Greenland ice core records [53, 69]. The cold climate—high dust relation holds over several

glacial interglacial cycles, as well as for millennial scale variability within glacial climates and during the last deglaciation [35, 47, 70, 71].

Several hypotheses have been proposed to explain glacial interglacial changes in paleodust records. Increased aridity [72, 73] and gustiness [51, 74, 75] are widespread conditions that could have enhanced dust emissions during glacial climate, when a general reduction of the hydrological cycle could also reduce wet scavenging and increase dust lifetimes [76, 77]. Additional mechanisms active at a more regional level could cause a characteristic geographic signature in specific paleodust archives in different geographical settings [78]. Source erodibility could be enhanced by reduced vegetation cover, linked to regional drying [79], and possibly reduced plant fertilization by CO₂ [80], and especially by increased sediment availability through glacial processes [39, 81], which is a well-established source of LGM dust at least in the Southern Hemisphere [82, 83], possibly also in connection to the exposure of continental shelves by lowered sea levels [40]. From an atmospheric transport perspective, reorganization of the atmospheric circulation between mid and high latitudes [35, 84], shifts in the intertropical convergence zone [85], and changes in the monsoonal variability [48] all contributed to shape the regional patterns of dust deposition in the LGM. In general, the LGM saw an expansion of mid and high latitude dust sources.

Global data compilations [59, 60, 80, 82, 86–89] allow for a generalized and consistent view of the geographical variability of the dust cycle, as well as constraining models, by comparing deposition fluxes from the models and mass accumulation rates for the observations in the same size range [86]. Model simulations including changes in dust sources (Fig. 1a) can be used in climate change experiments. The LGM climate constitutes an excellent test for dust; a model's spatial continuum can support the interpretation of paleodust archives [57], and yield quantitative estimates of the mass budget of the global dust cycle (Fig. 1c). Currently, only a few models, with diverse mechanisms to account for changes in dust sources, and diverse levels of validation against modern and paleodust data, have tried to simulate the LGM dust cycle [77, 82, 86, 90–98]; model emissions (loading) range from ~2400 to ~16,100 Tg a⁻¹ (23 and 71 Tg) for the LGM, and between ~1100 and ~7100 Tg a⁻¹ (8 and 36 Tg) for the corresponding pre-industrial/current climate control cases, with a median increase by a factor 2.0 (1.9) in the LGM (Fig. 1b, d).

The large spread is attributable to differences in the representation of dust emissions and deposition mechanisms, including from the dust schemes themselves, as well as differences in boundary conditions (including vegetation), the consideration of different aerosol size ranges, and lastly whether or not glaciogenic sources of dust (derived from glacier erosion) [81] were included. Each of these aspects contributes to the variability. Examples from specific studies illustrate and

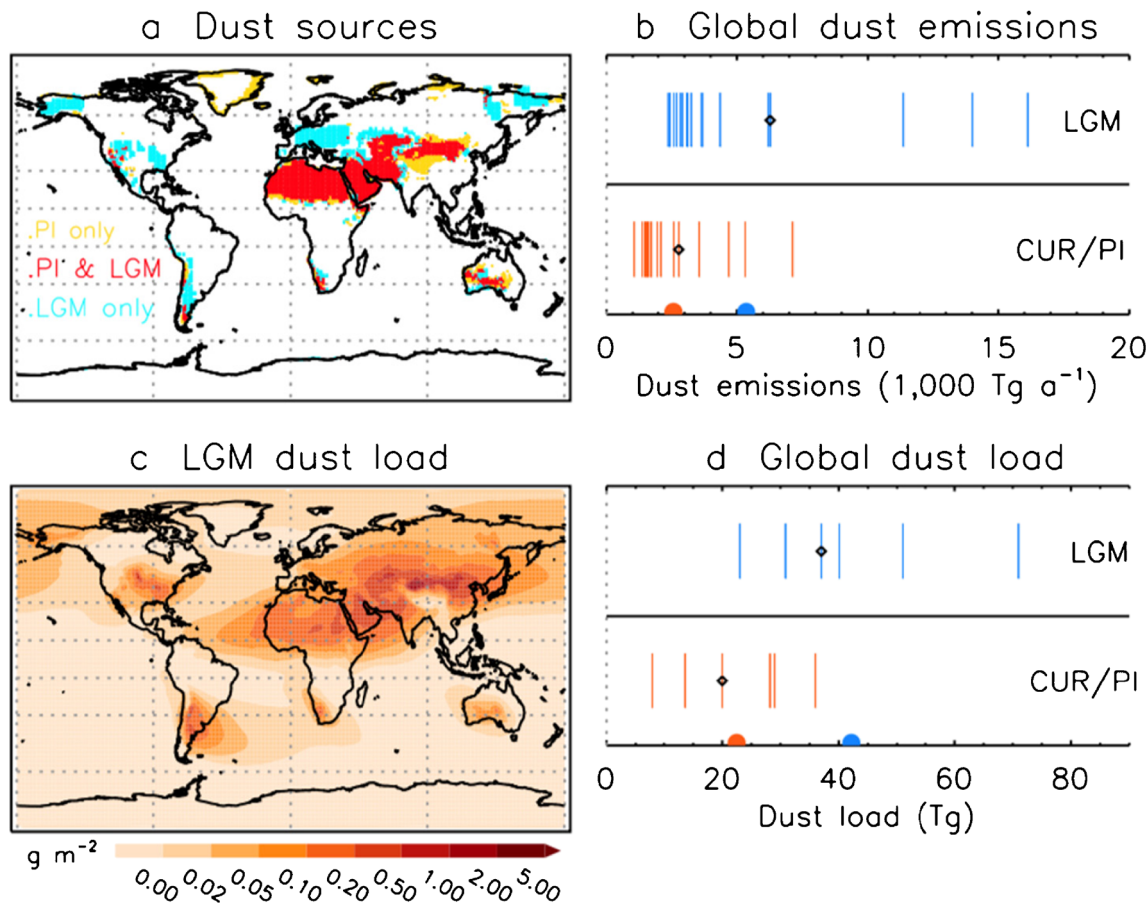


Fig. 1 **a** Map of active sources for dust emissions in the LGM and pre-industrial (PI) conditions [90]. **b** Comparison of global mass budgets of dust emissions for LGM (upper panel, blue) and corresponding PI or CUR (PI/CUR) control (lower panel, red) for different climate model simulations [77, 82, 86, 90–97]. **c** Example map of dust load in the LGM [86]. **d** Global mass budget of dust load for LGM (upper panel,

blue) and PI/CUR control (lower panel, red) simulations [77, 86, 90, 91, 93–95]. The black diamonds in **b** and **d** highlight the simulations displayed in **a** and **c** [86, 90]. The semi-circles on the x-axis in **b** and **d** mark the average LGM (blue) and control (red) of the respective model ensembles. The vertical gray dotted lines mark the zero value on the x-axis

quantify the impacts of, e.g., allowing for source changes and using different emission schemes [80, 82, 95]. Future improvements anticipated to positively impact the representation of the dust cycle include the representation of physical climate boundary conditions, vegetation cover [91, 95], and glaciogenic sources [82, 86]. Dust sensitivity experiments planned for PMIP4 will provide an excellent opportunity to coherently compare LGM dust simulations for the first time [99]. Importantly, by exploiting the concept of process-based modeling, dust can be also viewed as a tracer and used as an indirect test to evaluate other components in the climate system, including vegetation, atmospheric circulation, and precipitation [58, 90].

Sea Salt

Here, we focus on the inorganic component of marine primary aerosols, i.e., sea salt from both open water [100] and sea ice [101–103]. Information about LGM sea salt deposition is derived from the interpretation of signals preserved in polar ice

cores (Table 1). This limits the spatial coverage of this kind of paleo-records compared to dust. Nonetheless, the location of polar ice cores in the proximity of high latitude oceans, where sea salt is the dominant aerosol species, offers an interesting chance for model validation.

Soluble Na^+ ions, stable in snow and ice, are considered the more robust indicators of sea salt depositional fluxes in Greenland and Antarctic ice core records, because crustal sodium contributions are considered negligible in relative terms, and be corrected for [54, 69]. Beside the dominant NaCl , other sea salts (Na_2SO_4 , CaSO_4 , CaCO_3) contribute to the ionic budgets in polar snow and ice; in particular, it has been noted that Na_2SO_4 is a species present in open water sea salts, but almost absent from sea ice salts originated from frost flowers, which can allow for disentangling the two sources based on considerations of ion balance and corrections based on standard sea water content [17, 104]. A few recent studies based on the sublimation of ice samples and the combination of microscopy and spectroscopic analyses helped the characterization in their solid state of soluble aerosols trapped in ice

cores, as well as variations in relative abundance in different climatic periods [105, 106].

The relative influence of sea salts in specific ice cores varies with the geographical location and proximity to the edges of the ice sheets, hence to the open water and sea ice sources [54]. In general terms, ice core records indicate that deposition rates of Na^+ in Antarctica increased by a factor of 3 to 5 during the LGM, compared to the Holocene [107–109]. Smaller increases, between 1.5 and 3 times the Holocene average if looking Na^+ accumulation rates or concentration in ice, were observed in Greenland [53, 110]. About whether concentrations in ice, rather than deposition fluxes, should be regarded as more representative of aerosol burden, some have argued that, because of changes in snow accumulation and aerosol deposition mechanisms, concentrations in ice are more informative than deposition fluxes in areas where wet deposition of aerosol dominates over dry deposition, and vice versa [57, 69].

Changes in sources (sea ice cover), emission strength and transport pathways (wind speed, cyclogenesis), and residence time (reduced wet scavenging) have all been invoked as possible candidates to explain glacial-interglacial variability in sea salt deposition to polar ice sheets [69, 111]. Because only ice cores are available to compare against for the LGM, open ocean sea salts emissions from the mid and low latitudes, which should be the most important for radiative forcing, cannot be validated in these comparisons.

Very few modeling studies attempted to simulate the LGM sea salt aerosols. Experiments considering only an open ocean source estimated LGM (current/pre-industrial control) emissions of ~ 5700 (~ 6000) Tg a^{-1} [112], ~ 3400 (~ 4000) Tg a^{-1} [93], and ~ 4100 (~ 4300) Tg a^{-1} [113], respectively. The general slight decrease observed in those studies, as well as in others focusing more on the comparison with specific ice core records [114, 115], was generally attributed to an increase sea ice cover, which reduces the source areas and increases the distance from source to sink. On the other hand, by also including a sea ice source, Yue and Liao found an increase of almost 200% in their LGM global emissions, reaching $\sim 11,900$ Tg a^{-1} [113]. In general, studies incorporating a sea ice source found a significant increase in emissions, improving the comparison with, but still underestimating, sea salt deposition fluxes from ice core records in terms of Na^+ deposition rates [93, 112, 113, 115] or modern atmospheric concentrations [116].

Sulfur and Nitrogen Species

The major sources of natural sulfur aerosol in polar ice are phytoplankton's emissions of dimethyl sulfide (DMS), which is oxidized into methane sulfonic acid (MSA) and H_2SO_4 in the atmospheric environment. These two species can be measured in ice core samples via ion chromatography [54]. SO_4^{2-}

anions are stable in snow and ice, and those produced from the DMS oxidation pathway are a major contributor to the total SO_4^{2-} budget, which also includes contributions from volcanic activity and sea salt. The fraction of non-sea salt sulfates can be artificially separated from the total SO_4^{2-} budget by subtracting the sea salt fraction, based on the standard $\text{Na}^+:\text{SO}_4^{2-}$ ratio in sea water; the presence of sea ice salts depleted in sulfates complicates this kind of exercise [54, 107], adding uncertainty to a quantitative separation of different sources of sulfate aerosol in polar ice. On the other hand, it was shown that MSA has preservation issues in present-day conditions [117], whereas the presence of larger amounts of dust in the LGM is thought to have stabilized its preservation by offering the potential for fixation of the MS^- anion onto particulate material [107].

Ice core data suggests no significant glacial-interglacial variations in non-sea salt sulfate deposition fluxes to Antarctica [107], in contrast to increase in MSA LGM fluxes [118, 119], which are nonetheless affected by preservation issues. In contrast, an increase of ~ 2 of non-sea salt sulfate deposition fluxes was observed in Greenland [53]. In general, substantial uncertainties still exist in reconstructing and understanding the LGM sulfur cycle. Modeling studies [120] and present day observations [121] may help shed some light on these issues.

A few records of NO_3^- and NH_4^+ from polar ice cores also exist [122], although some concern arises from potential contamination and/or proven preservation issues, especially for nitrates [123]. The few existing records indicate an increase in LGM nitrate deposition fluxes in both Greenland [53] and Antarctica [124]. Very little is known about nitrate deposition on polar snow, although the analysis of NO_3^- oxygen isotopes has emerged as a viable tool to make inferences about the potential sources [125]. On the other hand, NH_4^+ records indicate larger fluxes during interglacials [122, 126]. In general, there is so far too little information to make robust inferences about the magnitude and causes of variations in nitrogen aerosol species on these time scales.

Since most of the radiative forcing from these short-lived aerosols will come from the concentration in tropical and mid-latitude regions, which are not sampled by polar ice cores, there are limits to how much information we can obtain [1].

Carbonaceous Aerosols

Black carbon is also referred to as elemental carbon (char and soot). In terms of carbon mass budget, the BC concentration in present day continental atmosphere is about one order of magnitude lower than OC [127]. Natural OC aerosol is made up by primary organic aerosol (POA), and foremost by secondary organic aerosol (SOA) originated from volatile organic compounds (VOCs) [128]. BC is preferentially emitted during the flaming phase of a wildfire, whereas OC aerosols mostly

originate under smoldering conditions. The highly heterogeneous chemical composition of carbonaceous aerosols may give rise to confusion in the nomenclature used in the scientific literature, where different operational definitions appear [129].

There is limited direct information on BC and OC, from ice cores. Measurements of total/dissolved OC and speciation of carbonaceous aerosols have been carried out on surface snow or shallow firn cores and alpine ice cores [130, 131], but deeper paleoclimate records are very rare at best [132]. In particular, we are not aware of any record spanning the LGM that provides estimates of mass accumulation rates of carbonaceous aerosol species. A recent paper on variations in relative abundance of different types of fluorescent organic matter in West Antarctica indicates a stronger imprint of humic-like material during the Holocene compared to the LGM, interpreted as more expansive vegetation cover and increased production and degradation of complex organic matter in terrestrial environments [133].

On the other hand, chemical tracers of past fire activity have been sought and measured in snow and ice; while some of these species may not be direct measurements of the most important carbonaceous aerosol species, they have the potential to be related to the major aerosol emissions from paleofires, upon knowledge of a present day characterization of fire emissions and deposition of its proxies onto snow and ice [134]. In particular, ammonium, formate, and levoglucosan seem to be the most promising species so far applied to ice cores dating back several millennia, at least in Greenland [55]. Ammonia tends to be the dominant nitrogen species emitted during boreal forest fires that would leave a trace in Greenland ice, by deposition on the form of ammonium formate; levoglucosan is only produced by combustion of cellulose under smoldering conditions, although its chemical stability during transport is still uncertain [55].

Complementary to ice core tracers, charcoal sedimentary records from lakes, peat bogs, and soil profiles represent a proxy for paleofires. The global patterns of charcoal abundance and the relative variations at specific sites provide an indicator of the frequency, intensity, and extent of past fires [135]. The Global Charcoal Database collects and organizes such type of records [56, 135]. Synthesis from the Global Charcoal dataset indicates a consistent pattern of low fire activity in the LGM compared to the Holocene, with a few localized exceptions [56]. This is generally consistent with an overall reduction of land vegetation biomass acting as fuel for fires—at high latitudes, the reduced fire activity is also linked to the presence of the Laurentide and Fennoscandian ice sheets. Indication of reduced fire activity in Northern high latitudes in the LGM is also consistent with the only ice core proxy record from Greenland, i.e., the ammonium record from the North Greenland Ice core Project (NGRIP) [126].

In terms of other biogenic emissions from land vegetation, global simulations with an Earth system model estimated

isoprene emissions between ~ 250 and 850 TgC a^{-1} in the LGM depending on the assumptions on CO_2 sensitivity and temperature boundary conditions, and a decrease by 42–44% in total SOA burden compared to their pre-industrial control for their central case [136, 137].

In synthesis, the overall knowledge on LGM carbonaceous aerosol is very limited. By combining information from the source of fires with information of past vegetation [138] and linking with past and modern data from sinks like snow and ice [134], we might have the tools to evaluate and constrain models simulating past fire activity, hence potentially the emissions of carbonaceous aerosols from fire [136, 139, 140] and vegetation [137]. Additional information is therefore needed, to provide a constraint on the sinks of carbonaceous aerosols, from polar (and potentially some alpine) ice cores spanning the LGM.

Aerosol Feedbacks on Climate

We now review the available information concerning the evaluation and quantification of aerosol feedbacks on climate during the LGM, in terms of direct and indirect effects in the atmosphere [1], as well as in terms of indirect impacts on biogeochemical cycles [141].

Direct and Indirect Impacts on Atmospheric Radiation

Numerical models are the main tool for studying aerosol impacts on atmospheric radiation, because it is very difficult to establish a causal relation for co-variations of aerosols and other climate proxies from the paleoclimate records, perhaps with the exception of very large volcanic eruptions. This is not the case for dust impacts on ocean biogeochemistry, as discussed in the following section.

Because of the large variations and the evidence of the strong dust-climate coupling imprinted in the paleoclimate records, most of the modelers' attention was focused on dust. Still, of the few studies simulating the LGM dust cycle, only some also included climate feedbacks. The case of dust is very interesting and very challenging because of dust interaction with both solar short-wave (SW) and terrestrial long-wave (LW) radiation [86, 142], which, combined with particle size distributions and the underlying surface albedo [63, 143–145], can result in geographically distinct patterns characterized by either a positive or a negative direct radiative forcing (Fig. 2a). On top of that, the LGM is challenging in particular because of the uncertainties in constraining dust emissions and changes in surface albedo [77, 86, 95].

The existing model studies that include both SW and LW dust-radiation interactions estimate the net top of the atmosphere (TOA) direct radiative forcing (RF), either instantaneous or effective, in a range between -0.02 and $-$

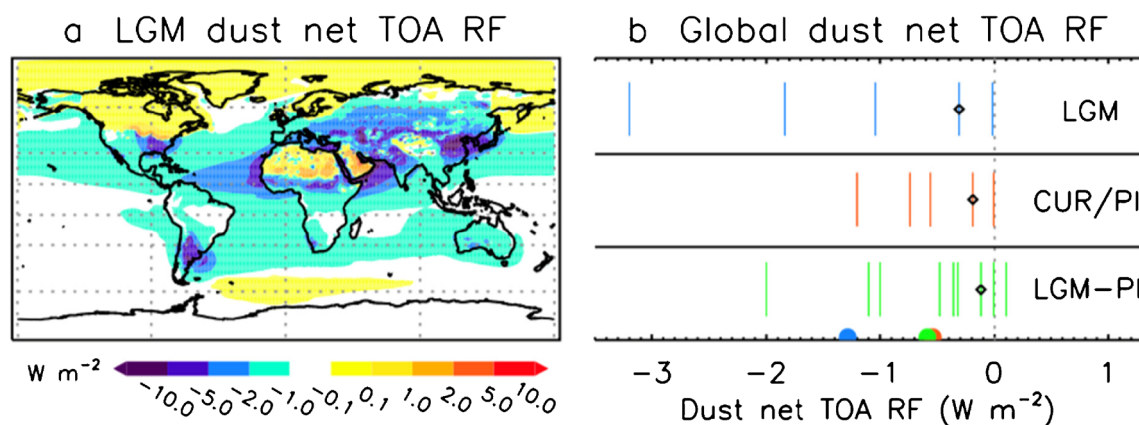


Fig. 2 **a** Example map of dust direct net (SW + LW) top of the atmosphere (TOA) instantaneous radiative forcing (RF) [86]. **b** Comparison of global estimates of dust direct net TOA RF from the literature, either in terms of instantaneous or effective RF, for LGM (upper panel, blue) and corresponding PI or CUR (PI/CUR) (middle panel, red) simulations [86, 90, 93, 95, 98, 146], and in terms of the LGM,

control climate anomaly (bottom panel, green) [86, 90, 93–95, 98, 146]. The black diamonds in **b** highlight the simulation displayed in **a** [86]. The semi-circles on the x-axis in **b** mark the average LGM (blue), control (red), and anomaly (green) of the respective model ensembles. The vertical gray dotted line mark the zero value on the x-axis

3.2 W m^{-2} for the LGM, and between -0.01 and -1.2 W m^{-2} for the corresponding pre-industrial/current climate control cases; LGM-control climate anomalies range from -2.0 to $+0.1 \text{ W m}^{-2}$ (Fig. 2b). Note that here we considered only the control cases for the LGM simulations, not other studies focusing on pre-industrial or present day climate. For reference, for present day climate, IPCC AR5 estimated net TOA direct RF from dust in the range -0.61 to $+0.10 \text{ W m}^{-2}$ [1], whereas a recent study re-evaluating some of the former estimates in light of new constraints indicates a range from -0.48 to $+0.20 \text{ W m}^{-2}$ [147]. The large spread in RF estimates is probably linked to the spread in estimating dust loads (Fig. 1), as well as to differences in assumptions on dust particle size and optical properties. Note that because of the spatial variability in RF, even models showing relatively low global averages of net TOA RF may predict strong regional forcing (Fig. 2a).

Far fewer studies exist looking at other types of climate impacts. Dust impacts on snow albedo could have been an important mechanism preventing the development of an ice sheet in Northern Asia [148], and accelerating the retreat of the ice sheets during glacial terminations [149]. A recent study estimated indirect effects of dust onto clouds through ice nucleation processes in an idealized LGM simulation, indicating a significant increase in cloud cover, and a corresponding net TOA RF of -1.1 W m^{-2} in the LGM, and -0.5 W m^{-2} for their control climate [97]. One study estimated that the net TOA direct RF of sea salt from open water sources is -0.92 W m^{-2} for their present day control simulation and -0.96 W m^{-2} for the LGM, associated to a surface temperature anomaly of -0.55 and -0.5K , respectively; if the sea ice source is also included, the LGM impacts become larger, i.e., -2.28 W m^{-2} and -1.27K [113]. The global impacts associated with aerosols changes from the interglacial

reference period to the LGM are non-trivial, if compared to the major climate forcings characterizing the LGM climate, i.e., greenhouse gases (-2.8 W m^{-2}) and ice sheets and sea level changes (-3.0 W m^{-2}) [150].

In synthesis, we have a few model studies targeting the dust cycle in the LGM and its direct climate impacts. While this ensemble provides a first-order constraint of LGM dust direct feedbacks, a coherent analysis of their differences was never carried out. It is expected that dust experiments in PMIP4 will provide an opportunity to make progress in this sense [99]. Notably, the consistent use of the same model configuration for Climate Model Intercomparison Project phase 6, i.e., the CMIP6-PIMP4 experiments under different climate scenarios, including a pre-industrial control case, will also provide a common reference scenario for the LGM—as well as for present and future climate [151]—that was not consistently available for models shown in Figs. 1 and 2. The inclusion of additional processes such as ice nucleation in a few models could be an important step forward in for understanding and testing past climate variability.

Indirect Impacts on Biogeochemical Cycles

Mineral dust is thought to be the main aerosol species of relevance for global biogeochemical cycles during the LGM. Due to its mineralogical composition, mineral dust effectively acts as a windblown carrier for chemical elements, which can be transported and eventually deposited far from the dust source areas, impacting land and aquatic ecosystems. On land, in particular, there is evidence that on long time scales, dust-borne phosphorus can compensate the basin scale losses in the Amazon, maintaining the balance of this major nutrient [152, 153]. On the other hand, ocean ecosystems are most notably

impacted by inputs of silica and especially iron. The latter in particular is a micronutrient that can limit primary production at the ecosystem level [154]. This can be of great importance for high nutrient–low chlorophyll (HNLC) regions, where the primary production is relatively low despite the excess in major nutrients available for phytoplankton growth. The “iron hypothesis” postulates that enhanced inputs of dust-borne iron during glacial climate stimulated an increase in productivity in HNLC areas which in turn lead to increased carbon sequestration in the deep ocean drawing down atmospheric CO₂ concentrations [155]. This mechanism has been proposed to be a potentially relevant contributor to the ~80–100 ppmv observed decrease in atmospheric CO₂ concentration during the LGM, compared to pre-industrial levels [107, 155, 156]. Artificial iron fertilization experiments have clearly shown an increase in productivity in HNLC zones in response to iron inputs, but a quantification of the consequent deep carbon sequestration remains unclear [157].

Marine sediment records have the potential to provide critical information on this subject, provided that it is possible to derive a record of export production, and ideally nutrient utilization, that this can be related to iron inputs, and that the source of iron can be identified. Export production represent the fraction of organic matter that “escapes” remineralization (recycling in the photic zone) and reaches the sea floor; it can be estimated directly through the sedimentation rates of organic matter, often challenging because of preservation issues, or more often by a proxy such as mass accumulation rates of opal [158] and biogenic barium [159], or through the Pa/Th ratio [158, 160, 161]. In general, the consistency of multi-proxy reconstructions of export production and iron fluxes can be interpreted as an indicator of iron fertilization [157]. An

additional constraint comes from the analysis of nitrogen isotopes in bulk sediment, foraminifera or diatoms, which is an indicator of nutrient utilization, and provides information on changes in the efficiency of major nutrient consumption [66, 162]. Finally, disentangling the role of different contributors to the iron budgets can clarify the actual mechanism of iron fertilization and allows for the quantification of the role of dust aerosol compared to other lithogenic inputs from the bottom, volcanic material, ice-rafted debris, or hydrothermal vents [163, 164].

The general view emerging from marine sediment cores [165], complemented by a number of recent studies (Fig. 3a), is that iron fertilization during glacial periods actually caused an enhancement of the efficiency of the ocean biological pump in specific regions [158, 183]. In particular, there is evidence that in the Southern Ocean, the most relevant HNLC area in terms of spatial scale and potential to influence the global carbon cycle, iron fertilization enhanced the efficiency of nutrient utilization [174]. In the subantarctic zone, this iron fertilization attributed to dust was also coupled to a net increase in export production, suggesting enhanced carbon sequestration in the deep ocean [167–169]. South of the polar front, a reduction of export production is observed, associated to diminished upwelling and increased stratification [165, 170, 183]. The other potentially relevant HNLC areas are the North and Equatorial Pacific (Fig. 3a). A study along the Line Islands in the central equatorial Pacific finds no evidence for iron fertilization by dust, nor increased export production or increased nutrient utilization, during the LGM [160]. A detailed investigation of three sediment cores, spanning the entire equatorial Pacific, showed that at each of the sites, biological productivity did not respond to increased dust

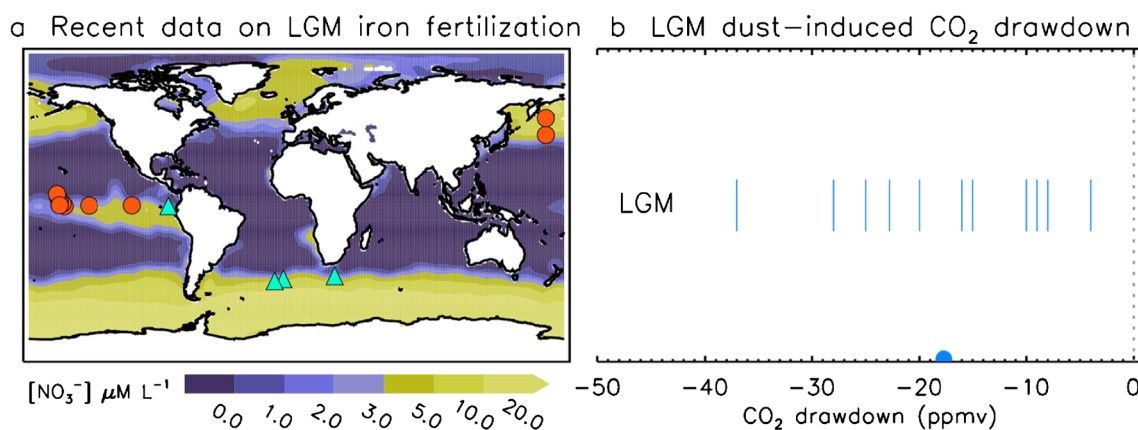


Fig. 3 **a** Map of present day high nutrient–low chlorophyll (HNLC) oceanic regions, indicated by patterns of nitrate concentrations in surface waters [166], along with the location of the most relevant recent core sites [159, 160, 167–173]. The shape of the symbols indicate whether the observations suggest an increase (triangle) or not (circle) in export production in the LGM. The color of the symbols indicate whether observations suggest dust-driven

iron fertilization in the LGM (green) or not (red). See also pre-existing global compilations of changes in ocean productivity in references [165, 174]. **b** Model-based estimates of CO₂ drawdown induced by increased LGM dust deposition [175–182]. The semi-circle on the x-axis in **b** marks the average of the model ensemble. The vertical gray dotted line mark the zero value on the x-axis

deposition during glacial conditions, thus arguing against dust fertilization [159]. Both of these studies are inconsistent with prior work in the region [184]. In the subarctic North Pacific, the very limited studies seem to indicate that increases in export production mark the beginning of the deglaciation, rather than the LGM, and seem unrelated to dust activity or in general to iron inputs [68, 171, 172, 185].

Ocean biogeochemistry models can help quantify potential impacts of iron fertilization, if the inputs of dust [86, 90, 175] and other sources of iron are reasonably prescribed [186, 187]. A few studies with three-dimensional ocean biogeochemistry models of different complexity targeted the LGM, applying a variety of assumptions/strategies in terms of dust inputs, iron solubility, presence of iron-binding ligands, carbonate compensation, and even climate boundary conditions for their LGM dust experiments. In general, the models indicate that LGM dust-induced iron fertilization could be responsible for increased export production and for the associated drawdown of almost 20 ppmv of atmospheric CO₂, despite the large spread in estimates (Fig. 3b) and the difficulty in establishing a coherent comparison given the diversity on experimental designs [175–182]. Box model studies (not included in Fig. 3b) tend to indicate larger estimates of CO₂ drawdown [156, 188].

In synthesis, marine sediment core data generally agree on the occurrence of dust-borne iron fertilization as a mechanism that enhanced the efficiency of the biological pump and resulted in increased export production, at least in the subantarctic Southern Ocean [167, 189]. Ocean biogeochemistry models indicate a reduction of ~20 ppmv CO₂ during the LGM in response to iron fertilization [182]. The quantification of this effect and its impact on the global carbon cycle remain however highly uncertain, most notably because of uncertainties in the representation of key process in ocean biogeochemistry models [157], in the quantification of dust [90] and other potential sources of iron to HNLC areas in particular [186], and because of uncertainty in quantifying the bioavailable fraction of iron [190]. By considering combined uncertainties in dust deposition and iron solubility alone, a recent study estimated differences of a factor ~5 in soluble iron inputs to the ocean globally, and up to two orders of magnitude in the glacial Southern Ocean [90].

Additional paleodata from marine sediment cores, as well as future improvements in ocean biogeochemistry models, also driven by data from initiatives such as GEOTRACES [191] or process-based field campaigns, are needed to better quantify iron solubility [192–194] and indirect effects of dust on the ocean carbon cycle. Recent developments in explicitly representing mineralogy and aging in dust models [195–197] will provide enhanced tools to derive dust-borne iron inputs to the ocean, especially if also used for paleoclimate experiments and notably for the LGM [99].

Conclusions

The preservation of certain aerosol species in natural archives, along with other paleoclimate records, offers the opportunity to study the response of these aerosols' life cycle to changing climate conditions, and in some cases, it provides clues about aerosol feedbacks onto climate. Past climate variability since the last interglacial can be seen as an envelope of the potential natural aerosol variability, in terms of changing emissions as well as feedbacks onto climate. In particular, paleoclimate records from the LGM seem to indicate a decrease in aerosol emission from land vegetation, whereas emissions of dust and sea salt at mid and high latitudes were significantly enhanced.

For the LGM climate, mineral dust has received most of the attention, because of its preservation in several natural archives worldwide, the amplitude of its temporal variability, and its potential to impact climate directly and indirectly. While significant uncertainties still persist, dust seems likely an important contributor to the LGM climate forcing; enough knowledge has emerged to combine several models and observations, offering a unique opportunity to improve our understanding of the mechanisms controlling the global dust cycle, as well as its feedbacks onto climate. The LGM climate is also an ideal target to test the inclusion of new processes in models, such as iron fertilization of the oceans and ice nucleation in clouds.

The global budgets and causes of variability of other aerosol species are far less well constrained, although at least sea salt from open water and sea ice sources seems to be a near-future potential target, because of its potential direct and indirect effects on the atmospheric radiation balance, and the availability of some quantitative constraint on its variability from polar ice cores.

Ongoing and future work on constraining and modeling vegetation cover is a key aspect for the representation of past and future natural aerosol emissions, because of its tight link with dust emissions as well as for direct and post-fire aerosol and aerosol precursors' emissions.

In general, more observations are needed, in order to enhance the geographically resolved, quantitative constraints of aerosol mass budgets, and the understanding of specific processes. The coherent organization of such data into global databases is a key tool, allowing a holistic view of biogeochemical cycles, and providing a benchmark for global Earth system models aiming at predicting climate change.

In addition to LGM equilibrium climate conditions discussed in this manuscript, abrupt changes related to the deglaciation as well as glacial variability imprinted in natural archives [31, 198] offer a unique opportunity to improve our understanding of aerosols-climate interactions, provided that adequate observational databases can support modeling experiments [59].

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Compliance with ethical standards

Conflict of Interest The authors declare no conflict of interest.

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