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Article

Keywords:

Posted Date: September 10th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10886013/v1>

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Additional Declarations: There is **NO** Competing Interest.

Multi-proxy records reveal rapid atmospheric reorganization during Dansgaard–Oeschger events

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September 2026

To be submitted to Nature Geoscience

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Abstract

Dansgaard-Oeschger (D-O) events refer to a series of rapid climate fluctuations in the Northern Hemisphere during the last glacial period. These fluctuations are recorded in Greenland ice cores by large, abrupt changes in both water isotopes and aerosol deposition, but the processes driving these changes remain unclear. Here we present new high-resolution, multi-proxy measurements from the Greenland GISP2 ice core spanning eight D-O events. In addition to multiple dust and sea-salt tracers, we provide the first high-resolution biomass burning proxy records from refractory black carbon (rBC) concentration and particle size during these transitions. Bayesian ramp fitting analysis shows that water-isotope and aerosol changes generally occurred within a narrow temporal window with no robust and reproducible sequence. Using a simplified aerosol transport and deposition framework, we find that changes in atmospheric transport and removal processes can explain much of the variability shared among aerosols from distinct source regions. Residual variability shows stronger relationships within than across source classes and, particularly for dust, differs between cold and warm climate states. These results suggest that rapid large-scale atmospheric and hydrological reorganization drove the common aerosol response, while changes in individual source systems contributed to the remaining variability. These new multi-proxy records provide valuable constraints for evaluating climate-model representations of abrupt climate transitions.

Introduction

Greenland ice-core records reveal pronounced rapid climate fluctuations during the last glacial period, known as Dansgaard-Oeschger (D-O) events^{1,2}. Palaeothermometry based on $\delta^{18}\text{O}$ and $\delta^{15}\text{N}$ isotopes suggests that Greenland warmed abruptly by 5 to 17 °C within a few decades during D-O event onsets³. Paleoproxy records and numerical simulations strongly suggest that these abrupt climate events were associated with changes in the strength of Atlantic Meridional Overturning Circulation (AMOC), with sea-ice retreat likely playing an important role in affecting the rapidity and amplitude of the transitions⁴⁻⁷. However, the full physical mechanisms underlying D-O events remain debated^{8,9}, and current climate models still struggle to reproduce such abrupt transitions while simultaneously capturing the full range of observed variability. This has raised concerns that current models may be overly stable, reducing confidence in projections of future climate behavior in the context of tipping points and abrupt climate change^{10,11}.

During D-O cycles, aerosol concentrations from multiple sources recorded in the Greenland ice core, such as mineral dust and sea salt, decreased dramatically during transitions from cold Greenland stadials (GS, with more negative $\delta^{18}\text{O}$ values) to warm Greenland interstadials (GI)¹²⁻¹⁴. Similarly anti-correlated relationships between aerosol proxies and water isotopes in ice have been observed over the last deglaciation in both Greenland and Antarctic ice cores¹⁵. These may reflect reduced transport efficiency from source regions to polar ice-core sites during cold-to-warm transitions and therefore provide valuable information on changes in large-scale atmospheric circulation and precipitation patterns. However, aerosol emissions themselves are also influenced by climate conditions. As a result, ice-core aerosol signals likely reflect a combination of transport, removal, and source changes, making their interpretation and mechanistic attribution challenging^{13,15-17}.

The relative timing of water-isotope and aerosol changes provides additional information on the atmospheric evolution of D-O transitions. Previous studies have used the duration and phasing of D-O transitions in different proxies to infer the sequence of changes in atmospheric circulation, sea ice, and Greenland temperature¹⁸⁻²¹. For example, one analysis suggested that dust and local precipitation changes preceded sea-salt and Greenland temperature changes by approximately one decade on average¹⁴, a result interpreted as atmospheric circulation changing before sea-ice retreat and Greenland warming. However, timing estimates are sensitive to fitting

uncertainty²², and subsequent studies have not identified statistically robust or reproducible leads or lags^{21,23}.

More broadly, observational constraints remain limited by the number of available high-temporal-resolution aerosol records and the restricted range of proxy types. Here we investigate water isotope and aerosol proxy variability during eight D-O events using new records developed from continuous measurements of ~260 m of the archived GISP2 ice core from Greenland. These measurements provide annual to multi-annual resolution records of a broad suite of proxies related to temperature and precipitation (i.e., $\delta^{18}\text{O}$, δD , and deuterium excess, hereafter *d*-excess), as well as aerosols from both terrestrial (dust and biomass burning, hereafter BB) and marine sources (sea salt) that greatly expand the breadth and resolution of data compared to the original ~20-year resolution GISP2 geochemical measurements²⁴. We also report aerosol size of refractory black carbon (rBC) in the form of geometric mean diameter (rBC_{diam}), providing an additional microphysical constraint complementary to concentration-based records for evaluating size-dependent aerosol processes during long-range transport. We use these records to examine the coherence and relative timing of water-isotope and aerosol transitions and to assess the roles of atmospheric transport, removal, and source changes in the observed variability. The records are interpreted using a physically based one-dimensional aerosol transport framework that includes size-resolved wet and dry deposition. Together, these results help distinguish a common transport-and-removal response from source-specific and climate-state-dependent variability, providing new insights into the atmospheric and hydrological reorganization during abrupt D-O transitions and new observations for their representation in climate models.

Results

Coherent aerosol and water-isotope variability in a high-resolution Greenland ice-core record

Here we present high-resolution stable water isotope and aerosol concentrations from the GISP2 ice core for the 30.7 to 47.4 ka BP interval (Fig. 1), measured using the continuous flow analysis system at the Desert Research Institute (Methods). Across the investigated D-O transitions, $\delta^{18}\text{O}$ increased 3.3 to 4.5‰ from GS to GI conditions, corresponding to approximately 10-14 °C of rapid warming at the Greenland ice-core site (T_{site}) over several decades^{25,26}. At the same time, d -excess decreased by approximately 3‰, indicating cooling of ~3-4 °C at the moisture source (T_{source})²⁵. The opposing yet closely aligned changes between T_{site} and T_{source} have been interpreted as evidence for a major reorganization of atmospheric circulation and moisture transport, with GS moisture weighted toward warmer, more southerly open-ocean regions and GI moisture toward cooler, more northerly source²⁵.

Our ice-core aerosol records reveal similarly pronounced and coherent variability across the D-O cycles (Fig. 1). The aerosol species reported here include BB tracers (rBC); dust tracers including non-sea-salt calcium (nssCa), cerium (Ce) and insoluble particles; and sea-salt tracers including sea-salt sodium (ssNa) and chloride (Cl). These aerosols originate from distinct source regions and are governed by different emission processes and potentially different transport pathways. Nevertheless, their concentrations consistently covary with one another and are strongly anti-correlated with $\delta^{18}\text{O}$ during the D-O events (Figs. 1 and S1a), with substantially higher concentrations during GS conditions. Although for some natural processes, such as BB, emissions might be expected to be higher under warmer conditions, higher aerosol concentrations in the ice core during GS conditions suggests that source changes alone cannot explain the record.

The responses of different aerosol classes show coherent timing but are not uniform in magnitude. For example, rBC concentrations are typically 3-5 times higher during GS than during GI conditions (Fig. 1d), dust tracer concentrations vary by a factor of 7-10 (Fig. 1h-j), and sea-salt tracers vary by a factor of 2-4 (Fig. 1f-g). These different magnitudes indicate source-specific variability superimposed on the shared multi-aerosol response.

In addition to aerosol concentrations, this study presents the first rBC particle size records across the abrupt D-O transitions. The observed rBC_{diam} is consistently approximately 10-15 nm larger during GS conditions (Fig. 1e). Modern observations show that rBC core-size distributions

vary substantially near source regions but become more uniform in regions remote from fire activity. This tendency arises because removal processes during long-range transport act as a size-selective filter, preferentially removing particles with less efficiently transported sizes^{27,28}. The recurrent 10-15nm shift of rBC_{diam} in Greenland is therefore difficult to explain by changes in source emissions alone and is consistent with a substantial contribution from altered size-dependent transport and removal during D-O transitions.

Together, the closely aligned temporal evolution of isotope and aerosol records, their distinct response in magnitudes, and recurrent rBC particle-size shift define a multi-proxy fingerprint of D–O transitions.

Rapid coupling of water-isotope and aerosol transitions

To examine whether this coherent multi-proxy response emerged within a narrow temporal window and whether any consistent lead–lag sequence occurred among proxy classes, we use a ramp-fitting framework following Slattery et al²² to fit data near abrupt transition periods (Fig. S2, Table S1) and determine transition timing characteristics, including onset time (Fig. S3, Table S2) and duration (Fig. S4, Table S3). Bayesian inference with prescribed prior uncertainty assumptions combined with Markov chain Monte Carlo (MCMC) sampling is used to estimate posterior parameter distributions and associated uncertainties (Tables S2-S7, Methods). We performed additional synthetic tests for each proxy record across the relevant parameter ranges to quantify potential fitting biases, which arise primarily at high noise-to-signal ratios when stochastic variability is difficult to distinguish from the true transition signal²².

For the water-isotope proxies (i.e., $\delta^{18}O$ and d -excess), the medians of the estimated transition durations are generally within 100 years (Fig. S4, Table S3), with d -excess records show a slightly shorter duration and narrower posterior uncertainty range. The median onset estimates generally differ by less than 10 years (Fig. 2, Tables S8-S15). The 5th to 95th percentile uncertainty ranges are broad and encompass both substantial $\delta^{18}O$ leads and lags relative to d -excess, suggesting no statistically robust lead-lag relationship between the two proxies. This conclusion holds even after accounting for the non-negligible ~10-year bias in d -excess onset estimates identified in the synthetic calibration tests (Table S16). Because the nominal posterior uncertainty does not fully capture this timing bias (Table S17; Methods), we explicitly account for the bias in the lead-lag interpretation in the following discussion. These results together suggest that changes in Greenland temperature and moisture sources occur within a relatively short interval, indicating

a rapid reorganization of the broader atmospheric and hydrological system on decadal-to-centennial timescales.

Aerosol transitions occurred within similarly narrow temporal windows. Within the same source classes, tracers show broad though nonuniform agreement. For dust, at least two of the three tracers show largely overlapping median timing estimates and uncertainty ranges across all the events. The two sea-salt tracers exhibit a clearer difference in the median timing estimates in some events (i.e., D-O #6 and D-O #10, Fig. 2b, e), but the direction and magnitude vary. Across events, the median timing difference remains small after accounting for the tendency of the fitting framework to estimate ssNa onset time slightly late (Table S16, Fig. 2h).

Across sources, dust, sea-salt, and BB generally transition close to the water isotope changes (Fig. 2, Tables S8-S15). Aerosol median onset estimates occur slightly after the isotope transitions in many events, but most of these offsets are less than ~ 10 years and comparable to the estimated bias of the ramp-fitting framework (Table S16). Therefore these should not be interpreted as robust evidence that isotope changes preceded the aerosol response. In addition, the relative timing among aerosol proxies is complex and varies across events, with no reproducible ordering of dust, sea-salt, and BB transitions (Fig. 2, Tables S8-S15).

The differences in timing distributions between ssNa and dust tracers tend slightly towards positive values – i.e., the ssNa changes appear to occur before those of the dust tracers (Fig. 2, Tables S8-S15), in contrast to a previous study suggesting that dust changes preceded sea salt changes²¹. However, the differences between the ssNa and dust timing are generally less than one decade considering fitting bias and are therefore highly sensitive to the fitting uncertainty. The results therefore do not support a systematic order between dust and sea salt, or the water isotope proxies, consistent with later studies^{22,23}. The addition of rBC records extends this comparison to a third aerosol class with distinct source regions and emission controls. However, neither rBC concentration nor particle size shows a consistent lead-lag relationship with dust, sea salt or the water isotopes. The timing of rBC concentration and size transitions is also variable among events, with concentration leading size in some events and size leading concentration in others.

We explore whether event specific variations might be larger than fitting uncertainty and therefore could reflect real timing differences. For example, Event #5.2 shows the median estimates of aerosol-isotope timing differences could be as large as 30 years (Fig. 2a, Table S8), while event #6 shows aerosol proxies appearing to transition earlier than the water isotopes (Fig.

2b, Table S9), even considering the possibility that the isotope onset time itself may be biased early (Table S16). Even though some individual offsets seem to be statistically meaningful compared to fitting bias, they do not combine into a reproducible sequence across D-O events, and the cross-event average timing differences remain small (Fig. 2h, Table S15).

Taken together, the absence of a reproducible sequence among aerosols with distinct emission, source, and transport characteristics argues against a progressive response of independent source systems and instead supports a rapid, tightly coupled adjustment of the large-scale atmospheric circulation and hydrological processes.

A common transport-and-removal response explains the shared variability

To examine whether a common transport-and-removal mechanism can account for this broadly coordinated response, we interpret these Greenland ice core records using a simplified aerosol transport and deposition model framework adapted from Markle et al.¹⁵. Details of the modeling framework and its validation are provided in Methods and Supplementary Information (Text S3). In brief, this model framework proposes that both aerosol and water isotope signals on climatological timescales are influenced by the cumulative cooling and condensation experienced by air masses during transport from source regions to polar ice-core sites. Progressive condensation removes water through precipitation, altering isotopic composition and removing aerosols through wet scavenging. Colder conditions produce lower $\delta^{18}\text{O}$ but less absolute moisture condensation along the pathway, reducing wet scavenging and allowing more aerosols to reach the ice-core site. This framework therefore provides a physical basis for relating Greenland temperature (reconstructed from $\delta^{18}\text{O}$) to ice core aerosol variability.

Figure 3 compares the observed aerosol records with relationships estimated by this framework as a function of Greenland temperature anomaly relative to the present day (ΔT_{site}). Note that two prescribed parameters, wet-scavenging efficiency, which controls the strength of aerosol removal, and source latitude, which affects the transport pathway and accumulated removal, could influence the modeled aerosol concentrations and their relationships with water-isotope and temperature changes (see Methods). Because observational constraints on removal efficiency remain limited, and the emission, transport, and removal processes are highly simplified, the modeled relationships should be interpreted as qualitative diagnostics of transport-and-removal effects rather than quantitative estimates of their exact contribution. We also perform sensitivity

tests on these parameters to evaluate whether these uncertainties affect the qualitative interpretation (Figs. S5-S6).

As shown in Fig. 3a-c, all three dust records show the clearest separation into two distinct groups in their relationships with Greenland temperature. We thus use k-means clustering to identify these groups objectively and apply the dust-based classification to the rBC and sea-salt records. The resulting groups broadly correspond to colder and warmer Greenland conditions (Fig. 3d-g).

While Markle et al focused on broader time spans encompassing millennial and glacial–interglacial variability ¹⁵, we show that the modeled relationships also capture a large portion of aerosol variability across the abrupt D-O transition at much finer event-scale resolution (Fig. 3). For the dust tracers, the model represents much of the temperature-dependent variability within each climate state but does not capture the separation between the groups. The modeled uncertainty range spans much of the variability within individual clusters but is unlikely to explain the full separation between them (Figs S5a, S6a). For rBC concentration, wet scavenging is simulated using a size-resolved removal scheme constrained by the measured particle-size distributions (see Methods), which reproduces the broad observed relationship under the assumption of constant emissions (Figure 3f). Variations in the model parameters have only a limited effect on the modeled relationship (Figs S5c, S6c). The rBC observations may also show a weak offset between the cold and warm states, but this separation is difficult to distinguish robustly based on modeled relation. For the sea-salt proxies, the modeled relationships agree reasonably well with observations, with relatively small variability around the central modeled relationship (Fig. 3d-e, Figs. S4b, S5b). When dust-based clustering is applied, the cold and warm groups follow broadly similar relationships with ΔT_{site} , providing little evidence on an additional state-dependent offset beyond the common transport and removal responses.

The rBC particle size record provides a complementary test because particle size is directly modified by size-dependent removal during the long-range transport. When assuming source-size distribution does not change, the model produces shifts in the deposited rBC core size of approximately 15nm solely through changes in cumulative precipitation and the resulting differential removal of particles of different sizes. This magnitude is broadly consistent with the observed difference between the median rBC sizes of the two climate states, despite substantial scatter among individual observations (Fig. 3g). The agreement in particle size therefore provides

additional evidence that changes in transport and removal contributed importantly to the rBC response.

After removing the modeled transport-and-removal component, the covariance between $\delta^{18}\text{O}$ and the aerosol residuals is reduced by 30%-60% relative to that for original aerosol concentrations (Fig. S1b), and the magnitude of this reduction remains similar across the tested parameters (Fig. S7). Covariance among tracers from different sources is also reduced. Together, these results indicate that transport and removal explain much of the variability shared across aerosol classes during D-O events, but do not account for all observed variability. We therefore examine the structure of the remaining variability and the extent to which it differs among aerosol source classes.

Residuals reveal source-related and state-dependent variability

Although covariance among tracers from different sources is reduced after removing the modeled transport-and-removal component, covariance among tracers within the same sources remains moderate to high (i.e., Kendall's $\tau > 0.7$ among three dust tracers and $\tau \sim 0.54$ between two sea-salt tracers, Fig. S1b, Fig. S7). This persistent within-class covariance is therefore less likely to arise solely from independent random model error and indicates that the residuals retain variability associated with common source behavior, source-specific transport pathways, or other processes not represented by the simplified framework.

For sea salt, the residual variability is smallest. If attributed entirely to changes in source strength (i.e., $C_{air}(\phi_0)$ in Eq. S15), the inferred changes provide an upper-bound estimate of GS-to-GI source variability, suggesting scaling factors of approximately 0.75–1.4 can reproduce much of the residual range (Fig. 4, Figs. S8-S11). For rBC, sensitivity to the assumed model parameters makes it difficult to determine whether a comparable state-dependent signal is present; under the same assumption, source-strength factors of approximately 0.6–1.8 can reproduce the broader residual range (Fig. 4, Figs. S8-S11). These ranges are intended as qualitative measures of residual magnitude rather than quantitative source reconstructions. Dust shows the clearest and most structured departure from the common transport-and-removal relationship, which persists across the tested model parameter sets (Fig. 4, Figs. S8-S11). We thus discuss in detail below.

All three dust tracers exhibit two distinct clusters that broadly correspond to GS and GI conditions. Although the clusters largely overlap in reconstructed Greenland temperature, their

concentration differs by up to an order of magnitude, indicating that a single continuous relationship represented by the simplified framework alone cannot explain the full dust variability.

With an unchanged effective source latitude, an approximately threefold increase in source strength shifts the modeled relationship from the warm-state cluster toward the cold-state cluster (Fig. 3a-c). Such a change is physically plausible, as studies focusing on glacial-interglacial differences suggest higher dust loadings under colder and more arid conditions²⁹, owing to factors such as stronger winds, reduced vegetation cover, expansion of source regions^{30,31}. However, these studies do not directly constrain the magnitude of source changes across abrupt D-O transitions.

Source-strength changes alone also cannot explain the weaker dependence on ΔT_{site} observed during the cold states (Fig. 3a-c). In the simplified model, shifting the effective source latitude northward by 5° produces a lower slope and closer agreement with the cold-phase cluster without requiring source strength change (Fig. 3a-c). This sensitivity experiment is not intended to reconstruct an actual shift in source location, but illustrates how changes in source configuration or effective transport distance can alter the dust–temperature relationship. Recent climate-model simulations raise the possibility of a related mechanism at the D-O scale, suggesting that millennial Greenland dust variability may involve circulation-driven changes in the contribution and poleward transport of North African dust associated with shifts of the tropical rainbelt³¹. Although those simulations did not evaluate the two-cluster structure identified here, similar changes in source mixture and transport pathways could plausibly contribute to the state dependent dust signal discussed here, though this hypothesis is yet untested. The observed cluster separation may therefore reflect a combination of enhanced emissions, expanded or redistributed source regions, and altered transport pathways. These mechanisms cannot be distinguished with the present simplified framework, but they represent testable hypotheses for future provenance analyses and coupled dust–climate simulations.

Implications for modeling abrupt climate change

The high resolution, multi-proxy ice core records across eight D-O events provide new constraints on aerosol responses to abrupt climate transitions in the Northern Hemisphere during the last glacial period. Transport and residual analysis reveal a two-layer structure in the D–O aerosol response. A rapid, common transport-and-removal adjustment affects aerosols transported from multiple remote source regions to Greenland, while source- and pathway-specific changes are superimposed on this common adjustment, with their relative importance differing among aerosol classes.

As a caveat, both the transport modeling and timing analyses rely on simplifying assumptions and are subject to substantial uncertainty. Further simulations using climate models that explicitly represent more complex atmospheric circulation, coupled atmosphere-ocean interactions, and aerosol-climate feedback would help interpret these results and improve understanding of the underlying processes. The multi-proxy GISP2 records provide process-level constraints for such evaluations. Models should reproduce not only the magnitude of Greenland temperature change, but also the rapid and coordinated atmospheric and hydrological circulation change that shift isotope and aerosol records, the common transport-and-removal response across aerosol classes, the state-dependent behavior of dust, and the observed changes in rBC particle size. A model that reproduces abrupt Greenland warming but not these accompanying atmospheric signatures may capture the climatic endpoint without representing the correct circulation and hydrological adjustment.

D-O events are not directly analogues for future climate change because they occur under different boundary conditions, but they provide valuable observational records of abrupt transition between distinct North Atlantic climate states. Evaluating whether models can reproduce the rapidity, coherence, and state dependence of these transitions can help constrain proposed interactions among AMOC, sea ice, atmospheric circulation, and the hydrological cycle. More broadly, these records provide benchmarks for assessing model stability and improving confidence in simulations of low-probability, high-impact abrupt climate changes under future forcing. Given the possibility that continued warming may increase the risk of crossing climate tipping point thresholds^{32,33}, increased understanding of the past abrupt transitions could help identify model limitations and strengthen assessments of future abrupt-change risks.

Materials and Methods

Ice core measurements

Contiguous, longitudinal samples from the archived Greenland Ice Sheet Project Two (GISP2) ice core (72.583 N, 38.467 W, 3203 m a.s.l.) taken from 2100 to 2365.0 m—corresponding to 30.7 to 47.4 ka before present (ka BP)—were analyzed at the Desert Research Institute (DRI; Reno, NV) using a unique, well-established continuous flow ice core analytical system^{34,35}. This system includes two Element2 High Resolution Inductively Coupled Plasma Mass Spectrometers (HR-ICP-MS) operating in parallel for elemental and chemical measurements including sodium (Na), sulfur (S), calcium (Ca), and rare-earth-elements (REE); an Abakus laser-based particle size analyzer for semi-quantitative size-resolved insoluble dust concentrations; a desolvating nebulizer system coupled to a Single Particle Soot Photometer (SP2, Droplet Measurement Technologies) for measuring rBC concentrations and particle size distributions; and a Picarro L2130 isotopic water analyzer for oxygen and hydrogen isotopes ($\delta^{18}\text{O}$ and δD). Deuterium excess was calculated from the measured oxygen and hydrogen isotope ratios as $d\text{-excess} = \delta\text{D} - 8 \times \delta^{18}\text{O}$. Details of the ice core measurements methods for rBC and other species made using this system are described elsewhere^{34,36,37}.

The longitudinal GISP2 ice samples were melted continuously and the meltwater split by ridges engraved in the ceramic melter head into three streams, corresponding to three ice core regions. The innermost 1 cm² of the 10 cm² cross-section (10%) was used for the elemental HR-ICP-MS measurements, the outer next 20% used for all other measurements reported here, and the outermost 70% discarded to prevent contamination^{34,36,37}. The ice measurements were dated using the Greenland Ice Core Chronology 2005 (GICC05) time scale³⁸.

Determining the timing of D-O transitions using ramp fitting

To quantify the timing of the rapid transitions during D-O warming events, we implemented a Bayesian ramp-fitting framework following Slattery et al²², which extends the original method of Erhardt et al.¹⁴ by considering possible gradual trends before or after the fast, abrupt transition itself. Our implementation is adapted for single-event analysis and designed to assess whether consistent patterns exist across events and proxies. We summarize the main steps here, with full technical details provided in Text S1.

We extracted seven 800-year time slices from the ice core records, corresponding to all covered D-O events except D-O #9, with each transition positioned near the center of its respective time

window (Table S1). D-O #9 was excluded from the ramp fitting analysis because of its relatively small warming magnitude and the subsequent cooling associated with Heinrich Stadial 4³⁹, which made the identification of the transition highly uncertain²¹. Prior to ramp fitting, all proxy records (except for $\delta^{18}\text{O}$, d -excess, and rBC_{diam}) were log-transformed and normalized using the 3rd and 97th percentiles (i.e., $x_{\text{norm}} = (x - P_3)/(P_{97} - P_3)$).

Each transition is modeled as the sum of a deterministic ramp ($\hat{x}$) and a stochastic noise term (ϵ) derived from an autoregressive model of order one (AR(1)) (Eqs. 1-3). The ramp is parameterized by four primary variables: the initial value (x_0), transition magnitude (Δx), onset time (t_0), and duration (Δt). Following Slattery et al.²², we introduce two additional parameters, s_{GS} and s_{GI} , to represent linear trends before and after the transition (Eq. 2). This relaxes the assumption of equilibrium states outside the transition period and allows for gradual but statistically significant background trends. The stochastic noise term (ϵ , Eq. 3) is characterized by the noise amplitude (σ) and an autoregressive coefficient (α) as a function of the decorrelation timescale (τ) (Eq. 4).

$$x_i = \hat{x}_i(t_0, x_0, \Delta t, \Delta x, s_{GS}, s_{GI}) + \epsilon_i(\alpha, \sigma) \quad (\text{Eq. 1})$$

$$\hat{x}_i = \begin{cases} x_0 - s_{GS}(t_0 - t_i), & \text{if } t_i \leq t_0 \\ x_0 + \Delta x \frac{t_i - t_0}{\Delta t}, & \text{if } t_0 \leq t_i \leq t_0 + \Delta t \\ x_0 + \Delta x + s_{GI}(t_i - t_0 - \Delta t) & \text{if } t_i \geq t_0 + \Delta t \end{cases} \quad (\text{Eq. 2})$$

$$\epsilon_{i+1} = \alpha_i \epsilon_i + \sigma \sqrt{1 - \alpha_i^2} \zeta_{i+1} \quad \zeta \sim \mathcal{N}(0,1) \quad (\text{Eq. 3})$$

$$\alpha_i = \exp\left(-\frac{t_{i+1} - t_i}{\tau}\right) \quad (\text{Eq. 4})$$

Where ζ_i represents a normally or lognormally distributed random variable with unit variance.

We use Bayes' theorem to infer the posterior distributions of the model parameters ($\theta = [t_0, x_0, \Delta t, \Delta x, s_{GS}, s_{GI}, \sigma, \tau]$) under prescribed prior uncertainty assumptions. An ensemble Markov Chain Monte Carlo (MCMC) sampler is used to generate representative samples from the posterior distribution (Text S2). For each fit, the sampler generates 4,000 posterior samples, each representing one plausible parameter set. We report the 5th-95th percentile range of these posterior distributions as the 90% credible interval.

From the fitting results, the residual autoregressive coefficient (α) is generally higher than 0.7 and relatively consistent across proxies and events (Table S4), suggesting substantial autocorrelation in the residual noise that should be considered in the ramp-fitting framework. The noise-to-signal ratio varies with species, with smaller values for dust tracers and larger values for ssNa and BC_{diam}, while remaining relatively consistent across different D-O events (Table S5). In some cases, the median fitted slopes in GS or GI periods adjacent to the abrupt transitions deviate from zero, suggesting gradual changes before or after the rapid transitions (Tables S6-S7). However, these slopes do not appear to show systematic dependence on either event or proxy type. The extended ramp model that includes pre- and post-transition slopes may therefore provide a more comprehensive representation of the transition structure and associated parameter uncertainties.

Tests with synthetic data suggest that, for most proxies, the true onset time lies within the 5th-95th percentile range of the fitted posterior distributions, indicating that the fitting framework is generally well calibrated and that the estimated uncertainty ranges are reasonable (Text S2, Table S17). An exception is *d*-excess, for which the fitted onset time distributions frequently do not encompass the true onset time and tend to occur earlier, indicating that the uncertainty estimates are less well calibrated for this proxy. Thus, in the lead-lag discussion, we consider both the Δt distributions calculated from the fitted onset-time ensembles of each proxy pair (i.e., Tables S8-S15) and the potential fitting biases identified by these synthetic data tests (Table S16). The median fitted onset times from the synthetic-data experiments show varying degrees of bias relative to the true values (Table S16). For *d*-excess, the mean bias is less than 10 years, despite the poorer coverage of its posterior uncertainty ranges. Biases are generally within several years for most variables including $\delta^{18}\text{O}$, rBC, Cl, insoluble mass, nssCa, and Ce, while larger biases, in some cases exceeding 10 years, are detected for ssNa and rBC_{diam}. These results suggest that inferred lead-lag relationships smaller than approximately 20 years may be sensitive to fitting bias and should therefore be interpreted with caution, whereas larger time differences are more likely to reflect robust lead-lag relationships.

Simplified aerosol transport and deposition model

We deployed a 1-D aerosol transport and deposition model modified from the approach in Markle et al. ¹⁵; a detailed description of the framework is provided therein as well as in the

Supplementary Materials (Text S3). In brief, the model framework considers 1-D transport of an air parcel from mid-latitude source regions toward the high-latitude ice core site. As the air parcel undergoes pseudo-adiabatic cooling during the meridional transport, the saturated vapor pressure decreases following the Clausius-Clapeyron relationship, causing excess moisture above saturation to condense and be removed as precipitation. The precipitation drives both water isotope distillation and aerosol wet deposition along the transport pathway. Comparisons with ice-core based reconstructions as well as climate model simulations suggest that the simplified framework captures the first-order relationship between temperature changes, cumulative water condensation, and precipitation along the transport pathway (Fig. S12).

The original framework for the 1-D aerosol transport and deposition model employed a bulk aerosol removal scheme. The relationship between the concentration of an aerosol in the atmosphere (C_{air}) and that in precipitation (C_{precip}) resulting from scavenging and rainout is quantified by the wet scavenging efficiency (Text S3). For dust and sea-salt aerosols, we used updated wet scavenging efficiency values referring to published values (Table S18) but also performed sensitivity tests by varying these values by ± 200 to assess the influence.

For rBC-containing particles from biomass burning, we extended this scheme by developing a size-resolved particle removal scheme considering both wet and dry depositions (Figs. S13-S14). The relatively small particle size of rBC makes dry deposition more important than for dust and sea-salt aerosols, and the availability of rBC size measurements means that the wet deposition scheme can consider size-resolved particle removal by in-cloud scavenging⁴⁰, below-cloud scavenging⁴¹, and cloud activation. The rBC particles emitted from BB typically are coated with organic matter (OM). Here we assumed a coating-to-rBC mass ratio of 10, consistent with recent field observations suggesting thick OM coating of aged BB rBC particles⁴². To calculate the original particle size in air (including coating), we assumed material densities of 1.8 g/cm³ and 1.5 g/cm³ for rBC and OM, respectively. We assumed that the initial rBC mass-diameter distribution follows a lognormal distribution, with a geometric standard deviation of 2 and a geometric mean diameter of 180 nm (and performed sensitivity tests with 160nm and 200nm), according to aircraft observations for BB emissions⁴³. The size-resolved wet deposition scheme was validated against rBC removal efficiencies derived from simultaneous measurements for rainwater and atmosphere (Fig. S14)⁴⁴.

The dry deposition scheme for rBC was based on the size-resolved dry deposition velocity implemented in GEOS-Chem with the TOMAS microphysics module⁴⁵. This scheme predicts removal efficiency decreasing with increasing particle size for submicron particles, with a lowest removal efficiency at $\sim 1 \mu\text{m}$ (Figs. S13-S14).

The transport pathway is represented by an effective source latitude, which affects the transport distance, cumulative moisture loss and precipitation along the pathway, and thus the extent of aerosol wet scavenging. We use an effective source latitude of 55°N for sea-salt tracers, representing major open-ocean and sea-ice sources in the North Atlantic, and 45°N for all continental species (i.e., dust and BB tracers), representing major BB sources in North America and dust sources in Asian deserts. Sensitivity tests were performed by varying each effective source latitude by $\pm 5^\circ$ to assess the influence of this assumption on the modeled relationships. The height of each curve along the ordinate is controlled by the assumed source strength, which was adjusted by visual inspection so that the modeled curve passed approximately through the center of the observed data distribution (for proxies with no clear clusters) or the cluster associated with warm periods (for proxy with two distinct clusters, such as the dust tracers).

Source-related residual reconstruction

The simplified aerosol transport and deposition model provides a framework linking aerosol deposition at the ice core site to source region aerosol loading through transport and removal processes. Here we combine this framework with the multi-proxy ice core records to estimate the residual aerosol variability after accounting for modeled transport and deposition effects.

We first calculate the sensitivity of the deposition strength at GISP2 site to source-region aerosol loading for each aerosol species. These modeled sensitivities depend on both site and source temperatures and vary across the GS-GI D-O transitions. We then applied the time-variant sensitivities to the measured aerosol concentrations in ice to estimate the residual variability not explained by the modeled transport and deposition changes. Under the simplifying assumptions of this framework, this residual variability may provide an upper-bound estimate of source-related changes, although it may also include contributions from source-region shifts or model structural uncertainty.

Figures

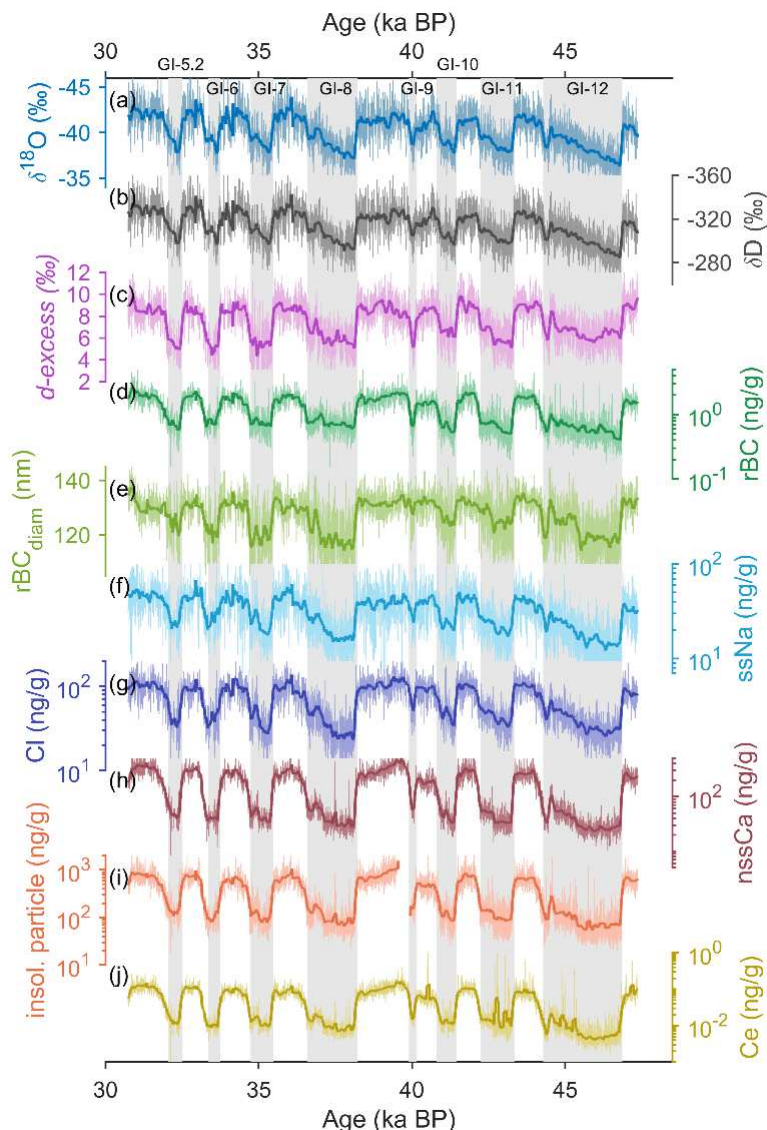


Figure 1. Water isotope and aerosol records measured in the Greenland GISP2 ice core for the 30.7-47.4 ka BP interval. Lighter-colored lines show data at annual resolution for (a) $\delta^{18}\text{O}$; (b) δD ; (c) deuterium excess ($d\text{-excess}$); (d) refractory black carbon mass concentration (rBC); (e) rBC geometric mean diameter (rBC_{diam}); (f) sea-salt sodium (ssNa); (g) chloride (Cl); (h) non-sea-salt calcium (nssCa); (i) total insoluble particle mass concentration; and (j) cerium (Ce). Darker-colored lines show 100-year moving averages. Grey shaded areas represent the Greenland interstadials (GI) as given by Rasmussen et al., 2014⁴⁶. Note the reversed axes in (a) and (b) to emphasize the anti-phase relationship of the water isotopes with the aerosol records.

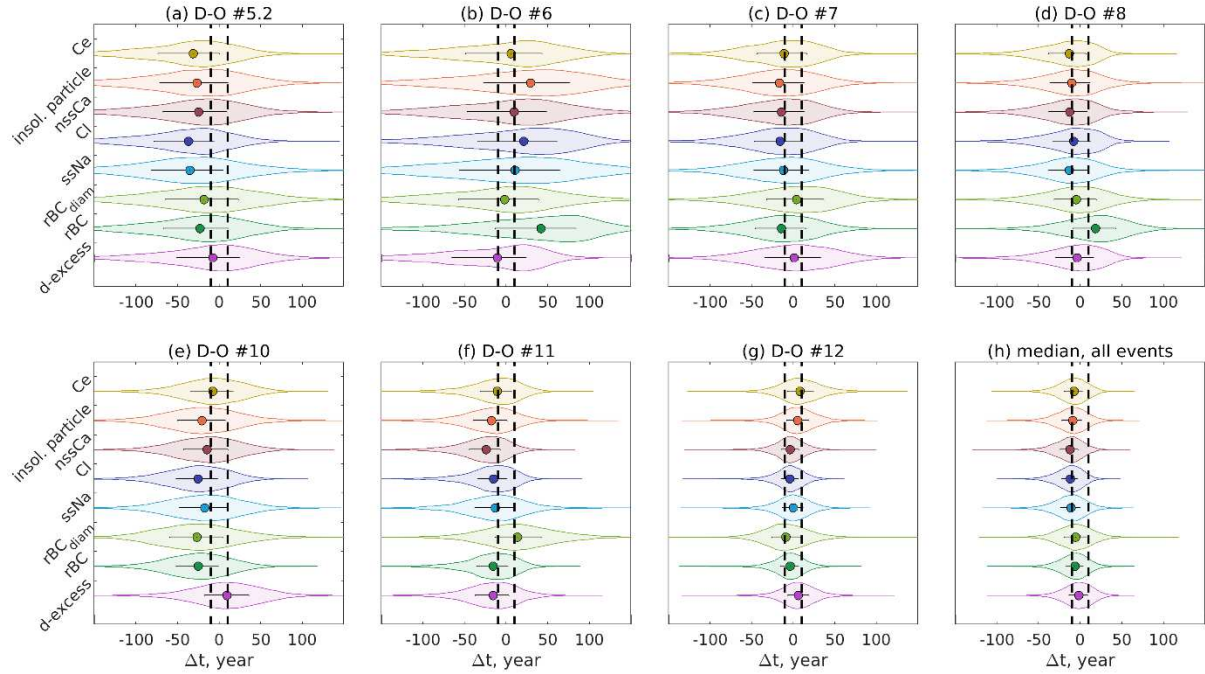


Figure 2. Onset timing differences between ice-core $\delta^{18}\text{O}$ and other proxies for the seven individual events examined in this study and the median across all these events. Positive values indicate that the proxy leads $\delta^{18}\text{O}$, while negative values indicate that the proxy lags $\delta^{18}\text{O}$. Colored markers show median estimates, error bars represent the 25th to 75th percentile ranges, and the shaded envelopes represent the full ranges of the estimates. Dashed lines indicate timing differences of ± 10 years, a reference scale comparable to the typical fitting bias estimated from synthetic tests (Text S2). Across events, most aerosol proxies changed within a narrow temporal window relative to $\delta^{18}\text{O}$, with no consistent lead-lag pattern among proxy classes, suggesting rapid atmospheric and hydrological reorganization. Note that D-O event 9 was covered by the observational record but was not included in the transition-timing analysis (see Methods).

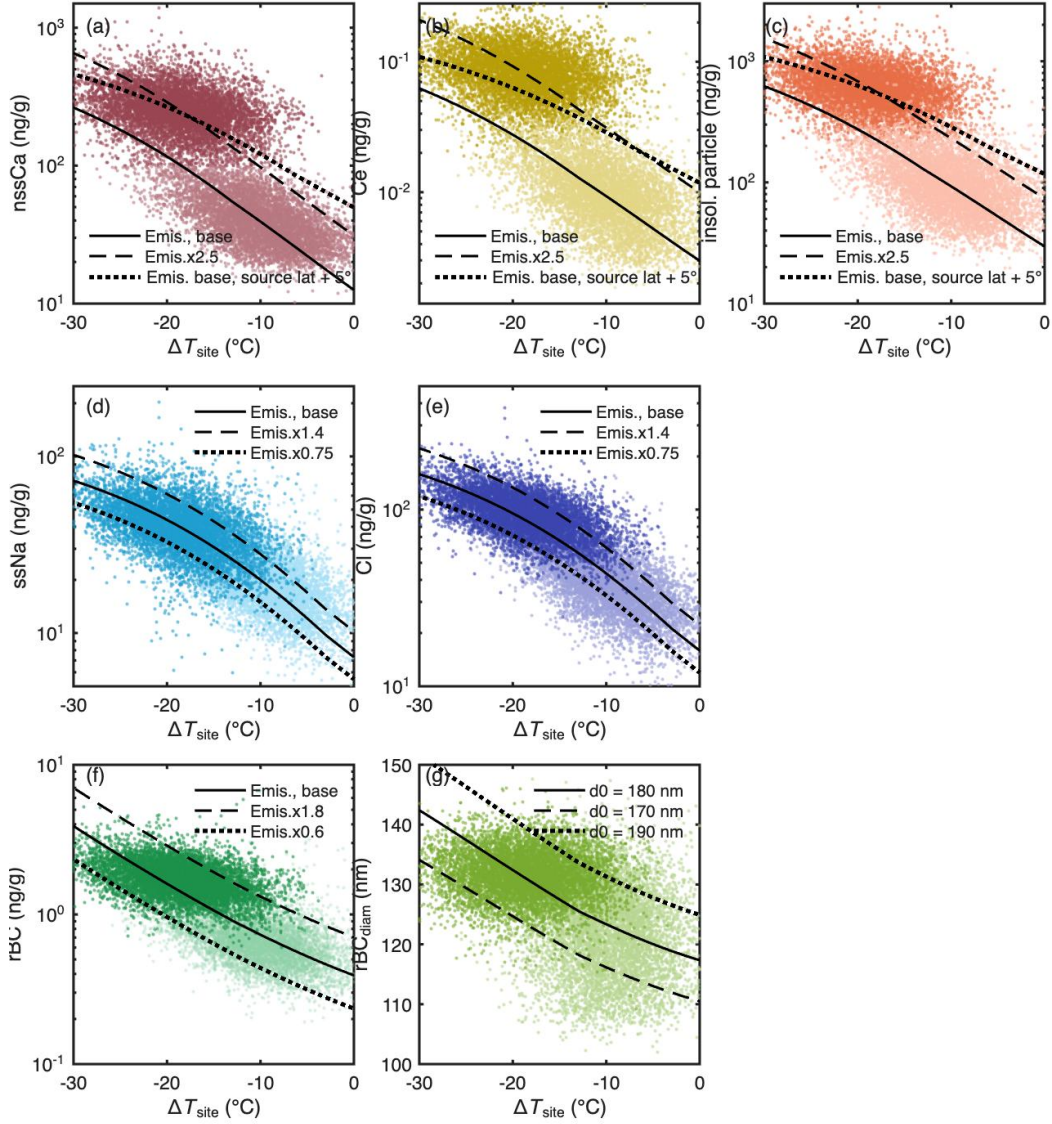


Figure 3. Comparison between modeled and measured aerosol-temperature relationships for different aerosol proxies. (a) nssCa; (b) Ce; (c) insoluble particles; (d) ssNa; (e) Cl; (f) rBC; (g) rBC_{diam}. Curves for the concentration proxies (a-f) show the base simulations (solid) and sensitivity runs for varied sources (dashed and dotted). In panel (g), the curves show sensitivity simulations with different initial rBC geometric mean diameters (d_0) assumed in emissions. In each panel, the darker and lighter colors indicate two clusters broadly corresponding to cold and warm states, identified using k-means clustering based on the nssCa records.

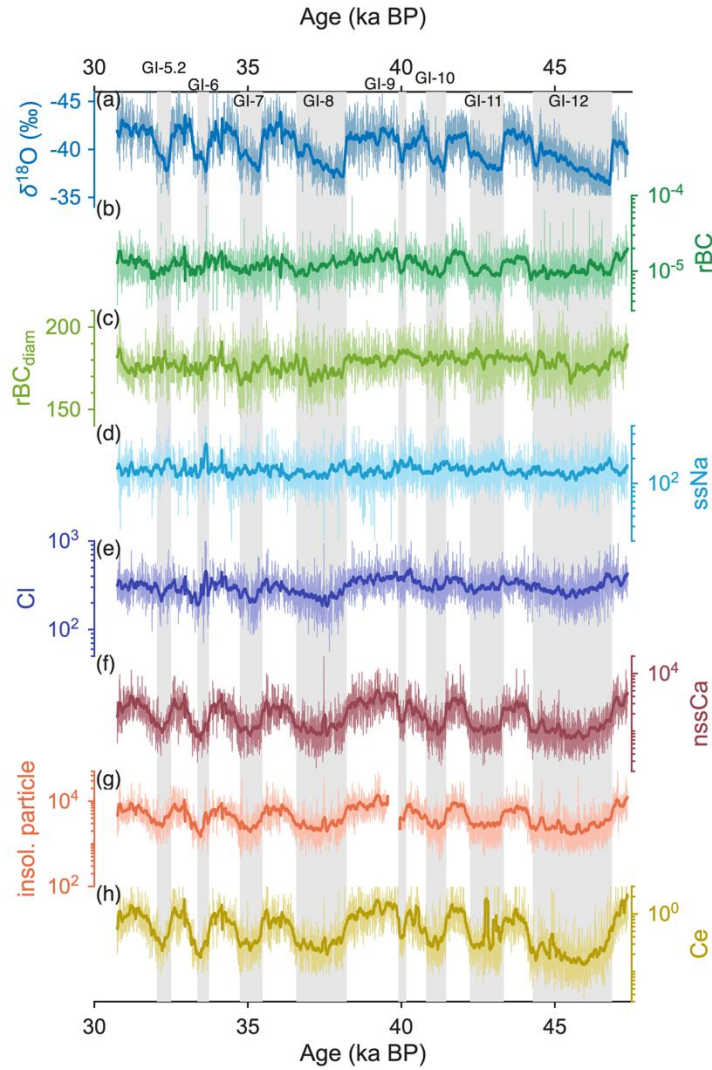


Figure 4. Residual variability in the ice core records after removing simulated effects of transport and aerosol deposition for (b) rBC, (c) rBC_{diam}, (d) ssNa, (e) Cl, (f) nssCa, (g) insoluble particles, (h) Ce. Residuals for the aerosol proxies are shown as dimensionless relative indices, such that proportional rather than absolute changes are physically meaningful. The $\delta^{18}\text{O}$ record in (a) is shown as a reference for temperature variability, with the y-axis reversed. Lighter-colored lines show data at annual resolution; darker-colored lines show 100-year averages. Grey shaded areas represent the Greenland interstadials (GI) as given by Rasmussen et al., 2014⁴⁶. By removing the modeled common transport-and-deposition response, the residual time series reveal variability with temporal patterns that differ among aerosol classes.

Data availability

All ice core data used in this work has been deposited in the Arctic Data Center under DOI: 10.18739/A29Z90D5T and will be made publicly available upon acceptance of the manuscript.

Acknowledgments

We thank Dr. Monica M. Arienzo and the students and staff of the DRI Ice Core Laboratory for their assistance with the ice core measurements, and we thank the U.S. National Science Foundation Ice Core Facility (NSF-2041950) for ice core sampling assistance and curation. This research was funded by the Geosciences Directorate of the NSF under grants AGS-2102917, 2102918, 1702814, and 1702830 and grant AWD-002731.

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