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Article

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Global chemical weathering dominated by continental arcs since the mid-Paleozoic

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Summary

Earth’s plate tectonic activity regulates the carbon cycle, and hence, climate, via volcanic outgassing and silicate-rock weathering. Mountain building, arc-continent collisions, and clustering of continents in the tropics have all been invoked as controlling the weathering flux, with arcs also acting as a major contributor of carbon dioxide (CO₂) to the atmosphere. However, these processes have largely been considered in isolation when in reality they are all tightly coupled. To properly account for the interactions between these processes, and the inherent multi-million-year time lags at play in the Earth system, we need to characterise their complex interdependencies. Here we analyse these interdependencies over the past 400 million years, using a Bayesian network to identify primary relationships, time lags and drivers of the global chemical weathering signal. We find that the spatial extent of continental volcanic arcs—the fastest-eroding surface features on Earth—exerts the strongest control on global chemical weathering fluxes. We find that the rapid drawdown of CO₂ tied to arc weathering stabilises surface temperatures over geological time, contrary to the widely held view that this stability is achieved mainly by a delicate balance between weathering of the seafloor and the continental interiors.

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Figure 1. Tectonic, atmospheric and ocean chemical changes over the past 400 Myr. a, Continental distribution with continental landmasses shown in pink, present-day coastlines in black, and the tropics (±20° of the equator) in beige; b, atmospheric CO₂ concentration (multi-proxy, black line) and phytane-based estimates in red; c, continental ice latitude is shown as the blue line (blue shaded regions denote glaciations); d, seafloor production rates (Methods); e, suture zone lengths; f, fragmentation index (i.e., continental perimeter/area, as black line), and total area of continents in the tropics (red line); g, \(\frac{87}{86}Sr/\frac{86}{86}Sr\) from marine carbonates, calculated as a ±0.25 Myr window in red; h, normalised \(\frac{87}{86}Sr/\frac{86}{86}Sr\) curve removing the signal caused by radioactive \(87\)Rb decay in the crust.
moving average using a ±0.25 Myr window (Fig. 1g). Given the large contrast in $^{87}\text{Sr}/^{86}\text{Sr}$ between radiogenic continents and unradiogenic oceanic crust, the $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ is thought to represent a globally integrated balance in weathering flux from continental surfaces and the seafloor. We tested this assertion by constructing a network to explore the relationship between $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ and partial pressure of atmospheric carbon dioxide ($p\text{CO}_2$) since 400 Ma. This analysis reveals a clear relationship between $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ and $p\text{CO}_2$ (empirical correlation = 0.57) at lag 0, which decreases with increasing lags (Extended Data Fig. 1), confirming that they are coupled. However, as there are clearly secondary controls on $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$, and some uncertainty in the CO$_2$ record used, the relationship is not straightforward; this is a key justification for analysing what drives these variations through time. We present our results in terms of $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$—the standard framework—but also test the sensitivity of our model to radioactive decay of $^{87}\text{Rb}$ (to $^{87}\text{Sr}$) in the crust through time.

**Building an Earth network**

Despite progress in linking variations in $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ to geodynamic and paleogeographic factors, it is unclear how processes combine to drive $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ variations. We constructed the network (Methods) with nodes for $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ and twelve predictor variables (Supplementary Data File S1), with lags from 0 to 50 Myr. We present three correlation measures that summarise the relationships between the variables and $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ (Figs. 2–3). First, the empirical rank (or Pearson product-moment) correlation ($C_{\text{Emp}}$) measures the linear relationship between two variables. Although informative, this does not account for autocorrelation, or the joint influence of other variables. Second, the BN rank correlation ($C_{\text{BN}}$) is the modelled representation of the empirical rank correlation. In an ideal case (i.e., a perfect model fit) this would be equal to $C_{\text{Emp}}$. Third, the conditional rank correlation ($C_{\text{Cond}}$) is the correlation between two variables conditional on any other parent variables (accounting for the effect of all nodes at shorter lags, and higher up in the network hierarchy; Methods).

We construct our network by starting with the variable with the highest empirical correlation (at lag 0), and systematically search the set of predictor variables to find maximum values of $C_{\text{Cond}}$ at increasing lags, up to 50 Myr (Methods). A variable is added to the network if its conditional correlation exceeds a confidence interval threshold (dependent on the number of original data points). The conditional correlation removes the influence of variables higher in the network hierarchy (and adds earlier lags), and provides a measure of the additional information each subsequent lagged variable provides in explaining $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ variation (Fig. 3; Extended Data Fig. 2). This approach is based on the method for partial autocorrelation, and efficiently accounts for multiple joint dependencies and lags (Methods). Whilst our focus below is on $C_{\text{Cond}}$, for context we also provide $C_{\text{Emp}}$ and $C_{\text{BN}}$ (Fig. 3).

**Identification of chemical weathering drivers**

We find that the spatial extent of continental volcanic arcs is strongly correlated with $^{87}\text{Sr}/^{86}\text{Sr}_{sw}$ ($C_{\text{Emp}} = -0.79$; $C_{\text{Cond}} = -0.7$; Figs. 2, 3a), increasing when we correct for crustal radioactive decay of $^{87}\text{Rb}$ (ref. 16; $C_{\text{Emp}} = -0.82$; Fig. 2a; Extended Data Table 2). This strong relationship (Fig. 2) suggests that periods of increased continental arc volcanism have favoured unradiogenic seawater compositions, and vice versa. Today, the global continental arc system is ~14,000 km long, and includes regions such as the Alaska Peninsula, the Cascades and the Andean Volcanic Belt (Extended Data Fig. 4). The global arc system was three times longer (~37,500 km) during the Mesozoic (Fig. 1c), reflecting a sharp increase in seafloor production (Figs. 1c–d). The ocean chemical response to changing arc extent is rapid, peaking in...
Figure 3. Simplified network structure showing key geological processes and correlations with seawater Sr

Graphical representation of our network, showing how the six dominant variables (a–f) influence \(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\)\(_{sw}\) (Extended Data Fig. 2). The plots summarise the relationships between the relevant variable and \(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\)\(_{sw}\) for all time steps in our analysis (n = 360). The plots show C\(_{\text{Emp}}\), C\(_{\text{BN}}\), and C\(_{\text{Cond}}\), at time lags from 0 to 50 Myr in 2.5 Myr intervals. A lag of 0 means the relevant process is occurring within the same 1 Myr time-step. The values shown in gray on the plots are the highest value of C\(_{\text{Emp}}\); if each process were considered in isolation this value would represent the dominant time lag. However due to autocorrelation and joint dependence, the dominant processes and time lags can be better identified by peak C\(_{\text{Cond}}\) (red). The horizontal dashed lines denote 99% confidence intervals.
Terrestrial weathering fluxes are highly sensitive to crustal deformation. It has been suggested that arc-continent collisions in the tropics led to enhanced weathering of ophiolites and CO₂ drawdown, driving Phanerozoic glaciations. Weathering of ultramafic lithologies on this scale should reduce (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\), reflecting unradiogenic inputs to oceans. To evaluate this, we incorporate existing suture length data. The land surface area within the tropics has increased (Fig. 1f; Extended Data Fig. 6), and integrate this in our net-

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It is well established that seafloor basalt alteration and hydrothermal venting decrease (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\), (i.e. toward mid-ocean ridge basalt [MORB] mantle (\(^{87}\text{Sr}/^{86}\text{Sr}\) ≈ 0.7035; ref. 22). Therefore, (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) is expected to scale inversely with seafloor production rate (Fig. 1d), which we calculate as the product of ridge length and spreading rate (Extended Data Fig. 7), adapting an existing plate model. We find that seafloor productivity is negatively correlated with (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) at short lags (Fig. 3d), reflecting the effects of early high temperature alteration of basalts along ridge axes. The seafloor weathering contribution becomes negligible ∼15–20 Myr after emplacement, suggesting that seafloor is not weathered appreciably after this time. This is strikingly consistent with hydrothermal models and observations of secondary minerals in ocean crust, which indicate that ~70–80% of fluid flux occurs in seafloor within 20 Myr of formation. We find that radiogenic continental weathering sources dominate the (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) signal at lags >20 Myr, explaining the switch to a positive correlation (Fig. 3d).

Glacial ice is also known to influence (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\). Investigating the effect of continental ice coverage, we find a strong empirical correlation between latitudinal extent of ice sheets (Fig. 1b) (as a proxy for the severity of glaciation and global climate) and (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) (Fig. 3c; Extended Data Fig. 8), supporting the notion that glaciations cause intensified weathering of continental crust. This is likely due to preferential weathering of radiogenic minerals like biotite in comminuted rock flour characteristic of glaciated catchments. The conditional correlation is low due to collinear-

High temperatures and precipitation usually favor high weathering rates in tropical regions. It has thus been hypothesized that a high proportion of continental landmasses within the tropics could strongly influence global weathering. We tested this by quantitatively evaluating how other processes combine to drive (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) observations, need to quantitatively evaluate how other processes combine to drive (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) than enhanced weathering of mafic lithologies.

Central role for volcanic arc weathering

Our analysis indicates that continental volcanic arc weathering exerts the strongest influence on (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) as a proxy for global chemical weathering, consistent with regional studies, and with the observation that chemical weathering of island arcs dominates the oceanic Sr budget today. Continental arcs are an important contributor to the atmospheric CO₂ inventory, owing to a propensity for decarbonation reactions in the continental lithosphere. The strongly negative correlation between continental arc length and (\(^{87}\text{Sr}/^{86}\text{Sr}\))\(_{\text{sw}}\) (Figs. 2 and 3a) is consistent with the hypothesis that their formation and spatial extent governs icehouse-greenhouse transitions.

The high CO₂ outgassing flux and greenhouse conditions associated with extensive continental arcs favour intensified chemical weathering. Today, continental volcanic arcs are among the highest topographic—and fastest eroding—surface...
features on Earth, supplying Ca-Mg silicates to the ocean, over tens of millions of years. Hydrothermal activity maximizes chemical denudation rates. For example, the present-day Andes (Extended Data Fig. 4) dominates dissolved ion fluxes to the Amazon River, fuelling Earth’s greatest offshore river plume. Today, continental arcs are predominantly unirradiogenic (Extended Data Fig. 4), with mean 87Sr/86Sr values of 0.7044±0.0025 (N = 5498; median = 0.704, mode = 0.7035), only slightly higher than typical MORB and ocean island basalts. Thus, the prolonged cycles of arc assembly, erosion and weathering, likely drove seawater toward the unradiogenic compositions we observe (Figs. 2–3). Greenhouse conditions linked to extensive arcs should promote increased bottom water temperatures and enhanced seafloor weathering, further reducing (87Sr/86Sr)sw.

Continental volcanic arcs are predisposed to acid-intermediate magmatism, favouring zircon production. The interpretation that continental arcs drive global chemical weathering fluxes (Fig. 2) is therefore consistent with an observed increase in detrital zircon abundance during greenhouse intervals. The latter implicates increased transport and weathering of arc detritus to ocean basins when continental arcs are longest. Our analysis confirms that the 87Sr/86Sr of zircon-bearng igneous rocks strongly correlates with (87Sr/86Sr)sw over the past 400 Myr (Extended Data Table 2), suggesting global chemical weathering is tightly coupled to the composition of continental igneous lithologies. The correlation between igneous 87Sr/86Sr and (87Sr/86Sr)sw is highest when arc systems are longest. It is therefore probable that the spatial extent of continental arcs drives (87Sr/86Sr)sw via the proportional availability of weatherable igneous catchments. This finding draws specific attention to the types of rock and climate conditions that offer the best potential for accelerated CO2 drawdown in enhanced weathering schemes designed to counteract current global climate change.

Summary

We have developed a new data mining approach based on conditional probability estimation, to disentangle complex interdependencies between solid Earth, hydrosphere, and atmospheric processes, over the past 400 million years. This approach has significant potential to aid interpretation of complex Earth data exhibiting high dimensional dependency on different spatial and temporal scales. It is widely accepted that continental arcs modulate atmospheric CO2 levels and represent a major agent of crustal growth through post-Archean Earth history via arc accretion processes. Our analysis indicates that arcs have also dominated global chemical weathering fluxes, which determine the Sr isotopic composition of seawater, (87Sr/86Sr)sw, over the past 400 Myr. This revises conventional concepts that (87Sr/86Sr)sw is driven by competition between weathering of the seafloor and continental interiors. Arc weathering causes reduction in (87Sr/86Sr)sw, while continental weathering causes increase in (87Sr/86Sr)sw. Thus, our findings of arc dominance in weathering help explain enigmatic low (87Sr/86Sr)sw during greenhouse climates, where the higher temperatures should according to conventional concepts have promoted greater continental crust weathering, driving increased (87Sr/86Sr)sw. Through this regulation of atmospheric CO2 levels over geological timescales, continental volcanic arcs played a central role in maintaining habitability over the course of Earth history even in the face of dramatic external drivers.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s12345-111-2222-3.

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Methods

1.0. Bayesian Network analysis

Here we outline the methodology for our Bayesian Network analysis. We use the UNINET COM library in Visual Studio C++ to perform data mining on the time-series detailed in section 2.0 (below). The approach enables the identification of dominant correlations for a range of geophysical and geochemical variables with the strontium isotope ratio of seawater through geological time as a proxy for global chemical weathering. All initial data processing and GIS analysis is performed.

Our analysis considers data for the time period from 410 Ma to 0 Ma, using regularised time-series with a time step of 0.5 Ma. These time-series are provided in Supplementary Data File S1 (Fig. 1). As many of the individual time-series are based on measurements, estimates or model values irregularly spaced in time, we interpolate to obtain regular (0.5 Myr) time-series to match the resolution of the CO2 record used. For cases with multiple observations in a given time interval, a moving average is applied (again with a 0.5 Myr window). The number of original data points before interpolation (and accounting for lags) is used to estimate confidence interval thresholds for each of the variables with respect to the baseline (87Sr/86Sr)sw. The CI (in this case the 99 percentile) threshold determines whether or not a particular lagged variable is retained in the model (described in detail in section 4.0). We also generate multiple lagged series using average values for each of the variables in 2.5 Myr intervals, up to 50 Myr. A lag of 0 means the processes occur within the same time step (t, Myr), a lag of 2.5 denotes the average value from t-2.5 to t-0.5 Myr etc.

It is important to note that we also repeated the BN data mining using 1 Myr interval input data, and the differences in the results were negligible. We impose CI thresholds based on the original number of observations, not the length of the interpolated time-series.

2.0. Network Nodes

The principal aim of our analysis is to identify the variables that are most strongly correlated with (87Sr/86Sr)sw (the ‘node of interest’), and at what time lags, in order to quantitatively understand the primary geological processes driving chemical weathering. We used the variation in (87Sr/86Sr)sw ratios derived from marine carbonates (Fig. 1g). These raw (87Sr/86Sr)sw data produce an irregular time-series with around 4,000 observations spanning 519.4 to 0 Ma, with a maximum interval of 10.2 Myr. To generate a regular time-series for our ‘node of interest’ we took a moving average of (87Sr/86Sr)sw using a rolling window of ±0.25 Myr. We do not consider lagged values for Sr here, as we are primarily interested in identifying the processes that lead (87Sr/86Sr)sw. We do however present a separate analysis of the relationship between lagged (87Sr/86Sr)sw and CO2 (Extended Data Fig. 1).

The variables investigated are:

1. Plate tectonic fragmentation: Continental areas were extracted from shapefiles generated by the open-source plate tectonic reconstruction software GPlates. We used the plate tectonic reconstruction of Matthews et al. for extracting latitudinal and time-sensitive data for our analysis. This plate model is a synthesis of the Domeier and Torsvik model for the Late Palaeozoic and the Müller et al. model for the Mesozoic and Cenozoic. All data were extracted with the plate model in a palaeomagnetic reference frame, and the output comprises georeferenced maps of continent boundaries at 1 million year (Myr) intervals from 410 Ma to the present.

Areas were calculated by taking continental polygons (which characterise the shape and location of all the continental landmasses) from the GPlates shapefiles, and splitting them into 6 bands according to latitude (90–20°N, 20–10°N, 10–0°N, 0–10°S, 10–20°S, 20–90°S). The Rsaga package function rsaga.intersect.polygons is used to split the continental shapes by latitude, working in WGS84 global reference system coordinates (EPSG4326). The total continental area within each latitude band is calculated by first ‘dissolving’ and cleaning the polygons using the rsaga.geoprocessor function shapes–polygons, then calculating the total area using the areaPolygon function from the R geosphere package. Area is calculated in m², and—accounting for the accuracy of shapefiles and the coordinate transformation from longitude/latitude—is considered accurate to approximately 2 significant figures. This is reasonable given the model uncertainty and resolution.

We measured the perimeter of continental landmasses through time using the perimeter function from the R geosphere package, and adapted the method of Cogné and Humler and Merdith et al. to compute the continental perimeter/area ratio—a measure of how “fragmented” the continents are through time (Fig. 1f; Supplementary Data File S1). In contrast to ref.26, we do not apply a minimum area threshold, as the time period we are considering, we only require the reconstructions of Matthews et al. and do not need to incorporate other plate models of variable spatial resolution. We do however remove ‘holes’ with areas <5 x 10¹¹ m², primarily to eliminate the extremely narrow void spaces (slithers) that occur where adjacent continental polygons never fully join due to their geometry and resolution in the model. Leaving these ‘holes’ in place significantly inflates the perimeter estimate at certain time steps, and they can be clearly identified as erroneous gaps from inspection of the individual shape-files. Units of fragmentation are m⁻¹ (perimeter/area), and are extracted at 1 Myr intervals, then interpolated to 0.5 Myr.

2. Land surface area within the tropics (a:±10°, and b:±20° of the equator): The area of the continental land surface (a) within 10° degrees of the equator (i.e., the tropical rain belt) and (b) ±20° of the equator were measured as described above, again using shapefiles exported from GPlates and processed in R. The latitudinal distribution of continental landmasses (within both ±20° (Fig. 1f) and ±10° bands) and the fractional areas of continental landmasses in the equatorial bands (i.e., area within the belt...
Seafloor production rates: We calculated seafloor production rates as the product of ridge length and spreading rate of each discrete spreading segment (i.e., each mid-ocean ridge segment separated by a transform boundary) through the segment midpoint (Extended Data Fig. 7). If the spreading direction (i.e., stage pole orientation) that passes through the segment midpoint and the great circle of the tangent to the midpoint of the spreading segment and measuring the angle between this and the great circle of the stage pole orientation (i.e., spreading direction) that passes through the segment midpoint (Extended Data Fig. 7). If this angle exceeds 70°, it is assumed to represent a spreading segment, and the full spreading rate was extracted and multiplied by the length of the segment. The sum of all segments × full spreading rate was calculated at 1 Myr intervals to give total seafloor production. Further details on this approach are provided in Extended Data Fig. 7. A key uncertainty in the construction of seafloor production rates is that very little oceanic lithosphere older than 200 Ma is preserved today. However, our analysis does not concern time sensitive evolution of oceanic lithosphere (such that is required for understanding how oceanic volcanism changes through time, or the delivery of volatiles to subduction zones, for instance). Instead, we just require a measured or inferred length value of the arcs (described in the Supplementary Information accompanying Cao et al.14). It must be noted that, for the present-day, these estimates omit several areas experiencing continental subduction (e.g., under Zealandia, flat slab subduction in the Andes). In most cases, the minimum and maximum lengths are equal, and in cases where these deviated we used the average length value—noting that the difference between the minimum/maximum and the average length is rather low (<11.5%) during the period of interest (Fig. 1c). The time-series (units: km) is regular with a 1 Myr interval, interpolated to 0.5 Myr.

Suture length: We used a database of suture zone length that records sites of ophiolite obduction during arc-continent collisions. Here, the suture zone lengths were estimated using the observed spatial extent of ophiolites based on published geological maps and global lithological compilations. Macdonald et al.5 reconstructed the locations of suture zones throughout the Phanerozoic using paleogeographic models. They estimated the duration of suture zone activity using the onset of ophiolite obduction (as evidenced by the first occurrence of arc exhumation), which they defined as the first appearance of ophiolite-derived detritus in the foreland; and the termination of foreland deposition was taken to mark the cessation of ophiolite obduction (procedure is described in the Supplementary Information accompanying ref.5). The time-series (units: km) is regular with a 5 Myr interval, interpolated to 0.5 Myr.

Atmospheric CO₂ concentration: We used a compilation of the partial pressure of atmospheric carbon dioxide (pCO₂) for the past 420 Ma derived from multi-proxy measurements (N=1241; from the literature and covering five independent techniques; ref.12). Foster et al.12 used a set of criteria to screen and standardise these records, and applied Monte Carlo resampling and a local polynomial regression (LOESS) fit to the resulting data series. We used the maximum probability pCO₂ data from ref.12 (Supplementary Data File S1), with associated 68 and 95 percentile ranges (Fig. 1b). We note that recent phytane-based measurements13 are in reasonable agreement with this long-term pCO₂ record (Fig. 1b). The time-series from Foster et al.12 (units: ppm) is regular with a 0.5 Myr interval.

Latitudinal ice extent: We used a recent compilation of
the latitudinal extent of continental ice sheets (e.g., 90°S = no ice sheets) from Macdonald et al. This compilation is based on a literature review of the geological constraints on glaciation during the Phanerozoic, and the inferred paleogeographic extent of continental ice using updated age constraints (a full discussion of this database is described in the Supplementary Information accompanying ref 5). The time-series (units: degrees) is regular with a 1 Myr interval interpolated to 0.5 Myr.

8. Subduction zone length: The total length of subduction zones (including oceanic arcs) through geological time, were taken from Matthews et al., and extracted using pyGPlates at 1 Myr intervals. Spatial and temporal constraints on the distribution and extent of subduction zones are provided by geological constraints such as, for instance, ophiolites, subduction-related magmatism and the occurrence of high pressure metamorphic lithologies that are consistent with subduction processes.

As noted by Merdith et al., there are some similarities in the trends of subduction zone and continental arc lengths, lending support to these independently-derived measures. The time-series is regular at 1 Myr intervals (units: km), interpolated to 0.5 Myr.

9. Active LIP area: We used the area of Large Igneous Provinces (LIPs) actively erupting at a particular 1 Myr time step from the compilation of Johansson et al. This database, which includes continental and oceanic LIPs, was compiled and digitised from the literature and their locations were reconstructed using GPlates software. An underlying assumption of this time-series is that the LIPs were active for a total period of 3 Myr after their accepted eruption age. The time-series is regular at 1 Myr intervals (units: km²), interpolated to 0.5 Myr.

10. Weatherable LIP area in the tropics: We also use the area of LIPs (active or inactive) exposed within 15° of the equator at a particular 1 Myr time step. Johansson et al. applied paleogeographic reconstructions to discriminate between continental and oceanic LIPs in order to isolate late exposed (continental) LIPs within the tropics. This regular time-series (1 Myr interval, units: km²) provides a minimum estimate of LIP area through time.

11. Igneous Sr ratio: We use the 87Sr/86Sr ratio of zircon-bearing igneous rocks (i-zig) over the last 400 Ma, from the compilation from Bataille et al. that spans 1000 Ma, to assess the relative contribution of continental igneous rock lithologies (i.e., dominantly continental volcanic arcs, which are the locus of zircon formation) to (87Sr/86Sr)sw. The authors applied a bootstrap resampling approach to correct for geographic/sampling biases in the detrital zircon record comprising 24,715 individual zircon grains. Bataille et al. used the relationship between the eHf component positions of zircons and the eSr of their igneous host rocks to estimate the secular variations in the (87Sr/86Sr)sw through time—reflecting the changing proportion of juvenile and reworked materials generated during orogeny.

We re-ran code from Bataille et al. using a modified smoothing window of 5 Myr, and increment and scale of 0.5 Myr (previously 10 Myr and 1 Myr, respectively, in Bataille et al.). Additionally, we applied an adaptive window (decreasing in size) for data points between 5 and 0 Ma, to enable extension of the time-series to 0.5 Ma.

3.0. Auto- and Cross-correlations

Many of the studied variables are strongly auto-correlated and cross-correlated (e.g., due to being different proxies for the same or related processes/states). This makes it very difficult to identify dominant driving processes and their time lags. It is straightforward to compute partial autocorrelations for individual parameters—a standard approach in time-series analysis, however the multivariate case cannot always be solved. We tested whether the multivariate partial autocorrelation could be computed for our data set using an R implementation of the PACE38 (function acf/pacf in the stats package). This function computes the partial lag autocorrelation matrix P(s) of Heyse and Wei (pp 411-414 in Wei57), where P(s) is the autocorrelation between Z and Zt-s after removing the linear dependency on the vectors at intervening lags Zt-1, Zt-2, ..., Zt-s-1. The elements are normalised correlation coefficients. Based on this analysis, we concluded that the multivariate PACF38 could not successfully be computed for our data.

To provide an alternative means of accounting for the combined effect of multiple parameters (at varying lags) on the variable of interest (87Sr/86Sr)sw, we developed a novel method based on conditional correlation, estimated using Uninet50. The approach we employ is similar in principle to the multivariate partial autocorrelation, but evaluates the conditional correlation for variables added iteratively to the BN at increasing time lags.

Code is written in C++ and uses the Uninet Windows COM library. Uninet is a software package for uncertainty analysis and dependence modelling for high dimensional distributions, originally developed for the CATS (Civil Aviation Transport Safety) project. It is available as a standalone application, and as a Windows COM library (the Unnet Engine) enabling alternative programming interfaces, including but not limited to: R, Matlab, and Visual Studio/C++ (used here). Uninet models empirical multivariate distributions by building a joint density function from a set of inputs (data mining). Joint dependency is represented by conditional rank correlation, using the joint normal copula.

4.0. Summary of the data mining algorithm

Input data are time-series for the variable of interest X, (which in this instance is (87Sr/86Sr)sw) and the 12 predictor variables A, B, C, ... L (observables listed in section 2.0), plus lagged values of those variables A-2,5, A-5,0 etc. Nodes are grouped and evaluated in order of increasing lag, giving a set of observables (A, B, C, ... L) at lag 0; (A-2,5, B-2,5, ... L-2,5) at lag 0.5 Myr to t-2.5 Myr etc., up to 50 Myr.

We construct the network by starting with the unlagged variable with the highest empirical correlation with (87Sr/86Sr)sw, then systematically search through the set of remaining predictor variables to find maximum values of conditional correlation.
(\(C_{\text{Cond}}\)) at increasing time lags. At each iteration we look for the largest \(C_{\text{Cond}}\) as this identifies the variable that individually provides the maximum additional information at each step. The variable with the largest \(C_{\text{Cond}}\) is subsequently added to the network if all of these following conditions are met:

1. Its conditional correlation \(C_{\text{Cond}}\) (the correlation with \((^{87}\text{Sr}/^{86}\text{Sr})_{sw}\), conditional on all other variables in the network) exceeds a specified confidence interval threshold. Here we use the 99% CI, with the threshold depending on the number of observations used to generate the time-series for the variable in question. This results in a higher threshold for lower resolution variables.

2. The difference between the (unconditional) empirical rank correlation \(C_{\text{Emp}}\) and BN (i.e., modelled) rank correlation \(C_{\text{BN}}\) with \((^{87}\text{Sr}/^{86}\text{Sr})_{sw}\) is less than 30%. This eliminates variables that cannot be represented accurately by the BN (using normal copulae) and prevents such nodes affecting the estimates of conditional dependence for subsequent nodes.

3. The variable is not highly correlated (a correlation of 0.8 or greater) with any existing variables in the network (i.e., nodes higher up in the network hierarchy). This reduces the effect of collinearity.

This procedure is repeated for each discrete time lag, resulting in the lagged variables being either added to the network (in order of decreasing conditional correlation and increasing lag, respectively), or rejected for not meeting one of the three criteria above. These steps ensure the construction of a parsimonious model where only the most informative nodes are retained. We present the calculated Empirical, BN (modelled) and Conditional (modelled) rank correlations for each variable and time lag in Figure 3 and Extended Data Figure 2.

The computational efficiency of Uninet means that this approach is suitable for application to large numbers (of the order hundreds to thousands) of variables that cannot be represented accurately by the BN (using normal copulae) and prevents such nodes affecting the estimation of conditional dependence for subsequent nodes. This prioritises inclusion of nodes that can be represented most accurately by the BN.

4.1: Itemised steps to construct the network:

1. Generate a saturated BN using all variables (nodes) with lag 0 (\(A_1, ..., L_n\)) and identify the node with the largest empirical correlation \(C_{\text{Emp}}\) with the variable \(X_i\) (node of interest \(X_i\)). This node (e.g., \(C_1\)) individually gives the most information about \(X_i\), so it is placed first in the network hierarchy. The variable of interest, \(X_i\), always remains last in the hierarchy, as we are interested in computing the probability of \(X_i\) given all the other observed variables. (NB: A saturated BN contains arcs linking every pair of nodes in the network). The first stage network is as follows:

2. Step through all the remaining nodes with lag 0 (\(A_1, ..., L_n\)) and add them one by one, as a second “test node” in the network (see figure below). Calculate the conditional rank correlation \(c\) for each test node in turn e.g., \(c = C_{\text{Cond}}(X_i, A_l | C_i)\).

This prioritises inclusion of nodes that can be represented most accurately by the BN.

3. Find the node that has the largest (absolute) value \(c\)’s \((c_{\text{max}})\). If the conditional correlation \(c\) is below the specified confidence interval threshold set for this particular node, or if \(p > 0.3\) (the BN and empirical correlations differ by more than 30%), the node is eliminated and not added to the network (in this case, move to step 5).

4. Reduce collinearity (high correlations between predictor variables) as follows: If the node giving \(c_{\text{max}}\) has not already been eliminated in step (3) calculate the empirical correlation of this node with all other nodes higher in the BN hierarchy (i.e., all nodes other than the node of interest, \(X_i\)). If this returns an empirical correlation greater than a given threshold (in this case 0.8) indicating high collinearity, the node is eliminated and not added to the network. If the empirical correlations are all below 0.8, add the node to the network and proceed to step (5).

5. Repeat steps (2) through (4) with all remaining lag 0 nodes, until they are all either added to, or eliminated from the network according to the steps above.

6. Repeat steps (2) through (5) using the set of nodes with lag 0.5–2.5 Myr (\(A_{r-2.5}, B_{r-2.5}, ..., L_{r-2.5}\)) etc., up to the maximum lag (50 Myr).

Using the resulting network, we can then compute and plot the conditional rank correlation for each variable at increasing lag, having effectively removed both the effect of shorter period lags, and other more informative predictor variables (see Fig. 3; Extended Data Fig. 2).

Please note that an additional reference is cited in an Extended Data Figure.

**Data availability**

All data generated or analysed during this study are provided in the online version of this article (Supplementary Data Files S1–S2) and in Extended Data Tables 1–2.
Source data

**Supplementary Data File S1:**

Time-series compilation of all data used in our network, spanning the period from 410–0 Ma. This includes (a) the predictor variables, which are: continental arc length, sutures zone length, latitudinal extent of continental ice sheets, continental area within 20° of the tropics (this study), continental area within 10° of the tropics (this study), plate tectonic fragmentation index (this study), subduction zone length, seafloor productivity (this study), atmospheric pCO2 (ref. 12), area of LIPs within 15° of the tropics, eruptive area of LIPs, 87Sr/86Sr of continental igneous lithologies (ref. 16), and (b) the node of interest, 87Sr/86Sriso (ref. 15), as well as a normalised version accounting for radioactive decay of 87Rb in the crust through geological time (ref. 16). The records were interpolated to obtain a regular (1 Myr interval) time-series, and in cases where multiple values occurred within a single time stamp we used a moving average with a 1 Myr window.

**Supplementary Data File S2:**

S2 lists the complete set of correlations for all test nodes with 87Sr/86Sriso. Note that CEmp = Empirical Rank Correlation; CBN = BN Rank Correlation; CCond = Conditional Rank Correlation. Nodes with CCond greater than the 99 percent confidence interval threshold (Cthresh, see the Methods for further details) were retained in the BN (highlighted in green in column F). The table of correlations includes maximum CCond for nodes that were subsequently eliminated (shown in red).

**Code availability**

The numerical modeling codes associated with this paper are available from the corresponding author (Thomas.Gernon@noc.soton.ac.uk) upon reasonable request.

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**Author contributions**

T.G. conceived the idea, led the study, interpreted the data and prepared the manuscript and figures. T.H. performed the modeling, designed the network and carried out the analysis, with input from T.G. A.M. calculated the seafloor production rates, and both A.M. and D.M. provided support with *GPlates* and *pyGPlates*. M.P. and C.B. provided support with Sr isotope interpretation, and C.B. provided normalised Sr data. G.F. provided CO2 data and both G.F. and E.R. assisted with paleoclimate interpretation. T.G. wrote the manuscript with input from all co-authors.

**Competing interests:**

The authors declare no competing interests.

**Additional information**

**Supplementary information** is available for this paper at https://doi.org/10.1038/s12345-111-2222-3.

**Correspondence and requests for materials** should be addressed to T.G.

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Tectonic, atmospheric and ocean chemical changes over the past 400 Myr a, Continental distribution with continental landmasses shown in pink, present-day coastlines in black, and the tropics (±20° of the equator) in beige; b, atmospheric CO2 concentration (multi-proxy, black line), and phytane-based...
estimates in red 13; continental ice latitude 5 is shown as the blue line (blue shaded regions denote glaciations); c, continental arc length 14; d, seafloor production rates (Methods); e, suture zone lengths 5; f, fragmentation index (i.e., continental perimeter/area, as black line), and total area of continents in the tropics (red line); g, (87Sr/86Sr)sw from marine carbonates 15, calculated as a 0.25 Myr window in red; h, normalised (87Sr/86Sr)sw curve removing the signal caused by radioactive 87Rb decay in the crust 16. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
Figure 2

Effects of continental arc extent on the strontium isotopic composition of seawater. a, Ranked ordered normalised \((^{87}\text{Sr}/^{86}\text{Sr})_{sw}\) versus ranked continental arc length (see Extended Data Fig. 3 for the non-normalised and unranked versions). Note that the smallest value that occurs in the data set is ranked 1. b, Probability density for continental arc length 14, identifying short (<16,100 km), intermediate (16,100–29,300 km), and extensive (≥29,300 km) arcs (note: these divisions denote approximately equal
quantiles); the distributions show that extensive continental arc systems favour low \((87\text{Sr}/86\text{Sr})_{sw}\), and vice versa.

**Figure 3**

Simplified network structure showing key geological processes and correlations with seawater Sr j

Graphical representation of our network, showing how the six dominant variables (a–f) influence \((87\text{Sr}/86\text{Sr})_{sw}\) (Extended Data Fig. 2). The plots summarise the relationships between the relevant
variable and $(87\text{Sr}/86\text{Sr})_{sw}$ for all time steps in our analysis ($n = 360$). The plots show $\text{CEmp}$, $\text{CBN}$, and $\text{CCond}$, at time lags from 0 to 50 Myr in 2.5 Myr intervals. A lag of 0 means the relevant process is occurring within the same 1 Myr time-step. The values shown in gray on the plots are the highest value of $\text{CEmp}$; if each process were considered in isolation this value would represent the dominant time lag. However due to autocorrelation and joint dependence, the dominant processes and time lags can be better identified by peak $\text{CCond}$ (red). The horizontal dashed lines denote 99% confidence intervals.

**Supplementary Files**

This is a list of supplementary files associated with this preprint. Click to download.

- NGS20201002584TS1.csv
- NGS20201002584TS2.xlsx
- NGS20201002584TExtendedData.pdf