Initiation of Snowball Earth with volcanic sulfur aerosol emissions

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After 1.5 billion years without significant glaciation, during the Cryogenian Period (720-635 Ma), ice extended to the equator at least twice and glacigenic sediments were deposited on every major continent\(^1,2\). Although it is generally agreed that these Snowball Earth events occurred via an ice-albedo runaway\(^3\), the drivers of cooling past the critical bifurcation point are unknown. Geochronological constraints indicate that eruptions associated with the Franklin Large Igneous Province (LIP) coincided with the onset of the first of the Cryogenian Snowball Earth events, the Sturtian glaciation\(^4\). The Franklin LIP was the largest Neoproterozoic volcanic event\(^5\) and was emplaced into a sulfur evaporite basin at the equator\(^4,6\). Geochemical data on sills indicate wall-rock entrainment of sulfur\(^7\), which would have outgassed as SO\(_2\) and H\(_2\)S and formed radiatively active sulfate aerosols. To constrain the magnitude and duration of an eruption needed to initiate a Snowball Earth, we performed coupled volcanic plume, aerosol evolution and climate modeling. We show that in a cool climate with a low tropopause, multi-year sulfur-rich fire-fountains at the equator can inject enough sulfur into the stratosphere to drive a runaway ice-albedo event. Coincidence has also been established between LIPs and mass extinctions\(^8,9\), but these events are variably associated with evidence for warming or cooling, and kill mechanisms are still unclear. Our results demonstrate that high latitude eruptions, such as the Siberian Traps, which is associated with the Permo-Triassic extinction\(^9\), cause less cooling, or even rapid warming if they erupt over high albedo surfaces. Thus, we suggest that depending on pre-existing geological and climate conditions, natural sulfur aerosol emissions on decadal timescales can drive extreme climate change, providing a link between LIPs, environmental destruction, and mass extinctions.

Snowball Earth events represent the largest episodes of planetary climate change recorded in the geological record\(^2\), both on Earth and throughout the Solar System, and yet we do not understand how or why these events occurred. It has been proposed that reduced volcanic CO\(_2\) outgassing\(^10\) or increased planetary weatherability due to the predominance of low-latitude continents mantled with flood basalts led to a cool Neoproterozoic climate\(^4,11-14\), and that Snowball Earth was initiated by additional short-term perturbations to the greenhouse gas inventory\(^11,15\) or planetary albedo\(^16-18\). Models invoking a short-term drawdown of CO\(_2\) or methane were motivated by an apparent relationship between perturbations to the carbon cycle and the onset of glaciation\(^11,15\). However, recent geochronology has demonstrated that the pre-Sturtian Islay carbon isotope excursion occurred more than 10 Myrs before the initiation of the Sturtian glaciation by 716.5 ± 0.2 Ma\(^4,13\), whereas the eruption of the Franklin Large Igneous Province (LIP) was synchronous (Fig. 1).

Volcanic rocks associated with the Franklin LIP cover an area > 3 Mkm\(^2\) over northern Laurentia and southern Siberia (Extended data Fig. 1)\(^5\), and erupted at equatorial latitudes\(^4,6\).
Victoria Island of the Canadian Arctic, the Franklin LIP is represented by basalt and gabbroic dikes and sills that intruded carbonate, organic-rich shale, and sulfur-evaporite of the Shaler Supergroup. Due to melting and assimilation of sulfur from the evaporites, many of the sills and dikes have extremely high sulfur concentrations, ranging from 100 to 100,000 ppm, and sulfur isotope compositions indicative of contamination (Extended data Fig. 2). Importantly, these sulfur-rich sills were dated at 716.3 ± 0.5 Ma, within error of the Sturtian glaciation onset between 717.4 ± 0.2 Ma and 716.5 ± 0.2 Ma (Fig. 1).

To evaluate the hypothesis that the emplacement of the Franklin LIP into a sulfur-rich basin liberated sulfate aerosols and initiated Snowball Earth, we first modeled the height to which sulfur-bearing volcanic plumes could be injected into the atmosphere during the eruption of a LIP, the microphysical evolution of aerosols in the stratosphere, and the radiative effects of a large sulfur-rich eruption at low-latitude. Unlike the short-lived injection of sulfate aerosols to the stratosphere from the 1991 eruption of the Pinatubo stratovolcano, fire fountains associated with LIPs can erupt for several years and drive turbulent, volatile-rich convective plumes into the atmosphere. We simulated the maximum height obtained by convective plumes using a steady-state model incorporating turbulent entrainment (see Extended Data). Plume height is a strong function of volume eruption rate, with values of 10^4-10^5 m^3/s leading to plume heights greater than the present-day tropical tropopause (~12 km) under Neoproterozoic atmospheric conditions (Fig. 2).

Volcanic sulfur is outgassed as a combination of SO_2 and H_2S. Once in the atmosphere, these gases react with O_2, H_2O and OH to form H_2SO_4, which condenses with H_2O onto condensation nuclei to form radiatively active sulfate aerosols. Weak plumes inject sulfur into the troposphere, where sulfate aerosols are consumed in days to weeks and have little long-term radiative effect. Conversely, strong plumes penetrate the tropopause and reach the stratosphere, where H_2SO_4 aerosols can have lifetimes of a year or more. The primary determinant of tropopause height is surface temperature (Fig. 2a); a warmer surface injects more water vapor into the atmosphere, which reduces lapse rate. Hence a given eruption is more likely to penetrate the stratosphere when the background climate is cold. This means that a pre-Snowball climate where CO_2 had already been drawn down to low levels would be more vulnerable to cooling via volcanic stratospheric aerosol injection. It also implies that once a volcanic eruptions begin, a positive feedback can occur: surface cooling by aerosols lowers the tropopause height, making it easier for subsequent eruptions to penetrate the tropopause and recharge the aerosol content of the stratosphere.

To evaluate the radiative forcing following stratospheric SO_2 injection, we modeled sulfur chemistry and aerosol microphysics, incorporating particle growth, coagulation, sedimentation and mixing in a box model of the stratosphere. Radiative forcing is calculated using a 1D radiative-convective correlated-k model. We find that the radiative forcing caused by episodic injections of SO_2 into the equatorial stratosphere increases with the quantity of SO_2 injected (Fig. 3a), but the increase is sub-linear due to particle coagulation, which increases mean particle size and sedimentation rate for large eruptions. For the case of episodic year-long eruptions, mean global forcing increases from ~2.7 W/m^2 given 20 Gt SO_2 injection (c.f. Pinatubo) to ~12 W/m^2 for 500 Mt SO_2 (sensitivity to parameters described in Extended Data). Estimating the rate at which sulfur was released from the Franklin LIP on a decadal timescale is challenging, but 500 Mt/yr is similar to estimates of SO_2 release rate from more recent LIPs.

The ability of volcanic aerosols to force a Snowball transition depends on the starting climate state and on the rate at which the mixed layer of the tropical ocean can cool.
Atmospheric CO₂ levels in the Neoproterozoic are not well constrained but were probably low³ due to the predominance of equatorial continents¹, which enhances weathering²⁸. The amount of metamorphic CO₂ released from the emplacement of gabbroic sills in the Franklin LIP was small relative to background levels and likely did not have a significant warming effect¹⁹. Moreover, relative to CO₂, only small quantities of reflective aerosols are needed to alter Earth’s global radiative balance²⁵. Based on radiative-convective modeling, we estimate a -10 W/m² global mean aerosol radiative forcing would be sufficient to cause runaway glaciation for CO₂ levels of 3000 ppm of below (Fig. 3b). For a representative mixed layer depth of 50 m, the cooling timescale is of order 3 years (see Extended Data). This timescale is compatible with geological observations: individual eruptions in recent mafic LIPs have been estimated to have lasted ~13 years with plume heights >15 km²⁶. Larger flows, such as those associated with the Deccan and Siberian Traps, may have lasted an order of magnitude longer²⁶ and had multiple eruptive centers. Hence a sequence of a few large, sulfur-rich eruptions of the type shown in Fig. 3A would have been sufficient to cause a Snowball transition (Fig. 3b).

If the Franklin LIP caused a Snowball Earth, then why are other LIPs not also associated with Snowball Earth events? The Siberian Traps are similar in size to the Franklin LIP, but instead of cooling, they appear to be associated with warming and extinction²⁸. Another LIP, the Central Atlantic Magmatic Province (CAMP), may have caused a temporary glaciation followed by warming⁶. The different outcomes are likely related to several factors: the background climate conditions, changes in planetary albedo with different paleogeography, the latitude of the eruptions, the composition of the country rock that the LIPs were emplaced into (a sulfate-rich basin, coal deposits, and an ancient mountain belt for the Franklin, Siberian Traps and CAMP, respectively), and timing of SO₂ injection relative to the cumulative release of greenhouse gases. Although Neoproterozoic paleogeography likely favored a cool climate¹,²⁷, the Siberian Traps and CAMP erupted during the ice-free Late Permian and Late Triassic, respectively, when warm conditions extended to Earth’s polar regions²⁹. Consequently, there was a higher ocean thermal inertia and higher mid-latitude tropopause that prevented stratospheric forcing by all but the most powerful volcanic eruptions (Fig. 2). Moreover, the Siberian Traps erupted at high latitude, whereas the CAMP and Franklin LIP erupted at equatorial latitudes⁶. Consequently albedo changes associated with the Siberian Traps would have primarily affected mid-to-high latitudes in one hemisphere, whereas the CAMP and Franklin LIP would have maximized albedo at low latitude, where solar forcing is highest. In general, because aerosol radiative forcing is highest given low surface albedo and a high latitude eruption in a cold climate occurs over snow or sea ice, an equatorial eruption is far more effective at forcing a Snowball transition than a high latitude one (see Extended Data).

The geological record preserves evidence for two Cryogenian glaciations, the Sturtian and the Marinoan, and although there is not evidence for a large LIP during the onset of the Marinoan glaciation, this may be a matter of preservation. The low-latitude rifting of Rodinia continued throughout the Neoproterozoic, and it is likely that these were associated with additional LIPs. Geochronological constraints suggest the Sturtian glaciation lasted ~58 Myrs and that there was a short recurrence interval between the glaciations, between 8.6 and 19.4 Myrs¹³. If planetary weatherability remained high during the Cryogenian due to continued low-latitude paleogeography and recently emplaced continental flood basalt provinces¹⁴, transport-limited weathering in the aftermath of the Sturtian glaciation may have returned the Earth on a 10 Myr timescale to a climate state sensitive to short-term perturbations in the concentration of greenhouse gases and albedo³⁰. After the Marinoan glaciation, the removal of a basaltic carapace
and drift of continents to higher latitudes\textsuperscript{1} likely reduced global weatherability and climate sensitivity.

We argue that the proximal trigger for the Sturtian Snowball Earth was the emission of volcanic aerosols and a sudden increase in planetary albedo. Over the Phanerozoic, the largest disturbances to surface environments coincided with LIPs and giant meteorite impacts. It appears that it was not the size of the LIP or the impactor that mattered most, but when and where it hit\textsuperscript{28}. LIPs and meteorite impacts both hold the potential to rapidly change the environment through the release of metals, volatile gases and reflective particles, but critically it is the rate of change that determines whether species adapt or are exterminated. Although there has been much focus on the cumulative effects of volcanic emissions, we suggest that multi-year to decadal perturbations to albedo and climate from LIPs may instead drive the highest rate of environmental change. Thus, a better understanding of these formative events may require annual resolution of events that occurred hundreds of millions of years ago, which presents a substantial challenge for the geological community, but one that can potentially be met by the richness of the stratigraphic record.


\textsuperscript{8} Schoene, B., Guex, J., Bartolini, A., Schaltegger, U. & Blackburn, T. J. Correlating the end-Triassic mass extinction and flood basalt volcanism at the 100 ka level. \textit{Geology} 38, 387-390 (2010).

\textsuperscript{9} Burgess, S. D. & Bowring, S. A. High-precision geochronology confirms voluminous magmatism before, during, and after Earth’s most severe extinction. \textit{Science advances} 1, e1500470 (2015).


Figure 1. Geochronological constraints on the onset of the Sturtian glaciation compared to the most precise date on sulfur rich sills of the Franklin Large Igneous Province. A) Neoproterozoic timeline with carbon isotope chemostratigraphy, modified from Cox et al.\textsuperscript{14}. B) Geochronological constraints on the onset of the Sturtian glaciation. C) Geochronological constraints on the Franklin large igneous province. Geochronological data and sources are in Extended Data.
Figure 2. Maximum plume height as a function of volume eruption rate under Neoproterozoic insolation and atmospheric conditions. Asterisks indicate tropopause height for each simulation. Black, red, green and blue lines correspond to simulations where the background CO$_2$ levels were 30, 300, 3000 and 30000 ppm, respectively. Grey box brackets maximum plausible volume eruption rate during formation of the Franklin LIP (see Extended Data).
Figure 3. The effect of volcanic sulfate aerosol emission on global climate. A) Radiative effect of yearly equatorial volcanic eruptions under Neoproterozoic conditions. Black, red and green lines show simulations assuming 20, 100 and 500 Mt of SO₂ injection to the stratosphere. Annual global mean forcings for each case are displayed. B) Bifurcation diagram of global mean temperature versus radiative forcing defined as the difference between outgoing longwave radiation and absorbed solar radiation. Dashed lines are unstable states. Starting from a mean surface temperature close to present-day (red star), a negative radiative forcing of < -10 W/m² is sufficient to push the Neoproterozoic Earth (green line) into a Snowball state (blue star). Starting from a colder initial state, the required radiative forcing is lower.
Supplementary Material: Initiation of Neoproterozoic Snowball Earth with volcanic sulfur aerosol emissions

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August 3, 2016

1 Materials and Methods

1.1 Geological Setting

Igneous rocks associated with the Franklin LIP are dominated by mafic dikes and sills that intrude the Canadian Shield and Proterozoic basins on the Arctic margin of Laurentia (Fig. S1), and may be correlative with similar aged intrusions on the southern margin of Siberia (1). The dikes presumably fed large basaltic provinces that have since been largely eroded. Preserved extrusive volcanic rocks include the Natkusiak Formation basalt of Minto Inlier, Victoria Island (2, 3), the Mount Harper Volcanics of the Coal Creek inlier in the Ogilvie Mountains (4, 5), Yukon, the Pleasant Creek Volcanics of the Tatontuk inlier, which straddles the Yukon-Alaska border (6), and the Kikiktak Volcanics of Arctic Alaska (7). These extrusive rocks are composed predominantly of tholeitic basalt to basaltic andesites, typical of continental flood basalts.

The Natkusiak magmatic assemblage consists of dolerite sills that intruded the Shaler Supergroup (8, 9), northwesterly striking dikes, and a succession of plateau basalts (2). Sedimentary strata of the underlying Shaler Supergroup were updomed, beveled and faulted before and during the emplacement of the Natkusiak magmatic assemblage (9, 10). Sills range in thickness from 5 to 100 m; some are differentiated with olivine accumulation at their base and pegmatites in their upper levels (2). Exposures of the Natkusiak Formation basalts have been separated into a northern lobe (3) and a southern lobe (11) of exposures preserved in synclines. The Natkusiak Formation basalts are thickest in the northern lobe, with a maximum preserved thickness of \(\sim 1100\) m, and has been divided into seven members (3). In the southern lobe, the Natkusiak Formation is up to 200 m thick and has been separated into four units that have been correlated with Jefferson et al.’s (1985) ‘basal member’, ‘pyroclastic member’, and ‘lower massive member in the northern lobe (11). Thus, it appears that units above the ‘Lower Massive Member’ in the northern lobe are not preserved in the southern lobe.

Nabelek et al. (12) estimated the amount of metamorphic CO\(_2\) released from the emplacement of the gabbroic sills associated with the Franklin LIP into carbonate strata of the Shaler Supergroup. Along with carbonate, the Shaler Supergroup includes organic rich shale and evaporite deposits at multiple horizons that are several tens of meters thick. Evans et al. (13) estimated a volume of 30,000 km\(^3\) in the upper evaporites of the Kilian-Redstone River deposits and 90,000 km\(^3\) in the lower evaporites of the Minto Inlet Formation. Although the total amount and rate of S release during the emplacement of sills in the Shaler Supergroup is poorly constrained, disseminated copper sulfides are present within basalt flows, outlining clasts in the pyroclastic member, and as veins
in all of the members (3). Additionally, many of the sills and dikes have extremely high sulfur concentrations (14, 15). Although dikes have been observed that feed the sills, feeder dikes for the basalts have not been observed. Yet, it is assumed that at least some of the dikes fed the basalts as is suggested by their similar chemistry (2, 16). Some of the sills are folded around a volcanic center near Kilian Lake that has been correlated with the pyroclastic member (14), suggesting that at least some of the sills formed before or very early in the eruption sequence. However, other sills are vesiculated and feed flows higher in the basaltic stack. Although fissure eruption centers have not been identified associated with the Franklin, the exposure is limited and it does not mean they were not there, and several circular to keyhole shaped eruption centers have been mapped [the largest one identified is $\sim 3 \times 2 \text{ km} (14)]$.

Paleomagnetic data from the Franklin LIP provide the most robust Neoproterozoic pole for Laurentia and is an important tie point for Neoproterozoic paleogeography (17). Data spanning six key regions, the Canadian Mainland, Victoria Island, Baffin Island, Devon Island, Ellesmere Island and northern Greenland, yield a grand mean pole that restores Laurentia and the Natkusiak assemblage of the Franklin LIP at ca. 717 Ma to within 9 degrees of the equator (17, 18) (Fig. S1).

U-Pb zircon and baddeleyite ages from the Franklin LIP rocks span ca. 725-710 Ma (Table S1). This spread could represent distinct pulses of magmatism; however, for younger, better dated LIPs, most of the volume is erupted in $<< 1 \text{ Myr}$ (19). Instead, this large spread may also be due to imprecise multigrain techniques and upper intercept ages, and we suggest that the most precise dates represent the best age constraint on the Franklin LIP. The most precise, single grain chemical abrasion-isotope dilution-thermal ion mass spectrometry (CA-ID-TIMS) on zircon and baddeleyite associated with the Franklin LIP yield ages between 717 and 716 Ma (Table S1). It has been also proposed that the ca. 725 Ma Dovyren Intrusive Complex in the Lake Baikal region of Russia represents a component of the Franklin LIP in SW Siberia (1). If true it would provide evidence that Franklin magmatism initiated well before the onset of the Sturtian glaciation and covered an area beyond that defined by the radiating dyke swarm in north-central to northeastern Laurentia. However, the Siberian age is imprecise and hence this correlation remains speculative.

A quartz-rich rhyolite flow in member D of the Mount Harper Volcanics was dated at 717.4 ± 0.2 Ma and a tuff interfinger with glacial deposits of the overlying Rapitan Group was dated at 716.5 ± 0.2 Ma, both with U-Pb CA-ID TIMS on zircon (20). These dates have been interpreted to bracket the onset of the Sturtian glaciation (20). Additional recent ages on the Franklin LIP and the onset of the Sturtian glaciation confirm this age model (Table S1). In China, an ash bed within stratified diamictite of the lowermost Chang-an Formation was dated with secondary ion mass spectrometry (SIMS) on zircon at 715.9 ± 2.8 Ma (21). In Arctic Alaska, a 719.5 ± 0.3 Ma CA-ID-TIMS age on zircon from volcaniclastic strata at the top of the Kikiktak Volcanics and directly below the Hula Hula diamictite is consistent with both a Sturtian-age of the Hula Hula diamictite and a tight temporal relationship between the Franklin LIP and the onset of the Sturtian glaciation (7). Sills that intrude evaporite rich-strata of the upper Shaler Supergroup dated at 716.3 ± 0.5 Ma (ID-TIMS on baddeleyite) (20) are indistinguishable from the constraints on the onset of the Sturtian glaciation (Fig. 1 in the main text). Importantly, the dated sill, sill 6a near Kilim Lake, is one of most sulfur-rich sills analysed to date (3).

1.2 Geochemistry

Subaerial continental flood basalt (CFB) volcanism is characterized by the repeated effusion of $\sim 10^3 \text{ km}^3$ of basaltic magma in less than 1 Myr. Each eruption comes from a dyke-fed fissure and
can last up to a decade or more (19). During eruptions, gas fluxes of > 1 Gt per year of SO\(_2\) and CO\(_2\) are possible (22). These volumes can increase when magmatic systems invade volatile-rich sediments as more volatiles are entrained into the melt. The occurrence of spatter, spatter-fed lava, and scoria mounds along eruptive fissures in the Columbia River CFB and in Icelandic fissure eruptions suggests the occurrence of violent fire-foaming events (23). It has been estimated that these fire fountains exceeded 1.5 km in height with convective plumes rising to heights in excess of 13 km (22).

Estimates of gas-release budgets with melt inclusion studies are limited to individual CFB eruptions in the mid-Miocene Columbia River CFB (24), the Deccan Traps (22), Siberian Traps (25), and studies of Icelandic fissure eruptions (26). These studies suggest that ~75% of volatile S species present in the rising melt are released at the vents largely as SO\(_2\) (22). The months-long Laki eruption in Iceland is estimated to have released ~120 Mt of SO\(_2\) with maximum fluxes reaching 6 Mt per day (26). Estimation of S gas release from ancient eruptions is complicated by weathering and ground water alteration, which leads to the removal of volatile elements. Nonetheless, using melt inclusions it has been estimated that a single flow with in the Columbia CFB released ~12 Gt of SO\(_2\) (24). Melt inclusion studies are beyond the scope of this study and it is not clear that there is material preserved in the Franklin LIP that is pristine enough for such a study.

In crystal-poor or altered CFBs, original S content of the lava can be estimated with the S content of undegassed dikes and sills normalized to TiO\(_2\)/FeO\(_2\) (27). S saturated melts from LIPs typically have between 1000 and 2500 ppm of S prior to degassing and flows contain much lower concentrations between 200 and 300 ppm (22). Major, minor, and trace element concentrations including S concentrations and S isotopes from sills and dikes associated with the Franklin LIP were compiled by Bedard et al. (15). S concentrations in sills range from 100-100,000 ppm and S isotopes show increasing variability with increasing concentration indicative of wall-rock assimilation (Fig. S2). At these high S concentrations, and with the late addition of S out of equilibrium with the melt, normalizations of S concentrations to TiO\(_2\)/FeO\(_2\) is both insignificant and not appropriate. If we assume ~75% of S remains in melt (22), and that the sills were variably degassed from original S concentrations less than 2500 ppm, the upper end of this range is greater than that of basaltic flows in the Columbia River CFB (24), the Deccan Traps (22), Siberian Traps (25), and the Icelandic fissure eruptions (26).

Crustal S is incorporated into mafic magmas predominantly via direct melting and assimilation of wall rock and xenoliths rather than by the liberation of sulfur during contact metamorphism in the thermal aureole (28). Although previous work has questioned the viability of releasing sulfur via intrusions into evaporites due to the high melting temperature of pure anhydrite (~1450°C), the melting temperature of mixtures of anhydrite and dolomite, which is probably closer to the composition of the wall rock intruded by the Natkusiaq sills, is much lower (~1000°C) (28). Moreover, hydrothermal systems associated with the Franklin LIP may have partially dissolved the evaporites, and if these fluids were saline, the solubility of anhydrite greatly increases with NaCl activity at temperatures between 600\(^\circ\)C and 800\(^\circ\)C (29). We suggest that excess S was incorporated into magmas primarily through the melting and dissolution of evaporite layers in the Minto Inlier and Killian formations of the Shaler Supergroup when intruded by sills, and then was rapidly degassed as bubbles in shallow intrusions, fissure eruptions, and in flows that were fed by these sills.

Petrographic analysis has revealed abundant disseminated pyrite and immiscible metallic sulfur in the sills. Bulk S isotope studies preserve a wide range of compositions (Fig. S2), differing from the value of mantle derived LIPs (~ 0‰ VCDT). Although Jefferson et al. (14) argued that trace
element compositions suggest that the sulfur was sourced from sulfidic shale of the Wynniatt Fm, the high S concentrations and the enriched S isotopic values of several of the sills are consistent with the S also coming from the entrainment of carbonate and sulfur evaporates into the sills (Fig. S2). However, because we are concerned with decadal fluxes of SO$_2$, the source and the total amount of S entrained and released in magmatic system are not critical. What is important is that the sills that are coincident with the onset of the Sturtian glaciation contain considerably more S than typical CFBs.

1.3 Radiative-convective climate model

To estimate the climate effect of Franklin LIP eruptions, we combine radiative-convective, aerosol microphysics and volcanic plume modeling tools. In the following sections, each of these components is described in detail.

To calculate Neoproterozoic tropopause height, we used a 1D radiative-convective model. The model atmospheric temperature structure is calculated as a function of surface temperature assuming adjustment to a moist adiabat in the troposphere, following the methodology in (30). The tropopause height is then defined using the standard Earth-based empirical measure as where the magnitude of the vertical temperature gradient drops below 2 K/km ($dT/dz < -2$ K/km). To relate CO$_2$ molar concentration to surface temperature, we performed iterative 1D radiative-convective calculations with moist adjustment in the troposphere and an assumed relative humidity of 0.77 (31). In the stratosphere, the temperature profile is allowed to evolve freely until thermal equilibrium is reached.

Atmospheric radiative transfer is calculated using a correlated-k approach (32). The absorption effects of CO$_2$ and H$_2$O (infrared and visible) and O$_3$ (UV) are taken into account; absorption by other gases is set to zero. Rayleigh scattering in the shortwave by N$_2$ and O$_2$ is taken into account, but the radiative effects of clouds are neglected. The surface albedo is set to 0.24, a value that allows the model to reproduce the present-day mean surface temperature (288 K) in simulations with present-day solar flux, surface pressure and atmospheric composition. Such an elevated surface albedo implicitly accounts for the effect of low-lying clouds (a standard assumption in 1D radiative-convective modelling). The radiative forcing due to clouds is uncertain in Earth’s paleoclimate (33,34), so inclusion of a fixed cloud forcing here would not have led to greater insight.

For the CO$_2$ and H$_2$O line absorption, we computed high resolution spectra over a range of temperatures, pressures and gas mixing ratios. We used an $6 \times 8 \times 8$ temperature, pressure and H$_2$O molar concentration grid with values $T = \{100, 150, \ldots , 350\}$ K, $p = \{10^{-3}, 10^{-2}, \ldots , 10^{3}\}$ mbar and $f_{H_2O} = \{10^{-8}, 10^{-7}, \ldots , 10^{-1}\}$, respectively. The correlated-k method was used to produce fast coefficients for the radiative-convective simulations, with the CO$_2$ mixing ratio chosen in advance for each calculation. The H$_2$O continuum was implemented using the MT-CKD parametrization (35,36). We used 80 spectral bands in the longwave and 82 in the shortwave, and sixteen points for the $g$-space integration, where $g$ is the cumulated distribution function of the absorption data for each band. Twelve new bands were added to the shortwave compared to previous studies (37) to give sufficient resolution in the ultraviolet to model absorption by ozone accurately.

Ultraviolet O$_3$ absorption cross-sections are derived from the MPI-Mainz UV/VIS Spectral Atlas (38) while the solar spectrum is taken from the VPL database (39). Calculations were performed with two O$_3$ molar concentration profiles: one assumed present-day values, while the other assumed an O$_3$ profile appropriate to an atmosphere with oxygen levels $1/10^{th}$ of today.
1.4 Aerosol radiative transfer

We use Mie theory to calculate aerosol extinction efficiency, single scattering albedo and asymmetry parameter as a function of wavelength for a given particle size distribution output by the microphysical model. The aerosol radiative forcing is calculated using the two-stream approximation, with Eddington scaling used in the shortwave and hemi-isotropic emission assumed in the longwave (40). We assume that absorption dominates in the longwave and that scattering dominates in the shortwave. Delta scaling is applied to the shortwave optical coefficients to ensure that the forward peak of the phase function is correctly captured at large particle size parameters. We describe the phase function using the Henyey-Greenstein approximation (41). For the aerosol particle refractive indices, we use the data given in (42), which were measured at stratospheric temperatures (\(\sim 200\) K). Solar zenith angle \(\theta_z\) was calculated by diurnal and seasonal averaging across the microphysics box domain, with weighting by the instantaneous insolation (43). For the equatorial eruption simulations (see next section), this yielded \(\theta_z = 43.8^\circ\), while for the midlatitude case the value was \(\theta_z = 55.4^\circ\). Finally, for the equatorial eruption simulations shown in Fig. 3A in the main text, we used a pre-aerosol tropical planetary albedo of 0.24. This was obtained by averaging albedo data from the Earth Radiation Budget Experiment (ERBE) mission (36,44) over the simulation domain. The same procedure was used to obtain the albedo value of 0.365 for the second mid-latitude eruption plot in Fig. S11.

1.5 Volcanic plume model

The volcanic plume model simultaneously solves the equations of mass, momentum and energy transport for an axisymmetric, steady-state circular plume that entrains the surrounding air as it rises. These equations are defined as

\[
\begin{align*}
\frac{df_1}{dz} &= \frac{d}{dz}(\rho_g ur^2) = 2\alpha \rho_a ur \\
\frac{df_2}{dz} &= \frac{d}{dz}(f_1 u) = g(\rho_a - \rho_g)r^2 \\
\frac{df_3}{dz} &= \frac{d}{dz}(f_1 c_{p,g} T_g) = 2\alpha \rho_a ur c_{p,a} T_a - \rho_a ur^2 g.
\end{align*}
\]

with

\[
\begin{align*}
f_1 &= \rho_g ur^2 \\
f_2 &= f_1 u \\
f_3 &= f_1 c_{p,g} T_g.
\end{align*}
\]

Here \(\alpha\) is an entrainment coefficient, \(z\) is altitude, \(u\) is the vertical speed of the plume, \(r\) is the plume radius, \(\rho_a\) is the density of ambient air, \(\rho_g\) is the mean density of gas in the plume, \(g\) is gravity, \(T_a\) is the local atmospheric temperature, \(T_g\) is the plume gas temperature and \(c_{p,a}\) and \(c_{p,g}\) are the specific heat capacity at constant pressure of ambient air and of the plume gas, respectively. Following (45), the plume specific heat capacity and gas constant are modified as it moves upwards to account for the entrainment of ambient air.

After boundary conditions at the surface are chosen (see below), (1-3) are integrated upwards until the vertical speed of the plume equals zero. The height at which this occurs is then defined as the maximum plume height. This plume model is based on the methodology of (45). It is a
generalization of the classic incompressible model of Morton et al. (46), which has been validated via numerous laboratory experiments and comparison with observations of real volcanic eruptions (47). For the numerical integration upwards we use a simple forward Euler approach, with vertical resolution increased to the point where the results converge (we use 1000 levels based on trial and error).

To initialize our model, we use the same approach as in a previous study that successfully predicted the plume heights of the historic Laki, Mauna Loa and Askja eruptions (47). Specifically, we link $Q$ to the volume eruption rate $V_{\text{erupt}} \, [\text{m}^3/\text{s}]$ via the expression

$$Q = \rho_{\text{magma}}V_{\text{erupt}}[(1 - q_f - q_v)c_{p,magma}\Delta T_c + (q_f c_{p,magma} + q_v c_{p,v})\Delta T_g].$$

Equation (8) assumes that the plume is heated by a combination of volatiles, fines and clasts, with a negligible contribution from fire fountain lava blobs. Here $\rho_{\text{magma}}$ is the magma density, which we take to be 2900 kg/m$^3$ — a value appropriate to basaltic composition, $q_f$ is the specific concentration of fines [kg/kg] and $q_v$ is the specific concentration of volatiles. $c_{p,magma}$ and $c_{p,v}$ are the specific heat of magma and volatiles (the latter is assumed to be dominated by water vapor), respectively. $\Delta T_c$ is the temperature drop of clasts in the fire fountain while $\Delta T_g$ is the temperature drop of volatiles and fines in the plume. See Table S2 for definitions and values of all model parameters.

We assume conservative values for the key unknown parameters: $q_f = 0.01$ kg/kg, $q_v = 0.02$ kg/kg, $\Delta T_c = 50$ K and $\Delta T_g = T_{g,0} - T_{g,1} = 1400 - 220 = 1180$ K (here $T_{g,1}$ is the assumed final plume temperature). We assume the dominant volatile by mass is water vapor. The contribution of sulfur and other comparatively minor species to the heat budget is neglected. The initial mass flux is calculated from the heat flux assuming a circular vent as (45)

$$f_{1,0} = \frac{Q}{\pi c_{p,v}(T_{g,0} - T_{a,0})}.$$

where 0 denotes initial (surface) variables. Fissure systems tend to produce lines of circular cones to key-hole shaped vents that range from tens of meters to kilometers in diameter, but more elongated eruptive centers also occur. Glaze et al. (2015) (48) found that elongated geometries produce larger plumes, but for simplicity we use a conservative circular geometry here.

1.6 Aerosol microphysical model

We model key aspects of the sulfate aerosol microphysics, focusing on those processes that play the most important role in determining total radiative forcing. Our model describes the evolution of $\text{SO}_2$ and $\text{H}_2\text{SO}_4$ gas, Aitken particles (aerosol condensation nuclei), and the aerosols themselves. Aerosol particles undergo nucleation onto Aitken particles, growth, sedimentation and coagulation.

In the microphysical model, the stratosphere is treated as a single box that exchanges material with the troposphere. The advantage of this idealized approach is that it captures the key processes governing aerosol optical depth and mean radius while allowing rapid exploration of sensitivity to poorly constrained parameters. Given the uncertainties, this approach is a more robust one than using a complex off-the-shelf model tuned to present-day Earth conditions.

Once they reach the tropical stratosphere, sub-tropical transport barriers cause aerosols lofted there to remain mostly confined to that region (49, 50). Orbital observations of stratospheric aerosol loading after Pinatubo showed that aerosol optical depth at 0.5 $\mu$m remained above 1/2
the peak value for around 45 weeks, with a strong peak in forcing in the 30°S-30°N latitude region \((51, 52)\). Modeling the stratospheric meridional circulation in the Neoproterozoic is challenging due to uncertainties in topographic wave forcing and stratospheric wave-mean interaction, but the aerosol transport following eruptions of the Franklin LIP should have been broadly similar.

In this study we treat the tropical and midlatitude stratosphere as distinct domains. For the tropical eruption simulations shown in the main text, we defined the domain base as 16 km and the domain volume as \(V = 2\pi r_E s \Delta z\), with \(r_E\) Earth’s radius, \(s = 1.4 r_E\), corresponding to a latitudinal extent from 30° S to 30° N, and \(\Delta z = 15\) km the height of the simulation domain. Our microphysical model is sectional, with 82 particle bins ranging from 0.01 \(\mu\)m to 4.14 \(\mu\)m radius. Bin spacing is defined geometrically, with the particle volume increasing by a factor of 1.25 each time the bin number increases. Once the microphysical model produces aerosol mass loading and size distribution data as a function of time, its results are fed into the radiative transfer model to yield net radiative forcing in the domain. Global net radiative forcing is then calculated by multiplying the results by a factor \(2\pi r_E s / 4\pi r_E^2\).

The model prognostic equations take the general form

\[
\frac{\partial \tilde{n}_k(t)}{\partial t} = P_k - L_k - \left( \frac{\partial \tilde{n}_k}{\partial t} \right)_{growth} - \frac{\tilde{n}_k}{\tau_{exchange}} \quad (9)
\]

\[
\frac{\partial n_i(r_i, t)}{\partial t} = \left( \frac{\partial n_i}{\partial t} \right)_{growth} + \left( \frac{\partial n_i}{\partial t} \right)_{coag} - \frac{n_i}{\tau_{sedim}} - \frac{n_i}{\tau_{exchange}} \quad (10)
\]

where \(\tilde{n}_k\) and \(n_i\) are molar concentrations of gas and aerosol species, respectively, and \(r_i\) is the aerosol particle size in bin \(i\). \(P_k\) and \(L_k\) are chemical production and loss terms, while the time derivative terms describe changes due to particle growth and coagulation. \(\tau_{exchange}\) and \(\tau_{sedim}\) are timescales for exchange of material with the troposphere and adjacent stratospheric regions, and for aerosol sedimentation. The equations are solved using the Klopfenstein-Shampine algorithm (\textit{ode15s} in Matlab).

The transformation of \(\text{SO}_2\) into \(\text{H}_2\text{SO}_4\) in the stratosphere proceeds by a chain of photochemical reactions, the most important of which are \((53)\)

\[
\text{SO}_2 + \text{OH} + \text{M} \rightarrow \text{HSO}_3 + \text{M} \quad (11)
\]

\[
\text{HSO}_3 + \text{OH} \rightarrow \text{SO}_3 + \text{H}_2\text{O} \quad (12)
\]

\[
\text{SO}_3 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_4 \quad (13)
\]

In our model, the chemical production and loss terms \(P_i\) and \(L_i\) are defined by the key sulfur reactions shown in Table S3. We neglect the outgassing of \(\text{H}_2\text{S}\) in our model. As we find that the \(\text{SO}_2\) injection rates to the stratosphere required to force a Snowball transition are orders of magnitude lower than the upper limits on Franklin \(\text{SO}_2\) outgassing rates, inclusion of \(\text{H}_2\text{S}\) would have added complexity to the model without fundamentally changing the nature of the conclusions.

The model is initialized by the injection of a given mass of \(\text{SO}_2\) gas \(m_{\text{SO}_2}\) into the box. Injected masses are calculated as multiples of the \(\text{SO}_2\) injected by Pinatubo [approx. \(m_{\text{SO}_2} = 20\) Mt; \((19)\)]. The number density of \(\text{H}_2\text{O}\) and the background gas density [M] are calculated from a standard present-day atmospheric profile \((54)\) (Table S2), while the OH number density is set to yield the characteristic \(\text{SO}_2\) to \(\text{H}_2\text{SO}_4\) conversion timescale observed after Pinatubo of 35 days \((55)\). The sensitivity of the results to changes in OH levels are described in the next section. At the first
model timestep, the aerosol distribution is set to consist of condensation nuclei only. We assume that condensation nuclei are transported into the stratosphere along with the sulfur-bearing gas. The number concentration in the first particle bin is initialized as \( n^0_1 = n_{Cn}(m_{SO_2}/m_{SO_2}, 0) \), where \( m_{SO_2}, 0 = 20 \text{ Mt} \). We find that \( n_{Cn} = 100 \text{ particles cm}^{-3} \) yields closest correspondence with Pinatubo observations. This value is similar to the observed condensation nuclei number density at the tropopause on present-day Earth \((56)\). Sensitivity of the results to variations in this parameter are also described in the next section.

**Aerosol particle nucleation and growth**

Aerosol particles grow through absorption of both \( \text{H}_2\text{SO}_4 \) and \( \text{H}_2\text{O} \). Equilibration with \( \text{H}_2\text{O} \) is rapid, however, and the variation of equilibrium \( \text{H}_2\text{O}:\text{H}_2\text{SO}_4 \) droplet weight ratios with height in the atmosphere is not high \((53)\). For simplicity, here we assume that droplets have a fixed \( \text{H}_2\text{SO}_4 \) specific concentration \( q_{\text{H}_2\text{SO}_4} = 0.75 \text{ kg/kg} \), a value appropriate to Earth’s present-day lower stratosphere \((53)\).

Aerosol particles are assumed to form on Aitken particles, which we treat as a monodisperse population of small \( r_p = 0.01 \mu\text{m} \) particles. The growth of particles by condensation of \( \text{H}_2\text{SO}_4 \) is given by

\[
g = \frac{dr_p}{dt} = \frac{m_{\text{H}_2\text{SO}_4}D_{\text{H}_2\text{SO}_4}(n_{\text{H}_2\text{SO}_4} - n_{\text{sat},\text{H}_2\text{SO}_4})}{r_p\rho_p q_{\text{H}_2\text{SO}_4}(1 + \lambda_p\text{Kn}_1)}
\]

where \( r_p \) is the particle radius, \( \rho_d \) is the particle density, \( m_{\text{H}_2\text{SO}_4} \) is the molecular mass of \( \text{H}_2\text{SO}_4 \), \( D_{\text{H}_2\text{SO}_4} \) is the molecular diffusion coefficient of \( \text{H}_2\text{SO}_4 \) in air \((57)\), \( n_{\text{H}_2\text{SO}_4} \) is the number density of \( \text{H}_2\text{SO}_4 \) molecules near the droplet and \( n_{\text{sat},\text{H}_2\text{SO}_4} \) is the \( \text{H}_2\text{SO}_4 \) condensation number density \((53)\). The latter is derived from the \( \text{H}_2\text{SO}_4 \) vapor pressure curve, for which we use the coefficients given in \((58)\). Growth rate was then converted to the rate of change of aerosol number concentration by a derivative with respect to particle radius

\[
\left. \frac{\partial n_i}{\partial t} \right|_{\text{growth}} = \frac{\partial}{\partial r} (g n_i).
\]

\( \text{Kn}_1 = l_{\text{H}_2\text{SO}_4}/r_p \) is the Knudsen number for \( \text{H}_2\text{SO}_4 \) diffusion in air, while we define \( \lambda_p \) here as

\[
\lambda_p = \frac{1.33 + 0.71/\text{Kn}_1}{1 + 1/\text{Kn}_1}.
\]

The \( \text{H}_2\text{SO}_4 \) diffusion coefficient \( D_{\text{H}_2\text{SO}_4} \) and mean free path in air \( l_{\text{H}_2\text{SO}_4} \) were calculated using standard methods \((57)\), with Sutherland’s formula

\[
\eta_a = 1.8325 \times 10^{-5} \left( \frac{416.16}{T_{\text{strat}} + 120} \right) \left( \frac{T_{\text{strat}}}{296.15} \right)^{1.5}
\]

used to calculate the dynamic viscosity of air as a function of temperature.

**Aerosol particle sedimentation**

Sedimentation is modeled using the Stokes velocity equation

\[
v_{\text{sedim}} = \frac{2}{9} f_C r^2 g \rho_p / \eta_a,
\]
where $g$ is gravity and $\rho_a$ is air density. Here

$$f_C = 1 + \text{Kn}_2(A + B e^{-C/\text{Kn}_2})$$  \hspace{1cm} (19)$$
is the Stokes-Cunningham low pressure correction factor, with $A = 1.249$, $B = 0.42$, $C = 0.87$, $\text{Kn}_2 = l_{\text{air}}/r_p$ the Knudsen number for air diffusion and $l_{\text{air}}$ the mean free path of air (57). The sedimentation timescale is then calculated as $\tau_{\text{sedim}} = z_{\text{fall}}/v_{\text{sedim}}$, where $z_{\text{fall}} = 5.9$ km is the distance that an average particle needs to fall to exit the stratosphere.

**Aerosol particle coagulation**

When they collide, aerosol particles may coagulate. Over time this process shifts the particle distribution towards larger radii, increasing the sedimentation rate and decreasing the radiative forcing per unit mass. We describe coagulation using the discrete equation

$$\left. \frac{\partial n_i}{\partial t} \right|_{\text{coag}} = + \frac{1}{2} \sum_{j=1}^{i-1} K_{j,i-j} n_j n_{i-j} - n_i \sum_{j=1}^{i} K_{i,j} n_j$$  \hspace{1cm} (20)$$

where $n_i$ is the aerosol number concentration in the $i$th bin, $t$ is time and $K_{i,j}$ is the coagulation kernel. Total particle volume conservation is ensured by use of a bin-dependent volume fraction correction term (59). We validated our coagulation scheme by comparison with analytical results for the constant $K_{i,j} = K_0$ case [Smoluchowski’s result; see e.g. (60)].

The coagulation kernel is calculated using the Fuchs equation, which is valid both in the high and low particle Knudsen number regimes. Specifically, for two particles of diameter $d_{p,1}$ and $d_{p,2}$, we define

$$K_{12} = 2\pi(D_1 + D_2)(d_{p,1} + d_{p,2}) \beta$$  \hspace{1cm} (21)$$

with

$$\beta = \left[ \frac{d_{p,1} + d_{p,2}}{d_{p,1} + d_{p,2} + 2\sqrt{g_1^2 + g_2^2}} + \frac{8(D_1 + D_2)}{\sqrt{(c_1^2 + c_2^2)(d_{p,1} + d_{p,2})}} \right]^{-1},$$  \hspace{1cm} (22)$$

$$g_i = \frac{1}{3d_{p,i}l_i} \left[ (d_{p,i} + l_i)^3 - (d_{p,i}^2 + l_i^2)^{3/2} \right] - d_{p,i},$$  \hspace{1cm} (23)$$

mean particle speed

$$c_i = \sqrt{\frac{8k_B T}{\pi m_i}},$$  \hspace{1cm} (24)$$

mean free path

$$l_i = \frac{8D_i}{\pi c_i},$$  \hspace{1cm} (25)$$

and particle diffusivity

$$D_i = \frac{k_B T f_C}{3\pi \eta_a d_{p,i}}$$  \hspace{1cm} (26)$$

with $m_i$ the particle mass and $k_B$ Boltzmann’s constant (60).
1.7 Bifurcation Plot

Figure 3B in the main text was produced using a classical Budkyo-Sellers type model of the ice-albedo feedback. We assume that surface albedo depends on temperature as

\[
A(T) = \begin{cases} 
A_i & T \leq T_i \\
A_0 + (A_i - A_0) \frac{(T-T_0)^2}{(T_i-T_0)^2} & T_i < T < T_0 \\
A_0 & T \geq T_0 
\end{cases}
\]

with \(T_0\) the temperature at which ice entirely disappears from the surface, \(T_i\) the temperature at which ice coverage is global, \(A_i\) the albedo of snow/ice and \(A_0\) the albedo of the ice-free planet. Justification of the terms in this equation are given in standard climate textbooks (36). Here we take representative values \(T_i = 260\) K, \(T_0 = 290\) K and \(A_i = 0.6\). \(A_0\) is set to be 0.24, as in the ice-free tropopause height simulations. The radiative-convective model was then run assuming a 1 bar \(N_2\) atmosphere and various \(CO_2\) molar concentrations to produce absorbed solar radiation and outgoing longwave radiation for a range of surface temperatures \(T_s\). The radiative imbalance \(\Delta F = OLR - ASR\) was then produced as a function of \(T_s\) via interpolation.

In some coupled 3D general circulation models with ocean dynamics included, Neoproterozoic Snowball glaciations occur close to lower \(CO_2\) partial pressures than those modeled here (61–63). While we do not attempt to capture the effects of sea ice and ocean dynamics here, we note that the basic effects of aerosol forcing would play out similarly starting from a lower \(CO_2\) climate state. As discussed in the main text, \(CO_2\) levels, while not well constrained prior to glacial onset, were probably low due to the predominance of low-latitude continents (64) and enhanced global weatherability (65).

1.8 Ocean Thermal Inertia

The energy balance equation for the tropical ocean mixed layer can be approximated as

\[
\frac{d}{dt}(\rho_w c_{p,w} T d) = ASR - OLR = \Delta F
\]

with \(\rho_w = 1000\) kg/m\(^3\) the density of seawater, \(c_{p,w} = 4200\) J/kg/K the heat capacity of seawater, \(T\) temperature and \(d\) the depth of the mixed layer (36). For a radiation imbalance \(\Delta F\) caused by aerosol forcing we can write the characteristic cooling timescale as

\[
\tau_{cool} = \frac{\rho_w c_{p,w} d \Delta T}{\Delta F}
\]

Given \(\Delta F = 10\) W/m\(^2\), a required global mean \(\Delta T\) of 5 K and a mixed layer depth of 50 m,

\[
\tau_{cool} = \frac{\rho_w c_{p,w} d \Delta T}{\Delta F} = 3.3\ y.
\]

While certainly missing many additional effects that could be resolved in a 3D coupled general circulation model, this simple calculation demonstrates that the radiative forcing produced by \(SO_2\) emissions from volcanism over 3 years or more is capable of tipping Earth into a Snowball state.
2 Model Validation and Sensitivity Tests

2.1 Radiative-convective climate model

To validate our radiative-convective model, we ran it assuming present-day globally averaged insolation and surface pressure, clear-sky conditions and a solar zenith angle of 60°. Results are shown in Figure S3. As can be seen, the model reproduces Earth’s present-day mean temperature profile reasonably accurately. Figure S4 shows the temperature profiles with varying atmospheric CO₂ used to create Figure 2 in the main text.

2.2 Volcanic plume model

We compared our plume model with analytic and numerical results from the literature (45–47). Figure S5 shows the results of a single simulation with moderate values of heat flux and initial plume velocity. This figure reproduces Fig. 2 in (45), validating our model. Figures S6-S7 show the results of the combined plume / tropopause height modelling for different atmospheric ozone and nitrogen abundances. These figures demonstrate that our conclusions are not dependent on uncertainties in these parameters.

2.3 Aerosol Microphysics

In addition to testing subcomponents of the model vs. analytic results (see previous section), we validated the aerosol microphysical model by comparison with observations of the Pinatubo eruption event. Given a single injection of 20 Mt SO₂ into the tropical stratosphere, our model predicts a global mean radiative forcing of -2.0 W/m² over two years, which falls in the -1 to -3 W/m² range observed in the two years after Pinatubo (51). Our predicted peak optical depth at 0.5 µm wavelength is 0.2 and our peak effective particle radius is around 0.5 µm, both of which are also close to the observed post-Pinatubo values (66). Finally, our peak aerosol sulfur mass burden is 8 Mt of S, which is close to the values obtained by complex 3D models with coupled microphysics and chemistry (Figure S8) (67). To test the response of the microphysical model to uncertain parameters, we also performed a range of sensitivity studies. See Table S4 for details.

Figure S9 shows the effective particle radius (as defined in (68)) and stratospheric aerosol S burden as a function of time for the three simulations displayed in Figure 3A in the main text. Figure S11 shows the radiative forcing from a similar set of simulations run in a domain centered on the mid-latitude northern hemisphere, assuming an ice-covered surface. Radiative forcing is significantly less negative in this case due to the increased surface albedo, which decreases the aerosol cooling effect. Aerosol transport rates from mid-latitudes to the tropics in the stratosphere are slow, so in contrast to the Franklin LIP, a high-latitude eruption would be unlikely to force a Snowball transition. Finally, we also tested the dependence of our radiative forcing predictions on the pre-aerosol planetary albedo. Results are shown in Figs. S10 and S11.
References


Table S1: Geochronological constraints on the onset of the Sturtian glaciation and the Franklin Large Igneous Province (69–83).
Table S2: Standard parameters used in the volcanic plume, climate and aerosol microphysics modeling.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plume entrainment coefficient $\alpha$</td>
<td>0.1</td>
</tr>
<tr>
<td>Specific heat capacity of atmosphere at constant pressure $c_{p,a}$</td>
<td>1040 J/K/kg</td>
</tr>
<tr>
<td>Specific heat capacity of volatiles in plume $c_{p,v}$</td>
<td>1900 J/K/kg</td>
</tr>
<tr>
<td>Specific concentration of fines in eruption $q_f$</td>
<td>0.01 kg/kg</td>
</tr>
<tr>
<td>Specific concentration of volatiles in eruption $q_v$</td>
<td>0.002 kg/kg</td>
</tr>
<tr>
<td>Magma density $\rho_{magma}$</td>
<td>2900 kg/m$^3$</td>
</tr>
<tr>
<td>Magma specific heat capacity $c_{magma}$</td>
<td>1100 J/K/kg</td>
</tr>
<tr>
<td>Solar flux $F_V$</td>
<td>1284 W m$^{-2}$</td>
</tr>
<tr>
<td>Surface pressure $p_s$</td>
<td>1.0 bar</td>
</tr>
<tr>
<td>Ice-free global mean surface albedo $A_0$</td>
<td>0.24</td>
</tr>
<tr>
<td>Ice-covered global mean surface albedo $A_i$</td>
<td>0.6</td>
</tr>
<tr>
<td>Total ice loss temperature $T_0$</td>
<td>290 K</td>
</tr>
<tr>
<td>Global ice coverage temperature $T_i$</td>
<td>260 K</td>
</tr>
<tr>
<td>Aerosol droplet $\text{H}_2\text{SO}<em>4$ specific concentration $q</em>{\text{H}_2\text{SO}_4}$</td>
<td>0.75 kg/kg</td>
</tr>
<tr>
<td>Sulfuric acid density $\rho_{\text{H}_2\text{SO}_4}$</td>
<td>1840 kg/m$^3$</td>
</tr>
<tr>
<td>Water density $\rho_{\text{H}_2\text{O}}$</td>
<td>1000 kg/m$^3$</td>
</tr>
<tr>
<td>$\text{H}_2\text{SO}<em>4$-air collision diameter $d</em>{\text{H}_2\text{SO}_4}$</td>
<td>$4.3 \times 10^{-10}$ m</td>
</tr>
<tr>
<td>Plume condensation nuclei parameter $n_{Cn}$</td>
<td>100 particles cm$^{-3}$</td>
</tr>
<tr>
<td>Troposphere-stratosphere exchange timescale $\tau_{exchange}$</td>
<td>1.0 y</td>
</tr>
<tr>
<td>Mean stratosphere temperature $T_{strat}$</td>
<td>218.6 K</td>
</tr>
<tr>
<td>Mean stratosphere pressure $p_{strat}$</td>
<td>$5.3 \times 10^3$ Pa</td>
</tr>
<tr>
<td>Mean stratosphere OH concentration $\tilde{n}_{OH}$</td>
<td>$5.5 \times 10^5$ molecules/cm$^3$</td>
</tr>
<tr>
<td>Mean stratosphere $\text{H}<em>2\text{O}$ concentration $\tilde{n}</em>{\text{H}_2\text{O}}$</td>
<td>$7.0 \times 10^{12}$ molecules/cm$^3$</td>
</tr>
</tbody>
</table>

Table S3: Chemical reaction rates used in the aerosol microphysical model. When used, M is assumed to be background air. The rate coefficient of reaction (1) was checked against the more general formula given in (84) and found to agree closely under the stratospheric conditions studied. The rate coefficient of reaction (4) was calculated by integrating loss rate values given in (56) over the altitude range of the box model.

<table>
<thead>
<tr>
<th>#</th>
<th>Reaction</th>
<th>Rate coefficient</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\text{SO}_2 + \text{OH} + \text{M} \rightarrow \text{HSO}_3 + \text{M}$</td>
<td>$8.2 \times 10^{-13}/(7.0 \times 10^{17} +</td>
<td>\text{M}</td>
</tr>
<tr>
<td>2</td>
<td>$\text{HSO}_3 + \text{OH} \rightarrow \text{SO}_3 + \text{H}_2\text{O}$</td>
<td>$1 \times 10^{-11}$ cm$^3$ s$^{-1}$</td>
<td>(56)</td>
</tr>
<tr>
<td>3</td>
<td>$\text{SO}_3 + \text{H}_2\text{O} + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_4 + \text{H}_2\text{O}$</td>
<td>$1.2 \times 10^{-15}$ cm$^3$ s$^{-1}$</td>
<td>(84, 85)</td>
</tr>
<tr>
<td>4</td>
<td>$\text{H}_2\text{SO}_4 + h\nu \rightarrow \text{SO}_2 + \text{products}$</td>
<td>$3.8 \times 10^{-12}$ s$^{-1}$</td>
<td>(56)</td>
</tr>
</tbody>
</table>
Table S4: Results from the aerosol microphysical model when key parameters are varied. \( \langle M_S \rangle \) and \( \langle r_{eff} \rangle \) are the time-mean total S aerosol burden and particle radius, respectively. All simulations are run for three years with yearly eruptions that inject 100 Mt of SO\(_2\) (5\( \times \) Pinatubo).

<table>
<thead>
<tr>
<th>Simulation</th>
<th>( \langle M_S \rangle ) (Mt)</th>
<th>( \langle r_{eff} \rangle ) (( \mu )m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard case, 5( \times ) Pinatubo</td>
<td>27.3</td>
<td>0.67</td>
</tr>
<tr>
<td>( \tilde{n}_{OH} = 5.5 \times 10^4 ) molecules/cm(^3)</td>
<td>11.2</td>
<td>0.68</td>
</tr>
<tr>
<td>( \tilde{n}_{OH} = 5.5 \times 10^6 ) molecules/cm(^3)</td>
<td>31.0</td>
<td>0.63</td>
</tr>
<tr>
<td>( n_{Cn} = 10 ) particles cm(^{-3})</td>
<td>24.3</td>
<td>0.83</td>
</tr>
<tr>
<td>( n_{Cn} = 1000 ) particles cm(^{-3})</td>
<td>28.3</td>
<td>0.63</td>
</tr>
<tr>
<td>( \tau_{exchange} = 0.5 ) y</td>
<td>17.1</td>
<td>0.57</td>
</tr>
<tr>
<td>( \tau_{exchange} = 2 ) y</td>
<td>35.8</td>
<td>0.75</td>
</tr>
</tbody>
</table>
Figure S1: Palaeogeographic reconstruction for Rodinia at 717 Ma and distribution of the Franklin LIP. Modified from Li et al., (64). I = India; SA = southern Australia; NA = northern Australia; T = Tarim; SC = South China; M = Mongolia; EA = East Antarctica; Si = Siberia; NC = North China; L = Laurentia; NS = North Slope; ES = East Svalbard; G = Greenland; B = Baltica; A = Amazonia; WA = West Africa; Aw = western Avalonia; Ae = eastern Avalonia; Sf = So Francisco; C = Congo; K = Kalahari; R = Rio Plata.
Figure S2: Sulfur concentration and isotope data from the Natkusiak magmatic assemblage, sedimentary rocks of the Shaler Supergroup, and contact metamorphic rocks. Data plotted from geochemical database of Franklin sills, Natkusiak Basalts and Shaler Supergroup rocks (15).
Figure S3: Radiative-convective equilibrium temperature profile for Earth’s atmosphere under cloud-free conditions with CO$_2$ molar concentration of 300 ppmv, solar zenith angle of 60$^\circ$, 1 bar atmospheric pressure and present-day insolation. The red line shows model results with ozone UV absorption included. Dashed and dotted lines show the US standard mid-latitude and tropical atmosphere, respectively.
Figure S4: Atmospheric temperature profiles used for the plume calculations displayed in Fig. 2 in the main text. In each case the climate model was run until radiative-convective equilibrium was reached.
Figure S5: (top left) Plume radius (top right) temperature (bottom left) plume vertical speed and (bottom right) density vs. altitude as simulated by the axisymmetric plume model. In the bottom right panel, the circle and asterisk indicate the maximum plume height and the neutral buoyancy height, respectively. For this simulation we assumed $Q_0 = 5.6 \times 10^7$ J/s, $r_0 = 3$ m, $T_{g,0} = 525$ K and $u_0 = 10$ m/s and the same thermodynamic parameters as ($45$).
Figure S6: Same as Fig. 2 in the main text but assuming 10% of present-day ozone abundance.
Figure S7: Same as Figure S6 but also assuming background $N_2$ pressure of 0.8 bar.
Figure S8: (top) Aerosol effective radius and (bottom) stratospheric sulfur loading as a function of time simulated by the aerosol microphysics model. The model initial conditions correspond to a single Pinatubo-like injection of 20 Mt SO$_2$ into the stratosphere.
Figure S9: (top) Aerosol effective radius and (bottom) stratospheric aerosol S burden vs. time for the standard set of 1×, 5× and 25× Pinatubo aerosol forcing simulations (corresponds to Fig. 3A in the main text).
Figure S10: Same as Fig. 3A in the main text, except for pre-aerosol tropical planetary albedo of 0.3 (top plot) and 0.2 (bottom plot). The lines in the plot correspond to tropical mean radiative forcing, while the values in the legend are global means.
Figure S11: Same as Fig. 3A in the main text, except assuming a tropopause height of 10 km and simulation box domain from 30° N to 80° N, corresponding to the situation following a high latitude basaltic fissure eruption. The pre-aerosol mid-latitude planetary albedo is 0.365 (present-day value) in the top plot and 0.5 (high ice/snow cover case) in the bottom plot. At the highest eruption rate in the bottom plot, the combination of large aerosol particles and high pre-aerosol albedo leads to a small net warming.