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Selective and limited environmental stress caused by magmatic sulfur emissions from continental flood basalt eruptions

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Abstract

Major periods of environmental crisis occurring throughout the past 260 million years have been related to episodes of continental flood basalt (CFB) volcanism[1-4]. However, the majority of the proposed causal relationships have been rather qualitative in nature, in particular for the effects of large emissions of magmatic sulfur to the atmosphere[3,5]. CFB provinces are typically formed by numerous individual eruptions, each lasting years to decades, with hiatus periods lasting hundreds to thousands of years[6-8]. Using a global aerosol-climate model, we quantify the sulfur-induced environmental effects of individual decade-long CFB eruptions representative of the 14.7 Ma Roza eruption and individual eruptions in the 65 Ma Deccan Traps[6-8]. For a decade-long Deccan-scale eruption, we calculate a decadal-mean reduction in global surface temperature of 4.5 K. However, unless climate feedbacks were very different in ancient climates, surface temperatures would have recovered within less than 50 years after such an eruption ceased. Acid mists and fogs could have caused damage to vegetation in regions of prolonged exposure, such as at high elevations[11]. However, in contrast to previous studies[3,5,9,10], we find that sulfur species deposited by even century-long eruptions would not have acidified the surface ocean or soils sufficiently to cause a global biotic crisis because these ecosystems are strongly buffered. Based on current knowledge of eruption magnitudes and hiatus frequencies, we conclude that the environmental effects of magmatic sulfur were too localized and/or too short-lived to explain global catastrophic extinction losses without the occurrence of additional environmental stressors such as marine regressions or asteroid impacts.

Main Text

Typically, hundreds to thousands individual and volumetrically large (on the order of 1000 km^3) eruptions made up a CFB province (total volumes 0.1-4.0 million km^3) emplaced over timescales of 100,000s of years[11] with highly uncertain hiatus periods[6-8]. These eruptions far exceeded even the largest historic eruptions in terms of lava volume, duration and the amount of gases and aerosol particles emitted into the atmosphere[7,8]. Intriguingly, the timing of the emplacement of four out of five CFB provinces in the last 300 Myr coincides with periods of severe environmental turnover including mass extinctions events[1,2,4]. This striking age correlation[4,11] led to the suggestion of a causal link between periods of CFB volcanism and periods of environmental turnover[1-4,7]. Yet after more than four decades of research this hypothesis remains equivocal and contested[3,12].

It is well known from observations of historic eruptions that emissions of magmatic sulfur dioxide (SO_2) and its oxidation products like sulfuric acid aerosol are the main agents able to induce profound environmental change[13,14]. Consequently, climatic cooling and environmental acidification due to the emission and deposition of large quantities of magmatic sulfur are the two most commonly proposed causal agents for environmental turnover during periods of CFB volcanism[3,5,10,15]. However, no previous study took into account the buffering capacities of soils and other ecosystems when assessing the effects of acid rain, hence until now this causal link remains elusive and unquantified. Similarly, to assess the climatic effects of CFB eruptions, previous studies either relied on extrapolations of the surface cooling caused by explosive volcanism[3], or used simple relationships between the mass of sulfuric acid aerosol particles generated from SO_2 and its cooling effects[6]. Both approaches do not account for two key factors that may reduce the aerosol-induced cooling: (i) limited oxidant availability, affecting SO_2 conversion to acidic aerosol, and (ii) particle growth to large sizes, reducing the particle light-scattering efficiency and shortening particle lifetime in the atmosphere due to sedimentation. The relative importance of these processes has been quantified for short-lived explosive eruptions[16-18], but never for CFB eruptions, which differ fundamentally in terms of eruption style, height and duration of the SO_2 emissions.

To constrain the environmental effects and consequences for habitability induced by magmatic sulfur emissions from individual decade- to century-long flood basalt eruptions we use numerical models including a global aerosol model, GLOMAP[19], a soil and freshwater acidification model, MAGIC[20] and an Earth system model, GENIE[21] (Online Methods). Our model experiments are based on the well-constrained 14.7 Ma (mid-Miocene) Roza eruption emplaced in the youngest CFB province on Earth, the Columbia River Basalt Group, and individual eruptions in the 65 Ma Deccan Traps coinciding with the Cretaceous-Paleogene (K-Pg) mass extinction. The 14.7 Ma Roza eruption (total volume 1300 km^3) is the only individual CFB eruption with a constraint on both duration and emission fluxes of about 1200 Tg of SO_2 per annum for a decade or two[6]. Individual eruption volumes in the Deccan Traps also reached volumes in excess of 1000 km[38], but individual eruption durations are unknown. Plume rise modeling for basaltic fissure eruptions suggests rise altitudes of 9-13 km[22,23], corresponding to the upper troposphere/lower stratosphere. We simulate a 'Roza-scale' eruption by emitting 1,200 Tg of SO_2 per year into 9-13 km altitude at 120°W, 45°N, and a 'Deccan-scale' eruption by emitting 2,400 Tg of SO_2 per year at 135°E, 21°S. The latter is considered an upper bound for the SO_2 emitted, assuming either greater mean lava discharge rates or that more than one flow field had been active at any one time (Online Methods, Extended Data Table 1).

We find that the net climate effect of magmatic sulfur emitted by individual CFB eruptions is to reduce surface temperatures (Figure 1), resulting from the combined climatic effects of acidic aerosol particles and SO_2. The increase in acidic aerosol particles exert a negative radiative forcing acting to cool the climate via the aerosol direct forcing and the aerosol indirect forcing (due to changes in cloud reflectance caused by changes in cloud droplet concentrations). In contrast, any unoxidized SO_2 acts as a greenhouse gas and absorbs ultraviolet radiation, which warms climate (positive forcing). We show that the relationship between the amount of SO_2 emitted and the magnitude of these two opposing climate forcings is highly non-linear. For example, a 20-fold increase in SO_2 release leads to less than a 6-fold increase in negative forcing (Extended Data Table 4). This non-linearity is caused by the combination of limited aerosol production and differences in particle growth with increasing SO_2 emissions, but also the striking saturation of the aerosol indirect forcing, and the offset of the negative aerosol forcings by the positive forcing from SO_2 (Extended Data Table 4). For instance, we find that for a Roza-scale eruption only 60% of the emitted SO_2 eventually forms volcanic aerosol (~1,490 Tg of sulfuric acid aerosol per year) due to the sustained depletion of atmospheric oxidants, in particular the hydroxyl radical, OH (Extended Data Table 2). The saturation of the indirect forcing is caused by increasing aerosol concentrations effectively decreasing the sensitivity of cloud reflectance to changes in aerosol loading[19]. A previous study on super-eruptions also suggested that the forcing from volcanic SO_2 may offset the aerosol cooling[17]. However, the greenhouse gas forcing by SO_2 is not normally considered in climate model simulations of volcanic eruptions or their geo-engineering analogues. Yet we show that for a Deccan-scale eruption the SO_2 forcing (+1.4 W m^{-2}) offsets about 8% of the global mean aerosol forcing (-17.6 W m^{-2}; Extended Data Table 4).

Our simulations show that the tempo, frequency and duration of individual eruptions as well as hiatus periods strongly affect the severity and longevity of the climatic effects of CFB eruptions. For the most probable individual eruption duration of a decade, the upper limit of global mean surface temperature reduction is 6.6 K (90% confidence interval of -7.66 K to -5.74 K) by the end of year 10 for Deccan-scale eruptions (Figure 1 and Online Methods). For context, simulations of the 74 ka Toba eruption suggest peak global mean temperature changes of between -3.5 K and -10 K[18,24]. Assuming present-day, century-scale climate feedbacks and ignoring potential carbon-cycle feedbacks, the mean temperature changes during the first decade are substantial: -3 K for a Roza-scale eruption and -4.5 K for a Deccan-scale eruption. However, Earth would have remained habitable mainly because the predicted temperature changes are short-lived on geological timescales. For the temperature reductions to reach equilibrium an individual eruption would have to last far longer than 150 years or eruptions would have to occur in quick succession without hiatuses longer than a decade (Figure 1), which is less probable than decade-long eruptions and longer-lasting hiatuses[6-8]. Our estimates are at the lower end of previous estimates of global mean surface temperature reductions for 14.7 Ma Roza[6], and in good agreement with temperature reductions in the mid-Miocene[25]. For the K-Pg, the survival of ectothermic tetrapods at mid-latitudes (but not at high-latitudes and with the exception of lizards)[12], appears to support our findings of surface temperatures potentially dropping and fluctuating significantly on decadal timescales, but prolonged or sudden drops to subfreezing temperatures are not supported by either the fossil record[12] or our model simulations.

A previous study suggested that the climatic impact of CFB and large explosive eruptions may be limited by the same atmospheric processes[16]. However, we find that the processes controlling the magnitude of climatic impacts differ fundamentally between CFB and explosive eruptions due to the difference in eruption style (Extended Data Figure 1). A sustained release of SO_2 into the upper troposphere/lower stratosphere during a CFB eruption provides a sustained source of sulfuric acid vapour, albeit self-limited by oxidant availability. The sulfuric acid nucleates to form many tiny particles less than 10 nm that, following condensation and coagulation, grow to radii of between 0.15 to 0.4 µm depending on eruption scale, but further growth is limited because the high removal rates in the troposphere limit the particle lifetimes to about two weeks (Extended Data Table 2). Conversely, for large explosive eruptions that inject SO_2 into the stratosphere, particles typically have time to grow to radii much larger than 0.4 µm[16,18] due to differences in atmospheric circulation that result in slow removal rates in the stratosphere. Importantly, at particle radii between 0.2 µm and 0.4 µm sulfuric acid aerosol particles scatter more incoming solar radiation back to space than at larger sizes and particle removal via gravitational settling is insignificant. Hence, in relative terms, aerosol optical depth (AOD, a dimensionless measure of the degree to which the transmission of light is reduced due to absorption and scattering by aerosol particles) and therefore climate are perturbed more efficiently for CFB eruptions even though the generated aerosol burden per unit mass of SO_2 emitted is lower than for explosive eruptions (Extended Data Table 3).

Environmental acidification can affect ecosystems either due to direct exposure to acidic species, or indirectly through the acidification of soils and stream waters. In contrast to previous studies that all neglected the acid buffering capacities of soils and other ecosystems[3,5,9,10], we find that the soil-mediated (indirect) effects due to volcanic sulfur deposition on vegetation and ecosystems are too limited in both magnitude and spatial extent to directly explain global-scale mass extinction events (Table 1, Figure 2a). Accounting for a wide range of acid-sensitive soils, soil depths and acid buffering capacities, we find that podzols are well buffered for centuries of continued deposition rates below 5 $\text{kmol}_c\ \text{ha}^{-1}\ \text{a}^{-1}$, which only occur in a small region near the volcanic vents. Localized vegetation damage due to soil acidification is likely to have occurred only in soils that are extremely acid-sensitive and highly weathered such as oxisols (Extended Data Table 5). For the K-Pg, there is no evidence of podzolization in the calcareous and smectitic paleosols found in Montana, USA[10,15], which places an independent limit on the degree of soil acidification in line with our simulated Deccan-scale acid deposition rates (zonal mean of up to ~5.2 kmol$_c$ ha^{-1} a^{-1}).

Acidification of stream waters with an equilibrium pH of 3.9 could have affected sensitive freshwater species such as molluscs[26] where acid deposition rates exceed 3 kmol$_c$ ha^{-1} a^{-1} for at least 50 consecutive years (Table 1), although the effects are spatially limited to an area of about 30 degrees latitude (Figure 2a). Our prediction of stream acidification occurring in limited parts of the world is supported by the vertebrate fossil record and survival patterns of pH-sensitive species such as alligators, turtles and frogs, which experienced only small reductions in their numbers at the K-Pg[12,15]. In fact, the survival patterns of fish and amphibians constrains the pH of freshwaters to no less than 4[26] in line with our findings.

The impact on ocean biogeochemistry of sulfur deposition from decade-long volcanic eruptions is also negligible (Online Methods). At Deccan-scale rates, we calculate that volcanic sulfur deposition would have needed to proceed continuously for almost three millennia to drive a surface ocean pH decline comparable to the current anthropogenic perturbation of ~0.1 pH units (Extended Data Table 6).

Based on our modeling results, we propose that the direct effects of acid mists and fogs on vegetation caused the most lethal and immediate vegetation damage on the timescale of years to decades, particularly at high elevations[27,28]. The fact that there is no soil intermediary or long-term exposure requirement (Figure 2a) and that acidity of mists is likely much greater than that of rainfall[27] makes this a potent mechanism affecting some but not all parts of the world (Figure 2b). Our findings corroborate contemporary records of regional damage of susceptible vegetation following the Icelandic 1783-1784 AD Laki eruption[29] – a smaller-scale flood basalt eruption that emitted at least an order of magnitude less SO_2 than the annual emissions in our scenarios. In the present-day climate the interception of cloud-water with the surface is mostly restricted to upland areas, and the presence of neutralizing species in the cloud-water (such as calcium or ammonia) can reduce the effects. Therefore, persistent and global damage from acid mists in deep times seems possible only if the cloud distribution or amount were entirely different in deep time climates. For the Roza-scale and Deccan-scale eruptions, critical levels for ground-level SO_2 are not exceeded on a scale sufficient to cause severe foliar damage or to affect sensitive tree species (Online Methods), but SO_2 concentrations strongly depend on the height at which volcanic SO_2 is emitted.

Based on current knowledge of the tempo, duration of individual CFB-scale and hiatus periods we conclude that, in isolation, environmental acidification due to magmatic emissions of sulfur is unlikely to have directly caused catastrophic global-scale extinctions. We did not account for potential increases in acidity and toxicity caused by magmatic emissions of halogens. Model simulations of magmatic halogens emitted during pulsed eruptions in the 270 Ma Siberian Traps[9] suggest that their effects are localized. Our calculated acid deposition rates may be underestimated 30-50% assuming a SO_2 to HCl ratio[8] of 1:0.29 and dispersion and deposition like SO_2 (Online Methods). More severe environmental acidification is expected only for CFB provinces where non-magmatic halogen emissions play a role[9,30], which is not the case in the Deccan Traps or for 14.7 Ma Roza. We find that the climatic effects of episodic magmatic sulfur emissions could have been large enough to impair habitability only if eruption frequencies and lava discharge rates were high and sustained for centuries or longer without hiatuses. Such a longevity and intensity of individual eruptions, hence cooling of climate cannot be demonstrated convincingly for any CFB province emplaced in the Phanerozoic. In fact, if individual CFB eruptions lasted centuries or longer, then the mean magmatic gas release rate may have been lower[31], resulting in lower eruption column heights[23]. This in turn would suggest a reduced effect from magmatic sulfur on climate and spatially even more confined and perhaps subdued environmental effects.

Figures (main text)

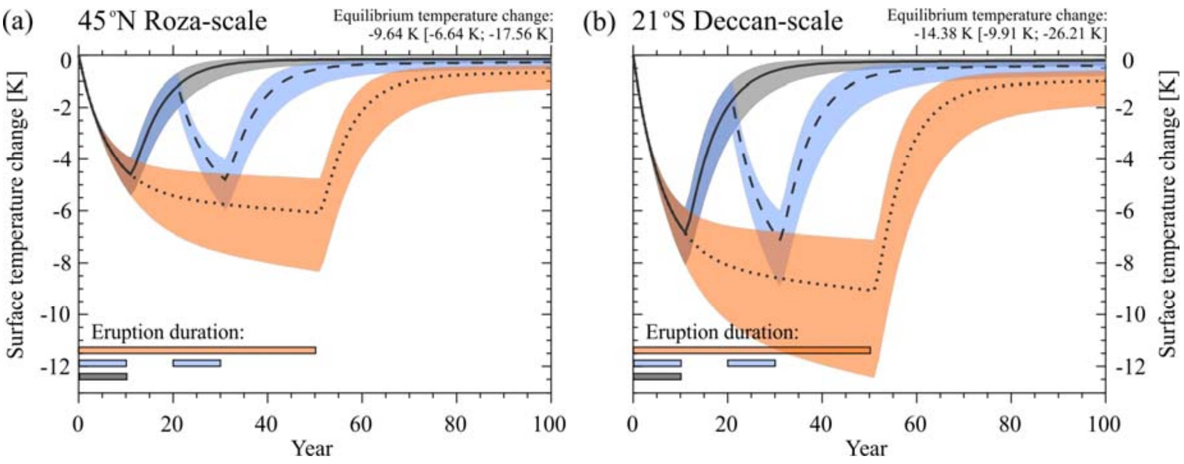

Figure 1. Global mean surface temperature change and its dependence on eruption duration and emission magnitude. (a) for a Roza-scale eruption emitting 1,200 Tg of SO_2 per year at 45°N and **(b)** for a Deccan-scale eruption emitting 2,400 Tg of SO_2 per year at 21°S. The eruption duration and hiatuses in each case are indicated by the colored bars (grey = 10 years of continuous eruption; blue = 10 years of continuous eruption followed by a 10-year hiatus followed by another 10 years of continuous eruption; and orange = 50 years of continuous eruption). The shading refers to uncertainty in surface temperature change based on 90% uncertainty range of the climate feedback parameter in CMIP5 models (Online Methods). The equilibrium temperature change including the 90% confidence interval is in the top-right corners and would require continuous SO_2 emissions for longer than 150 years.

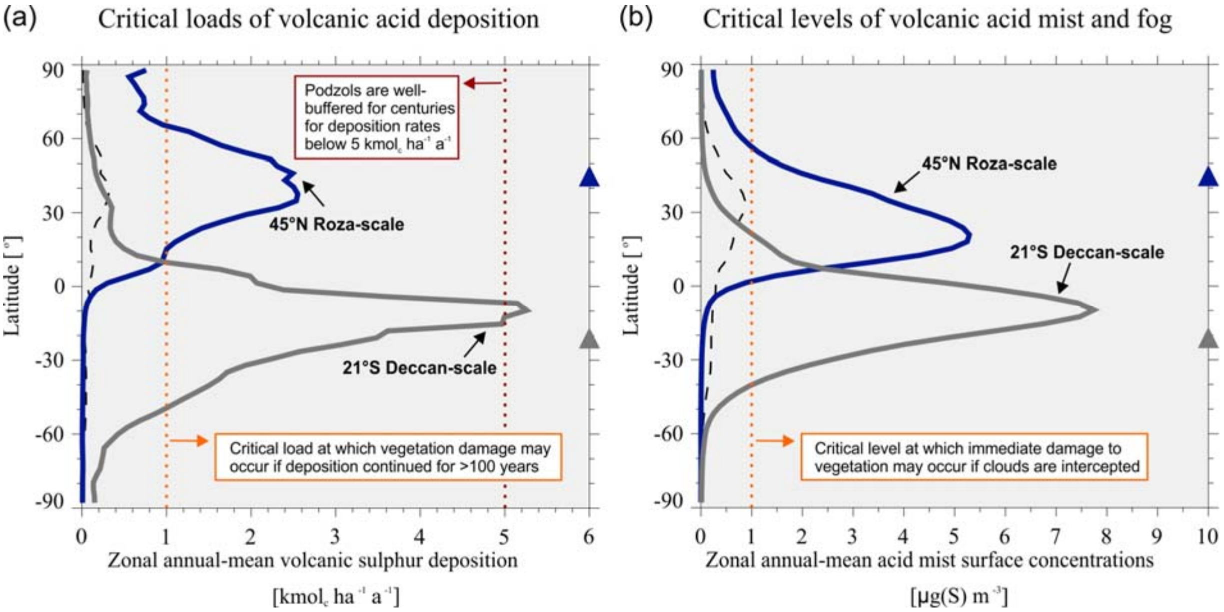

Figure 2. Annual zonal mean volcanic acid deposition rates and acid mist concentrations for CFB-scale eruptions in context with standards to protect soils, vegetation and stream water ecosystems from the effects of acid deposition ('critical loads') and direct exposure to pollutants ('critical levels')[26]. (a) Critical loads [kmol$_c$ ha^{-1} a^{-1}] for a 45°N Roza-scale eruption (blue line), a 21°S Deccan-scale eruption (gray line) and a year 2000 simulation with anthropogenic emissions only for context (dashed black line). In general, in our model the atmospheric dispersion of volcanic gases and aerosol particles is largely confined to the northern hemisphere an eruption at 45°N, whereas it is global for an eruption at 21°S. For both eruption scenarios critical loads of 1 kmol$_c$ ha^{-1} a^{-1} set to protect vegetation and forest ecosystems on the century scale[28] are exceeded on a hemispheric scale. We find that only very acid-sensitive soils such as Oxisols would be at risk due to deposition rates >1 kmol$_c$ ha^{-1} a^{-1}, whereas Podzols are well buffered for centuries below deposition rates of 5 kmol$_c$ ha^{-1} a^{-1} (Table 1). **(b)** critical levels [μg(S) m^{-3}] of acid mist concentrations for the same model experiments. The critical level of 1 μg(S) m^{-3} at which immediate damage to vegetation occurs if low-level clouds are intercepted[27] is exceeded on hemispheric scales for CFB eruptions of Roza-scale and larger making this a lethal mechanism to cause immediate vegetation damage where clouds are present.

Table (main text)

			Soil acidification parameters				**Stream water acidification parameters**				
			Soil- and vegetation-dependent, but BS ≤5% could be considered harmful		*Ca^{2+}:Al≤1 forest vegetation at risk of reduced growth, freezing injuries and dysfunction of fine roots*				*Acute effects on freshwater fish and amphibians*		*Acute effects on tolerant species if exceeded and pH<4.5*
Soil-type	**Initial soil and stream properties**	**Volcanic S deposition [$kmol_c \cdot ha^{-1} \cdot a^{-1}$]**	**Eq. BS [%]**	**Time to eq. [yr] / (Time to recover [yr])**	**Eq. Ca:Al**	**Time to fall below 1.0 [yr] / (Time to recover to 1.0 [yr])**	**Eq. stream pH**	**Time to eq. [yr] / (Time for full recovery [yr])**	**Time to reduce to pH<4.0 [yr] / (Time to recover to pH>4.0 [yr])**	**Eq. stream Al^{3+} [µeq L^{-1}]**	**Time to increase to 100 µeq L^{-1} [yr] / (Time to recover to 100 µeq L^{-1} [yr])**
Podzol Depth of 1.0 m	BS = 12.4 % Ca/Al = 5.6 Stream Al^{3+} = 0.0 µeq L^{-1} Stream pH = 6.85	**3**	6.2	1621 (2430)	1.1	-	4.10	300 (804)	-	73	-
		5	5.2	1014 (2590)	0.7	100 (16)	3.95	197 (865)	83 (4)	214	38 (8)
Podzol Depth of 0.25 m		**3**	6.2	791 (606)	1.1	-	4.10	75 (200)	-	73	-
Podzol 1.0m + Low sulfate adsorp.		**3**	6.2	1592 (2384)	1.1	-	4.10	98 (978)	-	73	-

Table 1. Indirect effects of volcanic sulfur deposition on soils and streams including damage threshold exceedances and their recovery timescales, accounting for the buffering capacities of these ecosystems. Orange shading indicates that thresholds to protect the ecosystem have been exceeded to a degree that harmful ecosystem effects occur. Green shading indicates the there are no threshold exceedances or harmful effects. The degree of soil acidification is too marginal for a wide range of soil parameters and different soil types (Extended Data Table 5) and spatially limited because deposition rates ≥5 $kmol_c$ ha^{-1} a^{-1} occur only in close proximity to the volcanic vent (Figure 2). Podzols are well buffered for deposition rates below 5 $kmol_c$ ha^{-1} a^{-1} and reach an equilibrium base saturation – the primary measure of soil acidification – of 6.2% at which no harmful effects are expected[26]. The Ca^{2+}:Al critical load for forest soils[26] and associated

vegetation is only exceeded if deposition rates ≥5 $\mathrm{kmol_c\ ha^{-1}\ a^{-1}}$ are applied for a century or longer, and recovery timescales are comparatively fast. In contrast to the marginal effects on soils, stream water acidification is more problematic. An equilibrium pH of 3.94 affecting sensitive freshwater species such as molluscs[26] occurs for acid deposition rates ≥3 $\mathrm{kmol_c\ ha^{-1}\ a^{-1}}$ applied for at least 50 consecutive years. For deposition rates ≥5 $\mathrm{kmol_c\ ha^{-1}\ a^{-1}}$ applied for about four decades the damage threshold for toxic inorganic monomeric aluminum (Al^{3+}) is exceeded, harming freshwater fish and other species if the pH drops below 4.5 (increasing the solubility of Al^{3+})[26].

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Author contributions:

A.S. and K.S.C. devised the study. A.S. ran and analyzed the model simulations and led the interpretation. A.S., T.T., S.S., M.W., R.A.S. and Andy.R designed model experiments. R.A.S. ran the soil and water acidification model simulations and interpreted the results together with A.S., and D.F. advised on the critical load calculations. Andy.R. run the GENIE model and interpreted the results. A.S. and P.M.F. calculated the SO_2 radiative forcing and ran the energy budget model. A.R. ran the radiative transfer code. AS led the writing and all authors contributed to the editing of the manuscript and approved the final version.

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