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Siberian Traps flood basalt magmatism coincided with the end-Permian mass 20 extinction approximately 252 million years ago. Proposed links between magmatism and 21 ecological catastrophe include global warming, global cooling, ozone depletion, and 22 changes in ocean chemistry. However, the critical combinations of environmental changes 23 responsible for global mass extinction are undetermined. In particular, the combined and 24 25 competing climate effects of sulfur and carbon outgassing remain to be quantified. Here we present results from global climate model simulations of flood basalt outgassing that 26 account for sulfur chemistry and aerosol microphysics with coupled atmosphere and ocean 27 28 circulation. We consider the effects of sulfur and carbon in isolation and in tandem. We find that coupling with the ocean strongly influences the climate response to flood basalt-29 scale outgassing. We suggest that sulfur and carbon emissions from the Siberian Traps 30 combined to generate systemic swings in temperature, ocean circulation, and hydrology 31 within a longer-term trend towards a greenhouse world in the early Triassic. 32 Geochemical proxies record profound shifts in carbon cycling, sea surface temperature 33 (SST), ocean chemistry and circulation, and weathering during the Permian-Triassic transition. 34 In particular, equatorial SST increased by 5-10 °C<sup>1-4</sup> in less than 100 kyr<sup>5</sup>; seawater <sup>87</sup>Sr/<sup>86</sup>Sr, 35 which reflects weathering of crustal material, underwent the most rapid increase in the 36 Phanerozoic<sup>6, 7</sup>; and carbon isotope data record a sharp negative excursion at the onset of the 37 end-Permian mass extinction, while boron isotope data record a pulse of ocean acidification 38 several tens of thousands of years after the carbon isotope excursion<sup>8</sup>. 39 Flood basalt provinces are enormous magmatic foci that recur at intervals of  $\sim 10^7$  years 40 on Earth<sup>9</sup>, emplacing magma volumes of 10<sup>5</sup>-10<sup>7</sup> km<sup>3</sup> accompanied by intense outgassing. In the 41

42 case of the Siberian Traps,  $\sim$ 7-15  $\times$ 10<sup>6</sup> km<sup>3</sup> intrusive and extrusive magmas were emplaced

beneath, into and on top of volatile-rich sedimentary basins<sup>10</sup>. Hypothesized environmental
consequences of flood basalts include release of toxic metals<sup>11</sup>, ozone depletion from halogen
emissions<sup>12, 13</sup>, enhanced weathering of fresh volcanic rocks<sup>14</sup>, and climate perturbations from
outgassing of sulfur and magmatic and sedimentary carbon <sup>10, 15-17</sup>.

We use the Community Earth System Model (CESM1), a three-dimensional (3-D) global 47 coupled climate model, to investigate the competing effects, on different timescales, of sustained 48 sulfur emissions and increases in atmospheric CO<sub>2</sub> (Figure 1c). Previous 3-D modeling 49 examined the atmospheric chemistry effects of Siberian Traps emissions alone<sup>13</sup>. Prior modeling 50 of the climate effects of flood basalt sulfur has employed present-day geography and offline 51 surface temperature calculation<sup>17</sup>, precluding investigation of the coupled atmosphere-ocean 52 response. Here we focus on climate consequences including hydrology and ocean circulation. 53 Our simulations include latest Permian paleogeography and the Community Aerosol and 54 Radiation Model for Atmospheres (CARMA), a sub-model of detailed aerosol microphysics 55 within the CESM1 framework, to capture sulfur aerosol growth and sedimentation (Methods; 56 Fig. ED2); both processes critically influence the climatic effects of volcanic aerosols<sup>17</sup>. 57

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### 59 Tempo and scale of Siberian Traps sulfur and carbon release

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The tempo and magnitude of outgassing are major sources of uncertainty. Sulfur concentrations in Siberian magmas are recorded in melt inclusions (see Methods). Geochemical evidence for pulsed magmatism from other flood basalt provinces suggests that time-averaged magma discharge rates vary substantially<sup>18</sup>. Comparison with recent fissure eruptions<sup>19</sup> and paleomagnetic data<sup>20</sup> suggest that during especially intense intervals, magma flux may have

exceeded the longer-term flux implied from geochronology<sup>21</sup> by 1-2 orders of magnitude, with 66 10<sup>3</sup>-10<sup>4</sup> km<sup>3</sup> of magma erupting in as little as 100 years. A unique aspect of flood basalt 67 eruptions is thus the potential for intervals of sustained, intense sulfur emissions lasting tens to 68 hundreds of years<sup>17</sup>. Eruption plume modeling indicates that during episodes of very high 69 eruption rate, columns could reach 13-17 km in height<sup>22</sup>. We consider upper troposphere-lower 70 stratosphere (UTLS; 12-14 km altitude) injection of 2000 Tg SO<sub>2</sub>/yr, spanning 10-200 years 71 (each year of emissions represents  $\sim 100 \times$  the SO<sub>2</sub> emissions of the Mount Pinatubo eruption in 72 1991). Our simulations span 4500 years, in contrast to the ~800 kyr duration of Siberian 73 magmatism<sup>21</sup>. Because the effects of sulfur outgassing are not cumulative beyond the lifetime of 74 sulfur in the atmosphere, stresses due to the most intense episodes of outgassing may be most 75 relevant to global biota<sup>23</sup>. We therefore focus on the climate response to such episodes, which 76 77 may have been repeated with varying intensity during the overall history of the large igneous province. 78

A central quandary of flood basalt carbon is that paleoclimate and geochemical records 79 are best explained through massive release of carbon with a temporally evolving isotopic 80 signature<sup>1, 8, 24</sup>, but carbon outgassing related to lavas, deep and shallow intrusive magmas, and 81 heating of surrounding rocks remains much more challenging to quantify than sulfur emissions. 82 This is in part because CO<sub>2</sub> saturation can occur deep in the magmatic system and carbon 83 solubility decreases strongly towards Earth's surface, partitioning CO2 into an exsolved volatile 84 phase<sup>10</sup>. While there are few direct petrologic constraints on Siberian Traps magmatic carbon, 85 comparison with Icelandic and Hawaiian basalts supports flood basalt magmas carrying up to ~1 86 wt% CO2<sup>10, 25</sup>, in addition to cryptic degassing from intrusive magmas<sup>26</sup> and CO2 release through 87 metamorphism and assimilation<sup>27</sup>. Recent measurements of CO<sub>2</sub> release during continental 88

rifting<sup>28</sup> also point to the continental lithospheric mantle as a potentially important, but poorly
known, source of carbon during flood basalt magmatism.

Motivated by  $pCO_2$  proxy data and inverse modeling of the carbon cycle<sup>2</sup>, we consider 91 progressive increases in  $pCO_2$  going from 710 to 2800 and then 5600 ppmv CO<sub>2</sub> (see Methods). 92 Such increases would each require 20,000-30,000 Pg of CO<sub>2</sub> release (Fig. ED3), consistent with 93 the carbon available from extraction of carbon from  $\sim 10^5$ - $10^6$  km<sup>3</sup> magma on decamillenial 94 timescales<sup>10, 24</sup> or from smaller magma volumes that also tap crustal carbon reservoirs. Only a 95 fraction of any such eruption interval would likely be sufficiently intense to inject sulfur into the 96 UTLS<sup>22</sup>. Furthermore, substantial assimilation or metamorphism of country rocks will enrich 97 gases more strongly in carbon than sulfur species<sup>27, 29</sup>. Thus, our combined sulfur and carbon 98 simulations encapsulate the effects of UTLS injection of sulfur during the most intense volcanic 99 activity (10-100 years) synchronized with more prolonged intervals ( $10^3$ - $10^4$  years) during which 100 101 carbon accumulated rapidly in the atmosphere due to volcanism, intrusion, and metamorphism. To account for uncertainty in the tempo of Siberian Traps outgassing and the degree to which 102 sulfur and carbon outgassing are synchronized, we present simulations in which sulfur and 103 carbon are considered independently before examining the potential combined effects. 104

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## 106 Climate response to Siberian Traps outgassing

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Our simulations demonstrate that as  $pCO_2$  reached ~2800 ppm, annually averaged equatorial sea surface temperatures were 29-30°C; at ~5600 ppm CO<sub>2</sub> annually averaged equatorial sea surface temperatures were ~32°C. Surface air temperatures were significantly hotter on land, in places reaching annual mean temperatures of ~38°C at 2800 ppm CO<sub>2</sub> and

almost 45 °C at 5600 ppm CO<sub>2</sub> (Figure 2b). The model reproduces the observed (Fig. 3) +8-10°C 112 anomaly in Permian-Triassic sea surface temperatures<sup>1, 4</sup> under the highest CO<sub>2</sub> levels we 113 consider, after ~3 doublings of CO<sub>2</sub>. Oxygen solubility is inversely related to salinity and water 114 temperature, but evidence for end-Permian anoxia is most prevalent at high latitudes<sup>30</sup>. While 115 high latitude SST is lower than tropical SST in all simulations, increasing  $pCO_2$  results in sharper 116 relative decreases in oxygen solubility at high latitudes due to a diminished equator-to-pole 117 temperature gradient (Fig. ED4). More sluggish ventilation under greenhouse conditions, 118 especially in the Tethys, would exacerbate oxygen depletion<sup>31</sup>. 119

In contrast to greenhouse gases, volcanic sulfur emissions quickly form sulfate aerosols 120 that exert a net cooling effect on climate<sup>17</sup>. Because Siberian Traps eruptions occurred at ~60 °N, 121 most sulfate aerosols remain in the northern hemisphere<sup>32</sup>. For UTLS injection of 2000 Tg/yr 122 SO<sub>2</sub> under constant baseline greenhouse levels, we find a maximum northern hemisphere optical 123 depth of ~5. Increased optical depth results in a global mean temperature decrease of 1.5-3 °C 124 (Fig. ED5b) after 10 years of emissions, with annual mean temperature decreases of ~10-15 °C 125 in the northern hemisphere on land (Fig. ED5a). While cooling of northern hemisphere 126 landmasses is severe, global mean cooling is more moderate than previous estimates of flood 127 basalt cooling (Fig. ED5b). We attribute this partly to the high paleolatitude of the Siberian 128 Traps, and partly to the strong ocean response to carbon and sulfur emissions. 129

We find that changes in ocean circulation play an important role in mediating changes in surface temperature due to prolonged flood basalt sulfur emissions. Changes in mixed layer density and the equator-to-pole thermal gradient due to changes in surface temperature and hydrology translate to changes in the strength of ocean overturning, with vigorous overturning in the sulfur-dominated climate mode and weaker overturning in the carbon mode. Strengthened

135 meridional overturning circulation (MOC) has been demonstrated due to cooling and diminished high-latitude runoff after eruptions of the past millennium<sup>33</sup>. The ocean circulation response to 136 flood basalt-scale sulfur emissions in our simulations is more pronounced by an order of 137 magnitude, probably due to prolonged cooling and correspondingly larger effects on ocean heat 138 content. A caveat is that the strength of the MOC response to volcanic eruptions has been shown 139 to vary across models<sup>34</sup> and to depend on background conditions<sup>35</sup>. Changes in ocean circulation 140 also affect the patterns of surface temperature change. For example, enhanced Panthalassic 141 overturning and poleward heat transport due to prolonged sulfur emissions cause regions of 142 warming in the northern Panthalassic ocean (Fig. 2a, Fig. ED5a). Within a year after sulfur 143 emissions cease, maximum northern hemisphere optical depth decays to less than 0.1. In 144 contrast, ocean circulation takes several decades to recover after sulfur emissions wane (Fig. 1), 145 indicating that changes in ocean circulation and heat content can extend the climate effects of 146 flood basalt sulfur emissions well beyond the lifetime of aerosols in the atmosphere. 147 Finally, it is well-known from the present day that aerosols and CO<sub>2</sub> cause competing 148 effects on the hydrological cycle due to the positive Clausius-Clapeyron slope of water vapor, 149 which indicates that to first order, increases in surface temperature correspond to increased 150 atmospheric water vapor concentrations<sup>36</sup>. Consequently, global greenhouse conditions lead to an 151 intensification of the hydrological cycle<sup>37</sup> on longer timescales (Fig. 2d). During intervals with 152 vigorous sulfur release, the patterns of precipitation shift in the opposite direction (Fig. 2c). 153 154 Predicted climate swings and comparison with the proxy record 155

The coupled changes in surface temperature, ocean circulation, and hydrology lead us to identify a cooler 'sulfur mode' that characterizes intervals of intense UTLS sulfur injection, and a warmer 'carbon mode' that prevails on longer timescales when high-altitude sulfur injection wanes. Multiple episodes of intense magmatism imply repeated swings between these sulfur and carbon modes. Given the idealized forcing and compressed timeline of our simulations, comparison of model and proxy data (Fig. 3) is performed to assess consistency of magnitude, sign, and dynamics, rather than to establish detailed alignment.

The maximum temporal resolution of available temperature proxy records from the PT 164 interval<sup>4</sup> is  $\sim 10^4$  years, too coarse to confidently resolve swings on the timescales discussed here 165 (Fig. 4). In this context, our results serve as predictions that could be most reliably tested with 166 higher resolution proxy records. However, significant evidence already exists for repeated shifts 167 in environmental conditions during flood basalt magmatism. Evidence for initial cooling 168 followed by warming during other flood basalt eruptions has been attributed to early liberation of 169 sulfur from cratonic lithosphere<sup>38</sup>. While release of sulfur and carbon from the mantle lithosphere 170 may well be an important process during continental flood basalt magmatism, our results show it 171 is not necessary to explain such apparent swings between sulfur-dominated and carbon-172 dominated climate modes. Uranium isotope-based records of marine redox conditions also show 173 marked fluctuations through the end-Permian mass extinction interval<sup>30, 39</sup>. Sulfur isotope 174 variations have been interpreted as evidence for variations in overturning vigor during this 175 time<sup>40</sup>. Such fluctuations are consistent with swings in oxygen solubility, runoff, ventilation, and 176 productivity. As with oxygen isotope records from the end-Permian, the temporal resolution of 177 available U-isotope datasets<sup>39, 41</sup> is limited to  $\sim 10^4$  years. If these records reflect the swings 178

described here, the signal must be sufficiently large that it is robust even with time-averaged
records, or individual data points represent snapshots of shifting conditions.

Over the long-term, our model predicts strong warming-induced increases in runoff, 181 especially at high latitudes as the equator-to-pole temperature gradient diminishes, providing a 182 mechanism to explain the steep increase in seawater <sup>87</sup>Sr/<sup>86</sup>Sr from the latest Permian to the early 183 Triassic (Figure 3). The increase in <sup>87</sup>Sr/<sup>86</sup>Sr has been interpreted as a consequence of intense 184 continental weathering at this time<sup>6, 7</sup>. In the northern high latitudes, eruption of the Siberian 185 Traps emplaced millions of square kilometers of fresh volcaniclastic rocks and early lavas 186 enriched in radiogenic <sup>87</sup>Sr, likely acquired through interaction with ancient Siberian continental 187 material<sup>42, 43</sup>. Consequent ocean fertilization and enhanced productivity may have contributed to 188 ocean anoxia<sup>44</sup>. 189

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### 191 Punctuated environmental stresses from Siberian flood basalts

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Even given uncertainties in the tempo of flood basalt outgassing and the origins of the 193 carbon that caused the end-Permian  $\delta^{13}C$  excursion and elevated pCO<sub>2</sub><sup>2</sup>, our results strongly 194 support several conclusions. The atmospheric residence time of sulfur aerosols is very short 195 relative to that of CO2<sup>15, 17</sup>, and flood basalt eruption column modeling<sup>22</sup> demonstrates that high-196 altitude sulfur injection is only possible during intense episodes that span a small fraction of the 197 overall duration of the extinction interval. Therefore, we predict swings between a cooler sulfur-198 driven climate mode during intervals of intense Siberian Traps fire-fountaining and a warmer 199 carbon-driven climate mode on longer timescales. Our modeling demonstrates that these swings 200 201 in climate encompass ocean circulation, marine oxygen solubility, and precipitation and runoff

patterns. Available temperature proxy data from the end-Permian mass extinction<sup>1, 3, 4</sup> are
insufficiently high-resolution to directly test for rapid swings in surface temperature, though
other proxies record fluctuations in ocean circulation<sup>40</sup> and redox<sup>30</sup>. If future very high-resolution
data from the end-Permian do not show the predicted climatic swings, this implies that most
Siberian Traps sulfur did not reach the UTLS or that fundamental gaps exist in understanding of
the climate response to flood basalt outgassing.

Extinction depends on the rapidity of environmental change relative to organisms' capacity to  $adapt^{45}$ . Variations in magma emplacement rate, volatile sources, and volatile concentrations over the life cycle of flood basalt provinces, along with differences in atmospheric lifetime, imply that environmental changes due to magmatism should be evolving rather than monotonic. By quantifying the contrary effects of CO<sub>2</sub> and SO<sub>2</sub> on different timescales, our simulations relate Siberian Traps magmatism to the punctuated deterioration of global ecosystems during the end-Permian mass extinction.

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# 218 Figures

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# Figure 1. Outgassing scenarios and changes in global surface temperature and ocean

- circulation. (a). Global temperature anomalies for increases in pCO<sub>2</sub> alone (panel c, blue curve)
- and for combined sulfur and carbon emissions (panel c, black and blue curves). Time intervals
- shown in Fig. 2 are indicated. (b). Maximum zonal mean strength of the meridional overturning
- circulation (MOC) at greater than 500 meters depth in the northern hemisphere. (c). SO<sub>2</sub>
- emissions scenarios and pCO<sub>2</sub> levels considered here.

229	Figure 2. CO <sub>2</sub> and SO <sub>2</sub> cause competing effects on surface temperatures and hydrology. (a).
230	Annual mean surface temperature anomalies relative to 710 ppm CO <sub>2</sub> simulation with combined
231	CO <sub>2</sub> increase and 2000 Tg/year SO <sub>2</sub> . Warming in northern Panthalassa results from enhanced
232	overturning and increased northward heat transport. (b). As in a, but with no sulfur emissions
233	and $\sim$ 3 doublings of atmospheric CO <sub>2</sub> to 5600 ppm CO <sub>2</sub> . Estimates of peak SSTs (circled
234	numbers) as noted in Table ED4. (c). Annual mean precipitation change on land (relative to 710
235	ppm CO <sub>2</sub> simulation) with combined CO <sub>2</sub> increase and 2000 Tg/year SO <sub>2</sub> . (d). As in $c$ , but with
236	no sulfur emissions and ~3 doublings of atmospheric CO <sub>2</sub> .

Figure 3. Summary of end-Permian proxy data and comparison with model results. (a.) 239 proxy data<sup>3, 4, 6, 8, 39, 41</sup> originating from Tethyan sections in Turkey, U.A.E., Iran, and South 240 China. Error bars for  $\delta^{238}$ U are two standard deviations. Due to uncertainties in Permian-Triassic 241 seawater  $\delta^{18}$ O, relative temperature anomalies from  $\delta^{18}$ O proxy estimates may be more robust 242 than absolute temperatures<sup>3, 4</sup>. (b.) Reconstructed localities for proxy data. (c.) Changes in sea 243 surface temperature, salinity, O<sub>2</sub> solubility, and runoff for simulations shown in Fig. 2a and 2c. 244 Tethys and Panthalassa water temperatures are averaged across the extent of each at the sea 245 surface and 750 meter depth. 246 247

251	Figure 4. Conceptual illustration of how combined flood basalt SO <sub>2</sub> and CO <sub>2</sub> emissions
252	cause repeated climate swings. This illustration shows an example eruption-climate history.
253	The sulfur mode triggered by intense UTLS sulfur injection during the most intense eruptions
254	swings to a carbon mode on longer timescales. Swings can occur repeatedly. Such swings are not
255	expected to be preserved in a proxy record with a temporal resolution of $\sim 10^4$ years, but will
256	rather average out to reflect the long-term shift towards carbon-dominated conditions. TS is
257	surface temperature.

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472	
473	Data availability
474	Model outputs used to generate Figures 1-3 have been archived at PANGAEA (PDI 18756).
475	Output files exceeding 100MB are available from the authors. Proxy data are available from
476	original sources as cited in the text.
477	
478	Code availability
479	The CESM code is available at cesm.ucar.edu.
480	

481 Author contributions

- 482 BAB conceived the research and analyzed the results. BAB and RRN designed and performed
- the simulations with input from JFL, JTK, CAS, MMM, CB, and LTET. All authors contributed
- 484 to writing the manuscript.

### 485 Methods

### 486 Sulfur and CO<sub>2</sub> forcing levels

487 Sulfur loading

488

Maximum pre-eruptive sulfur concentrations in Siberian Traps melt inclusions from lavas 489 are  $\sim 0.18$  wt%<sup>29</sup>. During episodes of intense magmatism, studies of paleomagnetic secular 490 variation suggest packages of 5-15 flows encompassing 10<sup>4</sup> km<sup>320, 46, 47</sup> erupted in 100 years or 491 less. Assuming a magma density of  $\sim 3000 \text{ kg/m}^3$  and a magma mass eruption rate of 265 492 km<sup>3</sup>/year (comparable to the  $\geq$ 216 km<sup>3</sup>/year mass eruption rate estimated by <sup>47</sup>) yields a total 493 sulfur budget of 2900 Tg SO<sub>2</sub>/year. The tropopause altitude is lower at high latitudes, and is 494 sensitive to climate, with increasing tropopause altitude as  $pCO_2$  increases <sup>22</sup>. In our simulations, 495 the altitude of the tropopause above the Siberian Traps is ~11-13km (Fig. ED8, ED10). This 496 altitude is higher in our microphysical simulations than at present due to the warmer climate 497 (3550 ppm CO<sub>2</sub>) assumed in our microphysical simulations. Plume modeling indicates that 498 during episodes of very high eruption rate, flood basalt eruption columns can reach 13-17 km in 499 height <sup>22</sup>. Assuming that during episodes of intense volcanism,  $\sim 70\%$  of total dissolved SO<sub>2</sub> is 500 released and reaches 12-14 km altitude (through gas release at the vent during vigorous fire 501 502 fountaining and lofting in buoyant columns) we obtain our applied injection rate during magmatic pulses of 2000 Tg SO<sub>2</sub>/year. Because our assumed injection altitude straddles the 503 tropopause, a large fraction of the sulfur emissions remain in the troposphere, which is reflected 504 in a sulfur mean atmospheric residence time of several weeks, comparable to previous upper 505 tropospheric simulations<sup>17</sup>. While the precise  $SO_2$  flux is subject to uncertainties given the 506 limited available information on volcanic tempo, our assumed flux is consistent with previous 507 estimates <sup>17, 47-49</sup>, and is further consistent with our goal of investigating the effects of 508 representative combined SO<sub>2</sub> and CO<sub>2</sub> emissions during the most intense magmatic pulses. The 509 pace and timing of gas release within eruptive pulses is uncertain; we therefore assume steady 510 511 SO<sub>2</sub> injection spread evenly annually and over the course of the eruption episode. Between episodes of peak magmatic intensity, we assume much more modest SO<sub>2</sub> injection at the same 512 altitude but a rate of 20 Tg SO<sub>2</sub>/year (Extended Data Table 2). To explore the sensitivity of our 513 results to the applied sulfur release rates, the continuity of emissions, and to emissions altitude, 514 we performed sensitivity simulations as listed in Extended Data Table 3 and as illustrated in 515 Extended Data Figure 9. 516

517

518 CO<sub>2</sub> levels

Evidence for very rapid, dramatic increases in atmospheric greenhouse gas levels (and 519 most likely in atmospheric  $CO_2$  levels) includes: (i) Oxygen isotopes, which suggest that low-520 latitude Sea Surface Temperatures (SSTs) increased by 6–10 °C during the extinction interval <sup>1, 3</sup>, 521 <sup>4</sup>, (ii) the pattern of marine extinction, which was particularly severe among poorly buffered or 522 heavily calcified organisms, implying that ocean acidification, warming, and anoxia contributed 523 to the extinction <sup>50-52</sup>, (iii) the brevity of the major negative marine carbon isotope excursion, 524 which had a duration of 2-18.8 kyr<sup>5</sup>, and (iv) a transient drop in ocean pH interpreted as the 525 result of a ~10,000 year pulse of CO<sub>2</sub> release (Extended Data Figure 3). Although the source of 526 increasing greenhouse gas levels remains the subject of discussion <sup>53</sup>, the overwhelming 527 evidence for rapid increases (on millennial timescales) in atmospheric pCO<sub>2</sub> motivates our 528

simulations of stepped increases in pCO<sub>2</sub>. The specific levels we consider (710, 2800, and 5600 ppmv CO<sub>2</sub>) are subject to some uncertainty, but are consistent with: (i) the stomatal index proxy for pCO<sub>2</sub> across the PTB <sup>54-56</sup>, (ii) carbon cycle modeling of transient spikes in pCO<sub>2</sub> due to injection of carbon <sup>53, 57</sup>, and (iii) pCO<sub>2</sub> levels required for the model to reproduce proxy constraints on paleo-temperature (Extended Data Table 4 and 5).

As discussed in the main text, classic volcanological estimates suggest flood basalt 534 magmas do not release sufficient mantle carbon to cause the observed changes <sup>58-60</sup>, leading to 535 the hypothesis that volatile-rich sedimentary rocks supply the bulk of the carbon <sup>27, 61</sup>. However, 536 recent estimates suggest that the mass of carbon released from the mantle could be much larger 537 than previously thought<sup>10, 62, 63</sup>. To achieve  $pCO_2$  increases similar to those we model requires 538 release of  $>10^4$  Gt of CO<sub>2</sub> on decamillennial timescales (Fig. ED3), constituting  $\sim 10-50\%$  of the 539 estimated Siberian Traps mantle-derived CO<sub>2</sub> budget of 64,000-320,000 Gt CO<sub>2</sub><sup>10, 12, 62</sup>. In this 540 context, the observed Permo-Triassic greenhouse conditions<sup>1, 3, 4, 64</sup> can be achieved primarily 541 through elevated CO<sub>2</sub> via a combination of the following possibilities: (1) mantle-derived CO<sub>2</sub> 542 release at the high end of estimates, (2) large additional quantities of  $CO_2$  released due to 543 assimilation, thermal metamorphism, and other interactions with Siberian coal, carbonates, and 544 545 hydrocarbon-bearing sedimentary rocks, or (3) a mechanism that is unknown or unrelated to magmatism. 546

The specific timescales we consider (2000 years of model time after each  $pCO_2$  increase) 547 are designed to allow investigation of transient effects rather than equilibrium climate. The 548 timescale for the atmosphere to equilibrate with the ocean is  $10^2 - 10^3$  years <sup>65</sup>. Therefore a pulse 549 of CO<sub>2</sub> release lasting  $<10^3$  years will result in a transient peak in atmospheric pCO<sub>2</sub>, followed by 550 an exponential decline in pCO<sub>2</sub> as equilibration—first with the ocean, then with sediments, then 551 through weathering—proceeds<sup>59, 66</sup>. For simplicity, and to investigate the specific, transient Earth 552 system response to a step change in atmospheric pCO<sub>2</sub>, we choose not to model this decline. 553 554 Instead, we fix atmospheric pCO<sub>2</sub> between pulses of outgassing.

Ultimately, the step changes in  $pCO_2$  in our simulations are representative of and allow us 555 to investigate the effects of several emissions scenarios: pulsed emissions of magmatic CO<sub>2</sub> if 556 magmas are CO<sub>2</sub> rich; pulsed emissions of metamorphic CO<sub>2</sub> and CH<sub>4</sub> if metamorphic gas 557 release occurs rapidly during brief intrusive and extrusive magmatic pulses; or pulsed emissions 558 of CO<sub>2</sub> and CH<sub>4</sub> from the deep ocean and/or from clathrates, if changes in ocean temperature and 559 circulation due to initial magmatic outgassing result in a follow-on release of gas from the ocean 560 as proposed here. Because all of these emissions scenarios are fundamentally related, changes in 561 atmospheric CO<sub>2</sub> probably in reality result from some combination of these processes. Our 562 idealized pCO<sub>2</sub> history is intended to investigate the climate response to rapid changes in pCO<sub>2</sub> 563 (with or without concomitant  $SO_2$  release) due to any or all of these scenarios. 564

As noted in the main text, we report greenhouse forcing in CO<sub>2</sub>-equivalent levels of 565 greenhouse gases. Release of methane and other greenhouse gases may have been significant 566 during the end-Permian<sup>27, 55</sup>. From a radiative perspective (neglecting the potential effects of 567 methane on atmospheric chemistry) our simulations can equally represent CO<sub>2</sub> dominated 568 greenhouse conditions or outgassing of CO<sub>2</sub> in lower abundances along with significant, 569 sustained quantities of methane (sustained because the lifetime of methane in the atmosphere is 570 ~10 years, barring significant drawdown of OH concentrations). While methane and other 571 greenhouse gases may have played a significant role, the relatively rapid oxidation of methane 572

favors  $CO_2$  as a greenhouse gas, because greenhouse conditions were relatively long-lived<sup>1, 3, 4</sup>,

<sup>64</sup>, and oxidation of realistic quantities of methane leads to only several hundred ppm increases in  $CO_2^{67}$ .

576

# 577 Modeling methods

Our modeling approach has been adapted from that of <sup>13, 31, 68</sup>. The Community Earth 578 579 System Model (CESM1) is a global climate model that tracks the coupled interactions between the Earth's atmosphere, ocean, land, and sea ice to yield a comprehensive view of coupled Earth 580 systems<sup>69, 70</sup>. In order to combine detailed microphysical simulations with long atmosphere-581 ocean coupled simulations, we utilize the modeling approach illustrated in Extended Data Figure 582 2, in which we use two configurations: the Community Atmosphere Model (CAM4) with the 583 Community Aerosol and Radiation Model for Atmospheres (CARMA), and CAM4 with 584 interactive ocean. We describe the model components and calculations of oxygen solubility 585 based on model outputs in more detail below. 586

587

588 CARMA

589

Robust aerosol size distributions are critical to accurate assessment of radiative effects. 590 and such size distributions can vary significantly for different styles of eruption<sup>17, 71</sup>. We 591 therefore employ the Community Aerosol and Radiation Model for Atmospheres (CARMA, a 592 sectional aerosol microphysical model<sup>72</sup> that includes processes such as nucleation, growth and 593 settling<sup>72-74</sup>. In our work, CARMA has been configured with 35 aerosol size bins, from 0.003 594 microns to 7 microns, and has been coupled to CESM1(CAM4) to permit the simulation of the 595 aerosol plume in three dimensions through time<sup>72, 75, 76</sup>. All size bins are included as tracers 596 within CESM1(CAM4). Heating of sulfates from tropical eruptions can influence the dispersal of 597 the sulfates<sup>77, 78</sup>, a process that is not accounted for in our simulations. The expected 598 consequences of aerosol heating might include self-lofting, slower removal of stratospheric 599 600 aerosols, and increased transport into the southern hemisphere. Therefore the climate effects we consider are representative, though conservative, given the assumptions involved. 601

602

603 *CESM1(CAM4)* 604

The configuration of CESM1(CAM4) used in this study has 26 vertical levels that extend 605 to ~40 km altitude with  $1.9^{\circ} \times 2.5^{\circ}$  horizontal resolution. CESM1(CAM4) handles the 606 representation of dynamics, physics, atmospheric chemistry, and coupling to the land and 607 prescribed SSTs within CESM while CARMA simulates only the sulfate aerosol microphysics. 608 The equilibrium climate sensitivity of CESM1(CAM4) is 3.2 °C for a doubling of CO<sub>2</sub>, 609 while that of CESM1(CAM5) is 4.0 °C<sup>79</sup>. The CESM2 was just released to the community and 610 its equilibrium climate sensitivity is 4.2 °C. Importantly, these are model estimates of Earth's 611 climate sensitivity. Analysis of paleoclimate data, present day observations and a hierarchy of 612 climate models yield an estimated range of climate sensitivity from 2.0 to 4.5 °C, in which the 613 probability distribution of all of these estimates peaks at around 3 °C<sup>80</sup>. Given uncertainty in the 614 precise observational value of Earth's climate sensitivity, and given that our model falls well 615 within the estimate range and agrees with the most probable value, CESM1(CAM4) is well 616 suited for investigation of end-Permian climate. If the true end-Permian climate sensitivity is 617

closer to 4.0 °C per doubling of CO<sub>2</sub>, rather than 3.0 °C per doubling of CO<sub>2</sub>, then lower levels 618 619 of atmospheric carbon dioxide would yield similar changes to those we simulate. We use a version of CAM4 with extended sulfur chemistry to allow for the accurate 620 transformation of the volcanic SO<sub>2</sub> plume into sulfate. The conditions for all simulations 621 presented here are summarized in Table S2. Appropriate boundary conditions and 622 paleogeography for the latest Permian were used to force the simulations (Extended Data Table 623 1). Stratospheric circulation is slightly slower in the CAM4 model compared with WACCM and 624 CAM5/6, resulting in an aerosol lifetime that is slightly longer in CAM4<sup>81</sup>. 625 626 Prescription of CARMA results in CESM1(CAM4) under end-Permian conditions 627 628 In our simulations with interactive ocean circulation, an atmospheric model with a 629 horizontal resolution of  $3.75^{\circ} \times 3.75^{\circ}$  was used to enable the relatively long simulations 630 presented here. The radiative properties of the sulfate aerosol plume calculated by CARMA were 631 coupled to CESM1(CAM4) by calculating the optical properties of the plume from the output of 632 WACCM-CARMA simulations. To accomplish this, we first calculate the wet aerosol mass and 633 size from the dry sulfate radius and mass output by CARMA by accounting for temperature and 634 water activity<sup>72, 82</sup>. Mie calculations are performed using this information to determine the 635 radiative properties of the plume required by CESM1(CAM4)<sup>83</sup>. CESM1(CAM4) uses this input 636 and the CAM-RT (71) radiative transfer scheme to determine the full radiative effects of the 637 sulfate aerosol distributions in four dimensions (space and time) during fully coupled simulations 638 with latest Permian paleogeography $^{31}$ . 639 640 Ocean model 641 642 We use prescribed SSTs based on a climatology derived from our equilibrium 643 simulations with CESM1(CAM4) in our CESM1(CAM4)-CARMA simulations. All other 644 simulations include the fully interactive Parallel Ocean Program (POP2) model<sup>69, 84</sup>, with 645 bathymetry from Kiehl and Shields<sup>31</sup>. 646 647 Land dataset 648 The latest Permian land surface dataset used in our simulations is from Kiehl and Shields 649 <sup>31</sup>, based on the surface types of Rees et al.<sup>85</sup>. The land surface types are illustrated in Figure ED 650 11. 651 652 Oxygen solubility

653

We compute oxygen solubility (Figure 3, Figure ED4) using Eqn. 8 in Garcia and Gordon 655 <sup>86</sup> in conjunction with temperature and salinity predictions from our model.

656 **The role of methane clathrate** 

657

658 Recent work on the Paleocene-Eocene Thermal Maximum<sup>24</sup> has suggested clathrate 659 dissociation due to volcanogenic warming could amplify carbon release and explain an excursion 660 in  $\delta^{13}$ C. Oxidation of methane in seafloor sediments and in the water column limits methane

clathrate release to the atmosphere<sup>87</sup>. However, clathrate methane need only reach the water 661 column to cause an isotope excursion, and high methane fluxes due to rapid dissociation may 662 enable a larger fraction of methane to bypass the sediment filter and enter the ocean<sup>87</sup>. During a 663 swing from a sulfur- to carbon-dominated climate regime, enhanced overturning continues for 664 around 50 years after the conclusion of a 200-year pulse with 2000 Tg/yr SO<sub>2</sub> injection even as 665 the surface ocean warms. Under these conditions, bottom water temperatures increase at rates 666 that briefly reach 0.1-0.5 °C/year at the seafloor on continental margins. This rate of seafloor 667 warming is much more rapid than under increasing pCO<sub>2</sub> alone (Figs. ED6 and ED7), potentially 668 catalyzing a clathrate-driven drop in the  $\delta^{13}$ C of the end-Permian ocean (and eventually the 669 atmosphere through air-sea exchange of carbon). The magnitude of the isotope excursion 670 requires ~4000 Gt C with  $\delta^{13}$ C $\approx$ -60<sup>67</sup>. An estimated 1800-5000 Gt C is stored in clathrate and 671 methane bubbles in modern marine sediments<sup>87</sup>. Suboxic conditions like those that prevailed 672 leading into the end-Permian<sup>30, 52</sup> enhance clathrate accumulation<sup>87</sup>, suggesting that the latest 673 Permian clathrate inventory may have been larger than the modern inventory despite late 674 Permian warmth, which is further supported by persistently high carbonate  $\delta^{13}$ C prior to the 675 extinction transitioning to lower long-term carbonate  $\delta^{13}$ C after the extinction<sup>67</sup>. Indeed, the 676 release of large quantities of methane into the ocean and subsequent oxidation in the water 677 column would have promoted anoxic conditions. This scenario, which depends on 678 synchronization of carbon and sulfur outgassing, explains abrupt carbon cycle perturbation near 679 the onset of warming<sup>4, 5</sup>, coincident with expansion of anoxia<sup>41</sup>, and agrees with evidence for 680 increases in methane abundance in seawater from enclaves in brachiopod shells<sup>88</sup>. 681