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# 1 A projected decrease in lightning under climate change

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9

10 **Lightning strongly influences atmospheric chemistry<sup>1-3</sup>, and impacts the frequency of**  
11 **natural wildfires<sup>4</sup>. Most previous studies project an increase in global lightning with**  
12 **climate change over the coming century<sup>1,5-7</sup>, but these typically use parametrisations**  
13 **of lightning that neglect cloud ice fluxes, a component generally considered to be**  
14 **fundamental to thunderstorm charging<sup>8</sup>. As such, the response of lightning to climate**  
15 **change is uncertain. Here, we compare lightning projections for 2100 using two**  
16 **parametrisations: the widely-used cloud-top height (CTH) approach<sup>9</sup>, and a new**  
17 **upward cloud ice flux (IFLUX) approach<sup>10</sup> that overcomes previous limitations. In**  
18 **contrast to the previously reported global increase in lightning based on CTH, we find**  
19 **a 15% decrease in total lightning flash rate with IFLUX in 2100 under a strong global**  
20 **warming scenario. Differences are largest in the tropics, where most lightning occurs,**  
21 **with implications for the estimation of future changes in tropospheric ozone and**  
22 **methane, as well as differences in their radiative forcings. These results suggest that**  
23 **lightning schemes more closely related to cloud-ice and microphysical processes are**  
24 **needed to robustly estimate future changes in lightning and atmospheric**  
25 **composition.**

26 Changes in climate over the next century are expected to alter atmospheric temperature,  
27 humidity, stability and dynamics<sup>11</sup>. The leading theory for electrical charge generation in  
28 thunderstorms<sup>8,12</sup> suggests that the occurrence of lightning depends on all these factors,  
29 through their effect on convection and colliding ice and graupel particles. Lightning is an

30 important source of nitric oxide (NO), a precursor of ozone and the hydroxyl radical (OH)  
31 which governs the lifetime of greenhouse gases such as methane<sup>1</sup>. Both ozone and  
32 methane are important greenhouse gases, and changes in their concentrations can lead to a  
33 warming or cooling of the atmosphere. Thus, lightning needs to be represented in chemistry-  
34 climate models to fully simulate the interactions and feedbacks between atmospheric  
35 composition and climate change. Future changes in lightning are also of importance for  
36 aerosol chemistry<sup>2,3</sup>, wild-fire ignition<sup>4</sup>, and damage to infrastructure and to human health.

37 Recent studies<sup>4,6,7,13,14</sup> simulating future lightning over the next century with the CTH  
38 approach have reported 5-16% increases in lightning flashes per degree increase in global  
39 mean surface temperature. Observational studies have shown lightning to be positively  
40 correlated with surface temperature on daily to decadal time scales, but such relationships  
41 become highly uncertain on longer time scales<sup>15,16</sup>. An alternative<sup>6</sup> to the CTH scheme, using  
42 cold cloud depth to parametrise lightning, suggested a smaller increase in lightning under  
43 climate change of  $\sim 4\% \text{ K}^{-1}$ . Furthermore, a decrease in future lightning has been found using  
44 a convective mass flux-based lightning scheme<sup>17</sup> in two recent studies<sup>6,18</sup>. However, this  
45 scheme has been found to perform poorly against observations<sup>6,10,19</sup>.

46 Only one study to date has used a lightning scheme dependent on cloud ice particles to  
47 project future lightning. This study found a decrease in lightning associated with an increase  
48 in temperature<sup>20</sup>. However, the study had a near-term focus on 2030 and the global surface  
49 temperature increase was less than 0.2K. It is not clear whether a similar response occurs  
50 for larger changes in temperature at the end of the century<sup>20</sup>.

51 In this study we use both the established CTH scheme<sup>9</sup> and the recently developed and  
52 evaluated IFLUX scheme<sup>10</sup> (see Methods) in a chemistry-climate model to simulate future  
53 lightning and its influence on atmospheric composition and radiative forcing. Atmospheric  
54 dynamics are decoupled from changes in atmospheric composition so that both lightning  
55 schemes use the same underlying meteorology. With the same model as used here, the  
56 IFLUX scheme has shown a more realistic representation of present-day global lightning and

57 tropospheric ozone than the CTH approach, especially in the tropics<sup>10,21</sup>. For instance, the  
58 spatial correlation of the global, annual lightning distribution compared to observations was  
59  $r=0.78$  using the IFLUX scheme and  $r=0.65$  using the CTH scheme. The temporal correlation  
60 of the annual cycle of southern/northern tropical upper tropospheric ozone against  
61 observations was  $r=0.93/0.65$  with the IFLUX scheme and  $r=0.79/0.26$  with the CTH  
62 scheme<sup>21</sup>. Whilst accurately representing present-day lightning does not guarantee that  
63 long-term trends can be captured, it does increase our confidence in the lightning scheme. In  
64 the IFLUX scheme, the upward ice flux is sampled at a specified pressure level. Shifts in the  
65 tropopause and vertical temperature profile (Supplementary Figure 1) suggest a shift in the  
66 vertical extent of deep convection and ice particle formation, and therefore a higher sampling  
67 level is found to be more appropriate under future climate change (see Methods).

68 Lightning  $\text{NO}_x$  emissions ( $\text{LNO}_x$ ) from existing lightning parametrisations scale linearly with  
69 changes in global mean surface temperature across chemistry-climate models<sup>18</sup>. Therefore,  
70 we use the year 2100 under Representative Concentration Pathway 8.5 (RCP8.5)<sup>22</sup> to obtain  
71 a clear lightning response to substantial climate change. We provide the first estimate of the  
72 future lightning response to long-term global warming in 2100 using a cloud ice-based  
73 lightning parametrisation, and compare with results from the widely-used CTH scheme.

74 We simulate a decrease in global total lightning of  $2.2 (1.9-2.5) \times 10^8 \text{ fl. yr}^{-1}$  by 2100 with the  
75 IFLUX scheme (Table 2 Global lightning and atmospheric composition properties, simulated  
76 for present-day and future with different lightning schemes and with no lightning. Percentage  
77 changes are relative to year 2000 for each approach.

78 Figure 1), where the range is the 95% confidence interval based on the simulated  
79 interannual variability. A sensitivity test diagnosing the ice flux at the same level as under  
80 present-day climate shows a decrease of  $5.8 (5.6-6.1) \times 10^8 \text{ fl. yr}^{-1}$  (Supplementary Figure  
81 2b), suggesting that the choice of level does not influence our conclusions. With the CTH  
82 scheme, we simulate an increase in the global flash rate of  $6.1 (5.9-6.2) \times 10^8 \text{ fl. yr}^{-1}$  in year  
83 2100.

84 Regionally, the IFLUX and CTH approaches result in increases in *total* lightning over the  
85 USA of 3.4 and 14.2 %K<sup>-1</sup>, respectively. Assuming total and cloud-to-ground lightning  
86 respond similarly to climate change, our results are consistent with a recent study that used  
87 convective available potential energy and precipitation to parametrise lightning<sup>23</sup>. In that  
88 study it was estimated that *cloud-to-ground* lightning over the USA would increase by 12 %K<sup>-1</sup>  
89 under RCP8.5<sup>23</sup> (with a range of 3-18%K<sup>-1</sup> across models). However, this increase does not  
90 apply to all mid-latitude locations. For instance, we find no significant change over most of  
91 Europe with either lightning scheme.

92 Whilst several studies have considered how climate variability, such as El Niño driven  
93 events, affects tropical lightning<sup>15,24</sup>, the impact of climate change on tropical lightning has  
94 not received much attention. This is despite ~80% of global lightning flashes occurring in the  
95 tropics and subtropics<sup>25</sup>. With the IFLUX approach, a decrease in tropical lightning is  
96 simulated, in contrast to an increase with the CTH approach. The tropical cloud top height,  
97 used to diagnose lightning in the CTH scheme increases by 900m (7%) in the future. This  
98 has a large impact on lightning due to the fifth-order dependence on cloud top height in the  
99 scheme. Furthermore, basing the change in lightning solely on cloud-top height disregards  
100 key changes in updraughts and ice content of the cloud, displayed in Figure 2, that govern  
101 lightning generation.

102 The cloud ice content and convective updraught mass flux decrease over tropical land in the  
103 mid-troposphere in the future (Fig. 2), where the thunderstorm charging zone is located. A  
104 shift in the distributions to higher altitudes is apparent, justifying the use of a higher sampling  
105 level in the future climate. Reductions in the convective and total cloud fraction throughout  
106 most of the troposphere in future are consistent with a ~20% reduction in the probability of  
107 lightning with the IFLUX scheme (Supplementary Table 1). A 28% reduction in the  
108 magnitude of total tropical flashes with the IFLUX approach (Supplementary Table 1) results  
109 from a combination of the change in probability of lightning flashes and decreases in cloud  
110 ice content and updraught mass flux.

111

112 The responses of the convective and cloud ice variables in the model to climate change are  
113 physically reasonable. For instance, the increase in cloud top height largely reflects the  
114 increase in tropopause height, a robust feature of global warming<sup>26</sup>. A reduction in cloud ice  
115 content, even when sampling at a higher altitude in the future climate (Fig. 2a), is consistent  
116 with an increase in tropospheric temperatures. The Intergovernmental Panel on Climate  
117 Change (IPCC)<sup>11</sup> reports a projected future decrease in mean updraught mass flux  
118 associated with weakened tropical ascent in the climate models, and a decrease in cloud  
119 fraction over tropical land except around the tropopause.

120 Despite the consistency between our results and IPCC models, the meteorological drivers of  
121 the IFLUX scheme, and their response to climate change, remain highly uncertain. For  
122 instance, many models underestimate tropical cloud ice content<sup>27</sup>. The formation of cloud ice  
123 depends on ice nuclei and secondary formation processes which are not well-represented in  
124 global models, and model resolution is generally insufficient to explicitly simulate storm-scale  
125 updraughts, highlighting the challenges in parametrising lightning at the global scale.

126 Nevertheless, it is evident from our results that these key drivers of the lightning response to  
127 climate change are not captured by the CTH approach.

128 Most studies report future increases in LNO<sub>x</sub>, as they employ the CTH scheme. One  
129 previous study found these future increases in LNO<sub>x</sub> more than offset the reduction in  
130 tropospheric ozone arising from lower anthropogenic emissions of NO<sub>x</sub> and other ozone  
131 precursors<sup>7</sup>. In our study, other ozone precursor emissions are kept constant so that  
132 changes in ozone burden due to changes in climate and LNO<sub>x</sub> can be quantified. In our  
133 model, each lightning flash produces 250 mol of NO. Given a present-day lightning emission  
134 of ~5 TgN yr<sup>-1</sup>, the responses to climate change using the IFLUX and CTH schemes are -  
135 0.15 TgN K<sup>-1</sup> and +0.44 TgN K<sup>-1</sup>, respectively. The CTH response closely matches results  
136 from a recent multi-model intercomparison of 10 models using the CTH scheme<sup>18</sup>.

137 Global lightning and atmospheric composition responses for model simulations are given in  
 138 Table 1. In the absence of lightning (ZERO simulations) there is a decrease in the  
 139 tropospheric ozone burden and methane lifetime under climate change. This occurs primarily  
 140 because increased water vapour in the warmer climate leads to greater loss of ozone<sup>28</sup>,  
 141 mainly via the increase in OH radicals through reaction of water with O(<sup>1</sup>D). The OH also  
 142 acts as a sink for methane, reducing the methane lifetime.

143 **Table 1** Global lightning and atmospheric composition properties, simulated for present-day and future with  
 144 different lightning schemes and with no lightning. Percentage changes are relative to year 2000 for each  
 145 approach.

Simulation	Global lightning ( $\times 10^9$ fl. yr <sup>-1</sup> )	Tropospheric ozone burden (DU)	Tropospheric ozone lifetime (days)	Methane lifetime (yrs)
ZERO-2000	0.00	209	18.7	12.5
ZERO-2100	0.00 (0%)	191 (-9%)	15.5 (-17%)	9.9 (-21%)
CTH-2000	1.41	271	20.1	9.9
CTH-2100	2.02 (+43%)	266 (-2%)	16.9 (-16%)	7.5 (-24%)
IFLUX-2000	1.42	266	19.8	9.9
IFLUX-2100	1.20 (-15%)	237 (-11%)	16.3 (-18%)	8.1 (-18%)

146

147 In addition to the direct effects of climate change, increases in LNO<sub>x</sub> increase ozone and OH  
 148 production. Therefore, using the CTH scheme, the tropospheric ozone burden decreases (-  
 149 2%) much less than in the simulations without lightning (-9%), almost offsetting the direct  
 150 effects of climate change, whilst methane lifetime decreases are proportionally larger (-24%).  
 151 In contrast using the IFLUX scheme, where LNO<sub>x</sub> decreases in future, tropospheric ozone  
 152 burden decreases are larger (-11%) than in the ZERO simulations but methane lifetime  
 153 decreases are smaller (-18%). Importantly, many of these changes occur in the tropical  
 154 upper troposphere (Supplementary Figure 3), where the ozone radiative forcing efficiency is  
 155 highest.

156 The link between lightning NO<sub>x</sub> and radiative forcing from ozone has been the focus of  
 157 several studies<sup>29-31</sup>. A positive feedback has been proposed through increased lightning,  
 158 ozone and radiative forcing (RF) producing further warming, and therefore more lightning.

159 However, the long-term net cooling effect<sup>32</sup> from reduced methane driven by the increase in  
160 LNO<sub>x</sub> is often neglected.

161 We provide the first estimate of the radiative forcing of LNO<sub>x</sub> under future climate  
162 considering both ozone and methane (Fig. 3). The radiative forcing by year 2100 without  
163 lightning, where changes in atmospheric composition are due to direct effects of climate  
164 change alone, is negative for both ozone and methane, as expected from the composition  
165 changes (Table 1).

166 Importantly, the two lightning schemes have opposite effects on radiative forcing from ozone  
167 and methane, arising from the different effects on composition. The difference in ozone  
168 radiative forcing between the two schemes is 83 mW m<sup>-2</sup> (Fig. 3, Supplementary Table S3),  
169 which is approximately a third of the total ozone radiative forcing between 2000 and 2100  
170 under RCP8.5<sup>33</sup>. This total forcing is an average of multi-model projections that use the CTH  
171 scheme. Therefore, the new IFLUX scheme suggests future total ozone radiative forcing  
172 may be substantially lower than previously estimated. For methane, there is a difference of  
173 54 mW m<sup>-2</sup> between the two schemes which accounts for ±5% in the total methane radiative  
174 forcing between 2000 and 2100 under RCP8.5. We find a net positive radiative forcing with  
175 the CTH approach, permitting a positive lightning-climate feedback as previously suggested.  
176 However, there is little net radiative forcing with the IFLUX approach, and therefore the  
177 results with this scheme do not support the positive feedback argument.

178 In conclusion, we find very different impacts on atmospheric composition and radiative  
179 forcing when simulating future lightning using a cloud-ice relationship and the commonly-  
180 used cloud-top height relationship. The latter approach is less closely related to the  
181 underlying ice-graupel collisions of cloud electrification, and may underrepresent this critical  
182 component in climate change projections. Therefore, quantification of future atmospheric  
183 composition and radiative forcing of methane and ozone should account for the uncertainty  
184 in the response of lightning NO<sub>x</sub> presented here.



185 Given the disagreement between schemes in future tropical lightning, field campaigns and  
186 long-term measurement studies focusing on tropical lightning are needed. Further research  
187 is also needed to evaluate simulations of cloud ice and quantify its response to climate  
188 change, and to test how global ice-based lightning schemes such as IFLUX perform against  
189 fully explicit fine-scale models of cloud microphysics and electrification. Alongside such  
190 work, implementation of the ice flux parametrisation in other chemistry-climate models would  
191 further enhance our knowledge of the response of lightning to climate change.

## 192 **Acknowledgements**

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195 model, and Lawrence Jackson for his advice regarding the calculation of significance.

## 196 **Author contributions**

197 DLF, RMD and OW designed the study and interpreted the results with input from other co-  
198 authors. OW and DS advised on the radiative forcing analysis. DLF performed the analysis,  
199 developed the code and ran the simulations. DLF prepared the manuscript with contributions  
200 from RMD and OW; all co-authors reviewed the manuscript.

## 201 **Competing financial interests**

202 The authors declare no competing financial interests.

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288 **Table 2** Global lightning and atmospheric composition properties, simulated for present-day and future with  
289 different lightning schemes and with no lightning. Percentage changes are relative to year 2000 for each  
290 approach.

291 **Figure 1** Changes in lightning flash rate between the 2000s and 2100s using lightning schemes based on cloud  
292 ice flux (top) and cloud-top height (bottom). Hatching shows areas with no significant change at the 5% level,  
293 determined using the simulated interannual variability.

294 **Figure 2** Mean vertical distributions of meteorological variables in the 2000s (solid) and 2100s (dotted), over  
295 tropical land. Dashed lines show the 440 (black) and 390 (grey) hPa sampling levels used in the present-day and  
296 future IFLUX simulations, respectively (see Methods).

297 **Figure 3** Estimated ozone, methane and the net radiative forcing between 2000 and 2100 resulting from climate  
298 change and LNO<sub>x</sub> emissions. Triangular points are estimates using high (upward triangle) and low (downward  
299 triangle) methane feedback (see Methods).

## 300 Methods

301 *Model.* The model used in this study is the UK Chemistry and Aerosols model (UKCA)  
302 coupled to the atmosphere-only version of the UK Met Office Unified Model (UM) version 8:  
303 UM-UKCA. The atmosphere component is the Global Atmosphere 4.0 (GA4.0)<sup>34</sup>. Both  
304 tropospheric and stratospheric chemistry processes are represented. The tropospheric  
305 scheme, most relevant to this study, is described and evaluated in another study<sup>35</sup>. There  
306 are 75 species with 285 reactions that include the oxidation of methane, ethane, propane,  
307 and isoprene. The model is run at horizontal resolution N96 (1.875° longitude by 1.25°  
308 latitude). Vertically there are 85 terrain-following hybrid-height levels between the surface  
309 and 85 km. The cloud parametrisation of GA4<sup>34</sup> uses the Met Office Unified Model's  
310 prognostic cloud fraction and prognostic condensate (PC2) scheme<sup>36,37</sup> along with  
311 modifications to the cloud erosion parametrisation<sup>38</sup>. PC2 uses prognostic variables for water  
312 vapour, liquid and ice mixing ratios as well as for liquid, ice and total cloud fraction. The  
313 cloud ice variable includes snow, pristine ice and riming particles. The model used is

314 identical to that used in an evaluation of the lightning scheme for present-day<sup>21</sup>, where  
315 further details can be found. However, cloud ice observations by satellite remains highly  
316 uncertain from satellite observations and is poorly represented in models<sup>27,39,40</sup>. The global  
317 representation of cloud ice, liquid and water have been evaluated in some configurations of  
318 the Met Office Unified Model at four pressure levels, including two levels in the mid to upper  
319 troposphere (215 and 600 hPa)<sup>27</sup>. At these levels the Unified Model configurations rank in  
320 the top 3 out of 19 models. We therefore have confidence that the simulated distribution of  
321 cloud ice is a useful one.

322 *Simulation setup.* Seven simulations were performed with different lightning schemes and  
323 representing either the year 2000 or the year 2100 under Representative Concentration  
324 Pathway 8.5 (RCP8.5). Following one year of spin-up from present-day initial conditions,  
325 each simulation is performed for a further 10 years using the same driving conditions for  
326 each year. The interannual variability of the simulation is used to provide 95% confidence  
327 intervals on the decadal-average changes (see main text). In addition, when calculating the  
328 significance level for Figure 1 and Supplementary Figure 2, an adjustment has been made to  
329 the sample size to account for temporal auto-correlation of lag 1 year, though the effect of  
330 this is small. In all simulations, the chemistry scheme uses the same anthropogenic and  
331 biomass burning emissions and Greenhouse Gas (GHG) concentrations representative of  
332 the year 2000<sup>41</sup>. Well-mixed GHG concentrations in the future scenario are altered in the  
333 model radiative scheme in order to represent changes in the radiative properties of the  
334 atmosphere, and hence climate under RCP8.5. Fixed present-day climatologies of ozone  
335 and aerosol are used in the radiative scheme. Methane mixing ratios in the chemistry model  
336 are fixed at present-day levels in all simulations using a prescribed lower boundary  
337 condition. A methane radiative forcing is calculated for the simulations, and this is described  
338 in the *Radiative Forcing Calculation* section of the methods. Sea surface temperatures  
339 (SSTs) and sea ice concentrations for present-day simulations are taken from decadal  
340 average climatologies based on 1995-2004 analyses<sup>42</sup>. For SSTs and sea ice in the future

341 scenario, decadal average anomalies from the Coupled Model Intercomparison Project  
342 Phase 5 (CMIP5) HadGEM2-ES simulations for 1995-2005 and 2095-2105 were applied to  
343 the present-day SST and sea ice analysis fields. In the model, the chemistry scheme does  
344 not feed back to the radiative scheme so that all model simulations within the same time  
345 period experience the same meteorology, and consequently the same changes in surface  
346 temperature between the two time periods.

347 *The IFLUX parametrisation.* The cloud ice flux based lightning scheme was developed using  
348 meteorological variables in reanalysis data and satellite observations of lightning<sup>10</sup>. The  
349 scheme uses cloud ice flux which is related to the collision of cloud ice particles, since this is  
350 the principle component of the leading theory for thunderstorm charging, the Non-inductive  
351 Charging mechanism<sup>8</sup>. The lightning flash rate (fl.  $m_{\text{cell}}^{-2} s^{-1}$ ) is calculated with:

$$352 \quad f = A\phi,$$

353 where  $\phi$  is the upward cloud ice flux at a sampling pressure level, and A is a constant  
354 ( $6.58 \times 10^{-7}$  fl.  $kg_{\text{ice}}^{-1} m_{\text{cloud}}^2 m_{\text{cell}}^{-2}$  over land, and  $9.08 \times 10^{-8}$  fl.  $kg_{\text{ice}}^{-1} m_{\text{cloud}}^2 m_{\text{cell}}^{-2}$  over ocean).  
355 The upward cloud ice flux in the model is calculated as:

$$356 \quad \phi = \frac{q\Phi}{c},$$

357 where  $q$  is the specific cloud ice water content ( $kg_{\text{ice}} kg_{\text{air}}^{-1}$ ),  $\Phi$  is the updraught mass flux  
358 ( $kg_{\text{air}} m_{\text{cell}}^{-2} s^{-1}$ ), and  $c$  is the fractional total cloud cover ( $m_{\text{cloud}}^2 m_{\text{cell}}^{-2}$ ). All variables are grid  
359 cell mean values and are interpolated to the sampling pressure level. The grid cell mean  
360 updraught mass flux is the product of the convective updraught mass flux and the convective  
361 cloud fraction, and both of these variables are shown in Figure 2. For present-day  
362 simulations a sampling level of 440 hPa is used. This pressure level is based on the  
363 definition of deep convective clouds by the International Satellite Cloud Climatology  
364 Project<sup>43</sup>. It is noted that more detailed lightning schemes are possible in mesoscale and  
365 cloud-resolving models that resolve microphysical processes in deep convection<sup>44-48</sup>.

366 *Details of the future IFLUX sampling level calculation.* For the future climate simulations, the  
 367 sampling level,  $p_{sample}$ , is adjusted for changes in the atmospheric temperature profile. This  
 368 adjustment is made relative to two reference levels and the sampling level is calibrated to  
 369 the relative position of the 440 hPa level between these under present-day conditions. The  
 370 lower reference level is global mean pressure of the 0°C isotherm,  $\bar{p}_{0C}$ , chosen because it  
 371 marks an approximate level at which ice can begin to form (a vital process for cloud  
 372 electrification). The upper reference level is the global mean tropopause pressure,  $\bar{p}_{trop}$   
 373 (determined using a combined isentropic-dynamical approach<sup>49</sup>), which was chosen  
 374 because vertical development of clouds becomes greatly inhibited above this height. The  
 375 equations used for the calculation, with  $t=2100$  for the future time period, are:

$$p_{sample}(t) = \bar{p}_{0C}(t) - K_{2000}(\bar{p}_{0C}(t) - \bar{p}_{trop}(t))$$

376 where

$$K_{2000} = \frac{\bar{p}_{0C}(t = 2000) - 440hPa}{\bar{p}_{0C}(t = 2000) - \bar{p}_{trop}(t = 2000)} \approx \frac{1}{3}$$

377 Using this approach, a sampling level of 390 hPa is calculated for the future simulation. An  
 378 alternative upper limit of the -40°C isotherm, based on the approximate top of the mixed  
 379 phase cloud region, also suggests a future sampling level of 390 hPa. All the sampling levels  
 380 discussed above are presented in Supplementary Figure 1. Simulations using the 440 hPa  
 381 sampling level in the future have been performed in order to test sensitivity to the sampling  
 382 level, and the results of these simulations are presented in the Supplementary figures and  
 383 tables. In addition, supplementary text discusses how the sampling pressure could be  
 384 refined within transient simulations.

385 *Lightning NO<sub>x</sub> scheme.* The lightning parametrisations provide the lightning flash rate. Each  
 386 flash corresponds to a NO emission of ~250 mol(NO)<sup>1,21</sup>. There is a total global present-day  
 387 emission of ~5 TgN using both lightning schemes<sup>1,21</sup>. The LNO<sub>x</sub> is distributed vertically  
 388 based upon prescribed vertical profiles<sup>50</sup> between the surface and the cloud top. Both  
 389 lightning schemes are normalised to give a global annual average of 46 flashes s<sup>-1</sup> (or 1.45 x



390  $10^9 \text{ fl. yr}^{-1}$ )<sup>51</sup> in a one-year present-day simulation, using factors of 1.57 for the CTH scheme  
391 and 1.11 for the IFLUX scheme. The same factors are used in the future climate change  
392 simulations but the global annual flash rate changes in response to the changing  
393 meteorology.

394 *Radiative forcing calculation.* With a fixed lower boundary condition for methane, the  
395 methane mixing ratio is heavily constrained and there is little adjustment to the oxidation rate  
396 as the OH concentration is modified by changes in climate and LNO<sub>x</sub>. The equilibrium  
397 methane mixing ratio can be calculated using the change in methane lifetime and a feedback  
398 factor which is typically around 1.30 in models<sup>33</sup> with a range in the literature of 1.19 to  
399 1.53<sup>7,33,52,53</sup>. This equilibrium methane mixing ratio is then used to determine the methane  
400 radiative forcing (RF)<sup>54</sup>. For ozone, the short-term radiative forcing is calculated using the  
401 differences in the annual mean spatial distribution of the tropospheric ozone column  
402 between each simulation and CTH in year 2000. These differences are multiplied by the  
403 horizontal spatial distribution of the radiative forcing efficiency of ozone ( $\text{mW m}^{-2} \text{ DU}^{-1}$ ), using  
404 a multi-model average spatial distribution from the Atmospheric Chemistry and Climate  
405 Model Intercomparison Project (ACCMIP) study<sup>33</sup>. The area-weighted mean over the  
406 distribution provides the global short-term ozone radiative forcing. As ozone is also  
407 influenced by changes in methane, there is an additional long-term ozone radiative forcing  
408 resulting from the equilibrium methane change. The tropospheric ozone response to a 20%  
409 reduction in methane from present-day levels across a range of models contributing to the  
410 Task Force on Hemispheric Transport of Air Pollution studies is  $0.95 \pm 0.25 \text{ DU}$ . This range is  
411 used to estimate the long-term ozone change associated with the inferred methane change,  
412 accounting for the non-linear response of ozone to methane changes<sup>55</sup>. The long-term ozone  
413 RF is calculated from this change, and the combined long and short term ozone radiative  
414 forcings provide the total ozone radiative forcing. The ozone and methane radiative forcings  
415 presented therefore correspond to radiative forcings after atmospheric composition has fully  
416 equilibrated with the perturbed LNO<sub>x</sub>. Parameter uncertainty in the RF estimate is

417 represented using three sets of parameters that represent a typical, low and high sensitivity  
418 to methane change, and are shown as bars, downward triangles and upward triangles in  
419 Figure 3. To isolate the effects of LNO<sub>x</sub>, we subtracted the radiative forcing in the absence of  
420 lightning (ZERO simulations) from that with each lightning scheme. The results of this are  
421 shown in Figure 3, while the original radiative forcing values for each set of simulations is  
422 given in Supplementary table 3.

423 *Data availability.* The data that support the findings of this study are available from the  
424 corresponding author upon request.

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