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A projected decrease in lightning under climate change 1 Declan L. Finney^{1,*}, Ruth M. Doherty¹, Oliver Wild², David S. Stevenson¹, Ian A. 2 MacKenzie¹ and Alan M. Blyth³ 3 ¹ School of GeoSciences, The University of Edinburgh, Edinburgh, UK 4 5 ² Lancaster Environment Centre, Lancaster University, Lancaster, UK ³ National Centre for Atmospheric Science, University of Leeds, Leeds, UK 6 * now at: Institute for Climate and Atmospheric Science, University of Leeds, Leeds, UK 7 8 Correspondence to: D. L. Finney d.l.finney@leeds.ac.uk 9 Lightning strongly influences atmospheric chemistry^{1–3}, and impacts the frequency of 10 natural wildfires⁴. Most previous studies project an increase in global lightning with 11 climate change over the coming century^{1,5–7}, but these typically use parametrisations 12 13 of lightning that neglect cloud ice fluxes, a component generally considered to be fundamental to thunderstorm charging⁸. As such, the response of lightning to climate 14 change is uncertain. Here, we compare lightning projections for 2100 using two 15 parametrisations: the widely-used cloud-top height (CTH) approach⁹, and a new 16 upward cloud ice flux (IFLUX) approach¹⁰ that overcomes previous limitations. In 17 18 contrast to the previously reported global increase in lightning based on CTH, we find a 15% decrease in total lightning flash rate with IFLUX in 2100 under a strong global 19 warming scenario. Differences are largest in the tropics, where most lightning occurs, 20 21 with implications for the estimation of future changes in tropospheric ozone and methane, as well as differences in their radiative forcings. These results suggest that 22 lightning schemes more closely related to cloud-ice and microphysical processes are 23 24 needed to robustly estimate future changes in lightning and atmospheric 25 composition. Changes in climate over the next century are expected to alter atmospheric temperature, 26 humidity, stability and dynamics¹¹. The leading theory for electrical charge generation in 27 thunderstorms^{8,12} suggests that the occurrence of lightning depends on all these factors, 28

through their effect on convection and colliding ice and graupel particles. Lightning is an

important source of nitric oxide (NO), a precursor of ozone and the hydroxyl radical (OH)
which governs the lifetime of greenhouse gases such as methane¹. Both ozone and
methane are important greenhouse gases, and changes in their concentrations can lead to a
warming or cooling of the atmosphere. Thus, lightning needs to be represented in chemistryclimate models to fully simulate the interactions and feedbacks between atmospheric
composition and climate change. Future changes in lightning are also of importance for
aerosol chemistry^{2,3}, wild-fire ignition⁴, and damage to infrastructure and to human health.

Recent studies^{4,6,7,13,14} simulating future lightning over the next century with the CTH 37 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 become highly uncertain on longer time scales^{15,16}. An alternative⁶ to the CTH scheme, using 41 42 cold cloud depth to parametrise lightning, suggested a smaller increase in lightning under climate change of ~4% K⁻¹. Furthermore, a decrease in future lightning has been found using 43 a convective mass flux-based lightning scheme¹⁷ in two recent studies^{6,18}. However, this 44 scheme has been found to perform poorly against observations^{6,10,19}. 45

Only one study to date has used a lightning scheme dependent on cloud ice particles to project future lightning. This study found a decrease in lightning associated with an increase in temperature²⁰. However, the study had a near-term focus on 2030 and the global surface temperature increase was less than 0.2K. It is not clear whether a similar response occurs for larger changes in temperature at the end of the century²⁰.

In this study we use both the established CTH scheme⁹ and the recently developed and evaluated IFLUX scheme¹⁰ (see Methods) in a chemistry-climate model to simulate future lightning and its influence on atmospheric composition and radiative forcing. Atmospheric dynamics are decoupled from changes in atmospheric composition so that both lightning schemes use the same underlying meteorology. With the same model as used here, the IFLUX scheme has shown a more realistic representation of present-day global lightning and

tropospheric ozone than the CTH approach, especially in the tropics^{10,21}. For instance, the 57 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 scheme²¹. Whilst accurately representing present-day lightning does not guarantee that 62 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).

Lightning NO_x emissions (LNO_x) from existing lightning parametrisations scale linearly with changes in global mean surface temperature across chemistry-climate models¹⁸. Therefore, we use the year 2100 under Representative Concentration Pathway 8.5 (RCP8.5)²² to obtain a clear lightning response to substantial climate change. We provide the first estimate of the future lightning response to long-term global warming in 2100 using a cloud ice-based lightning parametrisation, and compare with results from the widely-used CTH scheme.

We simulate a decrease in global total lightning of 2.2 (1.9-2.5) x10⁸ fl. yr⁻¹ by 2100 with the
IFLUX scheme (Table 2 Global lightning and atmospheric composition properties, simulated
for present-day and future with different lightning schemes and with no lightning. Percentage
changes are relative to year 2000 for each approach.

Figure 1), where the range is the 95% confidence interval based on the simulated
interannual variability. A sensitivity test diagnosing the ice flux at the same level as under
present-day climate shows a decrease of 5.8 (5.6-6.1) x10⁸ fl. yr⁻¹ (Supplementary Figure
2b), suggesting that the choice of level does not influence our conclusions. With the CTH
scheme, we simulate an increase in the global flash rate of 6.1 (5.9-6.2) x10⁸ fl. yr⁻¹ in year
2100.

Regionally, the IFLUX and CTH approaches result in increases in total lightning over the 84 USA of 3.4 and 14.2 %K⁻¹, respectively. Assuming total and cloud-to-ground lightning 85 86 respond similarly to climate change, our results are consistent with a recent study that used convective available potential energy and precipitation to parametrise lightning²³. In that 87 study it was estimated that *cloud-to-ground* lightning over the USA would increase by 12 %K⁻ 88 ¹ under RCP8.5²³ (with a range of 3-18%K⁻¹ across models). However, this increase does not 89 apply to all mid-latitude locations. For instance, we find no significant change over most of 90 91 Europe with either lightning scheme.

92 Whilst several studies have considered how climate variability, such as El Niño driven events, affects tropical lightning^{15,24}, the impact of climate change on tropical lightning has 93 not received much attention. This is despite ~80% of global lightning flashes occurring in the 94 tropics and subtropics²⁵. With the IFLUX approach, a decrease in tropical lightning is 95 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.

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 increase in tropopause height, a robust feature of global warming²⁶. A reduction in cloud ice 114 115 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 Change (IPCC)¹¹ reports a projected future decrease in mean updraught mass flux 117 118 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 instance, many models underestimate tropical cloud ice content²⁷. The formation of cloud ice 122 123 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.

Most studies report future increases in LNO_x, as they employ the CTH scheme. One

129 previous study found these future increases in LNO_x more than offset the reduction in

130 tropospheric ozone arising from lower anthropogenic emissions of NO_x and other ozone

131 precursors⁷. In our study, other ozone precursor emissions are kept constant so that

132 changes in ozone burden due to changes in climate and LNO_x can be quantified. In our

model, each lightning flash produces 250 mol of NO. Given a present-day lightning emission

134 of ~5 TgN yr⁻¹, the responses to climate change using the IFLUX and CTH schemes are -

135 0.15 TgN K⁻¹ and +0.44 TgN K⁻¹, respectively. The CTH response closely matches results

136 from a recent multi-model intercomparison of 10 models using the CTH scheme¹⁸.

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 28 ,

141 mainly via the increase in OH radicals through reaction of water with O(¹D). The OH also

acts as a sink for methane, reducing the methane lifetime.

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

Simulation	Global lightning (x10 ⁹ fl. yr ⁻¹)	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_x 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_x 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_x and radiative forcing from ozone has been the focus of

157 several studies^{29–31}. A positive feedback has been proposed through increased lightning,

ozone and radiative forcing (RF) producing further warming, and therefore more lightning.

However, the long-term net cooling effect³² from reduced methane driven by the increase in LNO_x is often neglected.

We provide the first estimate of the radiative forcing of LNO_x under future climate considering both ozone and methane (Fig. 3). The radiative forcing by year 2100 without lightning, where changes in atmospheric composition are due to direct effects of climate change alone, is negative for both ozone and methane, as expected from the composition 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 radiative forcing between the two schemes is 83 mW m⁻² (Fig. 3, Supplementary Table S3), 168 169 which is approximately a third of the total ozone radiative forcing between 2000 and 2100 under RCP8.5³³. This total forcing is an average of multi-model projections that use the CTH 170 171 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 54 mW m⁻² between the two schemes which accounts for ±5% in the total methane radiative 173 174 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

forcing when simulating future lightning using a cloud-ice relationship and the commonlyused cloud-top height relationship. The latter approach is less closely related to the underlying ice-graupel collisions of cloud electrification, and may underrepresent this critical component in climate change projections. Therefore, quantification of future atmospheric composition and radiative forcing of methane and ozone should account for the uncertainty in the response of lightning NO_x presented here.

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

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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-
- authors. OW and DS advised on the radiative forcing analysis. DLF performed the analysis,
- 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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287 List of Tables/Figures

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

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

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

Figure 3 Estimated ozone, methane and the net radiative forcing between 2000 and 2100 resulting from climate
 change and LNOx emissions. Triangular points are estimates using high (upward triangle) and low (downward
 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)³⁴. Both
- 304 tropospheric and stratospheric chemistry processes are represented. The tropospheric
- 305 scheme, most relevant to this study, is described and evaluated in another study³⁵. There
- are 75 species with 285 reactions that include the oxidation of methane, ethane, propane,
- and isoprene. The model is run at horizontal resolution N96 (1.875^o longitude by 1.25^o
- 308 latitude). Vertically there are 85 terrain-following hybrid-height levels between the surface
- and 85 km. The cloud parametrisation of GA4³⁴ uses the Met Office Unified Model's
- prognostic cloud fraction and prognostic condensate (PC2) scheme^{36,37} along with
- 311 modifications to the cloud erosion parametrisation³⁸. 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

identical to that used in an evaluation of the lightning scheme for present-day²¹, where 314 315 further details can be found. However, cloud ice observations by satellite remains highly uncertain from satellite observations and is poorly represented in models^{27,39,40}. The global 316 317 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 troposphere (215 and 600 hPa)²⁷. At these levels the Unified Model configurations rank in 319 the top 3 out of 19 models. We therefore have confidence that the simulated distribution of 320 321 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 the year 2000⁴¹. Well-mixed GHG concentrations in the future scenario are altered in the 332 333 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 average climatologies based on 1995-2004 analyses⁴². For SSTs and sea ice in the future 340

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.

The *IFLUX parametrisation*. The cloud ice flux based lightning scheme was developed using meteorological variables in reanalysis data and satellite observations of lightning¹⁰. The scheme uses cloud ice flux which is related to the collision of cloud ice particles, since this is the principle component of the leading theory for thunderstorm charging, the Non-inductive Charging mechanism⁸. The lightning flash rate (fl. m_{cell}⁻² s⁻¹) is calculated with:

352 $f = A\phi$,

where ϕ is the upward cloud ice flux at a sampling pressure level, and A is a constant (6.58x10⁻⁷ fl. kg_{ice}⁻¹ m_{cloud}² m_{cell}⁻² over land, and 9.08x10⁻⁸ fl. kg_{ice}⁻¹ m_{cloud}² m_{cell}⁻² over ocean). The upward cloud ice flux in the model is calculated as:

$$356 \qquad \phi = \frac{q\phi}{c},$$

where q is the specific cloud ice water content (kg_{ice} kg_{air}⁻¹), Φ is the updraught mass flux 357 (kg_{air} m_{cell}⁻² s⁻¹), and c is the fractional total cloud cover (m_{cloud}² m_{cell}⁻²). All variables are grid 358 359 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 Project⁴³. It is noted that more detailed lightning schemes are possible in mesoscale and 364 cloud-resolving models that resolve microphysical processes in deep convection^{44–48}. 365

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 adjustment is made relative to two reference levels and the sampling level is calibrated to 368 369 the relative position of the 440 hPa level between these under present-day conditions. The lower reference level is global mean pressure of the 0°C isotherm, \overline{p}_{0C} , chosen because it 370 371 marks an approximate level at which ice can begin to form (a vital process for cloud electrification). The upper reference level is the global mean tropopause pressure, \overline{p}_{trop} 372 (determined using a combined isentropic-dynamical approach⁴⁹), which was chosen 373 374 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:

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

376 where

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

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.

Lightning NO_x scheme. The lightning parametrisations provide the lightning flash rate. Each flash corresponds to a NO emission of ~250 mol(NO)^{1,21}. There is a total global present-day emission of ~5 TgN using both lightning schemes^{1,21}. The LNO_x is distributed vertically based upon prescribed vertical profiles⁵⁰ between the surface and the cloud top. Both lightning schemes are normalised to give a global annual average of 46 flashes s⁻¹ (or 1.45 x

10⁹ fl. yr⁻¹)⁵¹ in a one-year present-day simulation, using factors of 1.57 for the CTH scheme
and 1.11 for the IFLUX scheme. The same factors are used in the future climate change
simulations but the global annual flash rate changes in response to the changing
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_x. The equilibrium 397 methane mixing ratio can be calculated using the change in methane lifetime and a feedback factor which is typically around 1.30 in models³³ with a range in the literature of 1.19 to 398 1.53^{7,33,52,53}. This equilibrium methane mixing ratio is then used to determine the methane 399 400 radiative forcing (RF)⁵⁴. 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 horizontal spatial distribution of the radiative forcing efficiency of ozone (mW m⁻² DU⁻¹), using 403 404 a multi-model average spatial distribution from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) study³³. The area-weighted mean over the 405 406 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±0.25 DU. This range is 411 used to estimate the long-term ozone change associated with the inferred methane change. accounting for the non-linear response of ozone to methane changes⁵⁵. The long-term ozone 412 413 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 equilibrated with the perturbed LNO_x. Parameter uncertainty in the RF estimate is 416

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_x , 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	2 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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