

1 Historical (1960–2014) lightning and LNO_x trends and their 2 controlling factors in a chemistry–climate model

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7 **Abstract.** Lightning can cause natural hazards that result in human and animal injuries or fatalities, infrastructure destruction,
8 and wildfire ignition. Lightning-produced NO_x (LNO_x), a major NO_x (NO_x=NO+NO₂) source, plays a vital role in
9 atmospheric chemistry and global climate. The Earth has experienced marked global warming and changes in aerosol and
10 aerosol precursor emissions (AeroPEs) since the 1960s. Investigating long-term historical (1960–2014) lightning and LNO_x
11 trends can provide important indicators for all lightning-related phenomena and for LNO_x effects on atmospheric chemistry
12 and global climate. Understanding how global warming and changes in AeroPEs influence historical lightning–LNO_x trends
13 can be helpful in providing a scientific basis for assessing future lightning–LNO_x trends. Moreover, global lightning
14 activities' responses to large volcanic eruptions such as the 1991 Pinatubo eruption are not well elucidated, and are worth
15 exploring. This study employed the widely used cloud top height lightning scheme (CTH scheme) and the newly developed
16 ice-based ECMWF-McCAUL lightning scheme to investigate historical (1960–2014) lightning–LNO_x trends and variations
17 and their influencing factors (global warming, increases in AeroPEs, and Pinatubo eruption) in the framework of the
18 CHASER (MIROC) chemistry–climate model. Results of sensitivity experiments indicate that both lightning schemes
19 simulated almost flat global mean lightning flash rate anomaly trends during 1960–2014 in CHASER (Mann-Kendall trend
20 test (significance inferred as 5%) shows no trend for the ECMWF-McCAUL scheme, but a 0.03 % yr⁻¹ significant increasing
21 trend is detected for the CTH scheme). Moreover, both lightning schemes suggest that past global warming enhances
22 historical trends of global mean lightning density and global LNO_x emissions in a positive direction (around 0.03% yr⁻¹ or 3%
23 K⁻¹). However, past increases in AeroPEs exert an opposite effect to the lightning–LNO_x trends (-0.07% yr⁻¹ – -0.04% yr⁻¹
24 for lightning and -0.08% yr⁻¹ – -0.03% yr⁻¹ for LNO_x) when one considers only the aerosol radiative effects in the cumulus
25 convection scheme. Additionally, effects of past global warming and increases in AeroPEs on lightning trends were found to
26 be heterogeneous across different regions when analyzing lightning trends on the global map. Lastly, this report is the first of
27 study results suggesting that global lightning activities were suppressed markedly during the first year after the Pinatubo
28 eruption shown in both lightning schemes (global lightning activities decreased by as much as 18.10% simulated by the
29 ECMWF-McCAUL scheme). Based on simulated suppressed lightning activities after the Pinatubo eruption, findings also
30 indicate that global LNO_x emissions decreased after the 2–3-year Pinatubo eruption (1.99%–8.47% for the annual percentage
31 reduction). Model intercomparisons of lightning flash rate trends and variations between our study (CHASER) and other

32 Coupled Model Intercomparison Project Phase 6 (CMIP6) models indicate great uncertainties in historical (1960–2014)
33 global lightning trend simulations. Such uncertainties must be investigated further.

34 **1 Introduction**

35 Lightning, an extremely energetic natural phenomenon, occurs at every moment somewhere on Earth: its average occurrence
36 frequency is approximately 46 times per second (Cecil et al., 2014). Lightning generation is associated with electric charge
37 separation, which is mainly realized by collisions between graupel and hail and hydrometeors of other types within
38 convective clouds (Lopez, 2016). As a natural hazard, lightning can cause human and animal injuries and fatalities,
39 infrastructure destruction, and wildfire ignition (Cerveny et al., 2017; Cooper and Holle, 2019; Jensen et al., 2022;
40 Veraverbeke et al., 2022). Lightning-produced NO_x (LNO_x) accounts for around 10% of the global tropospheric NO_x
41 (NO_x=NO+NO₂) source. It is regarded as the dominant NO_x source in the middle to upper troposphere (Schumann and
42 Huntrieser, 2007; Finney et al., 2016b). Moreover, LNO_x plays a crucially important role in atmospheric chemistry and
43 global climate by affecting the abundances of OH radical, important greenhouse gases (GHGs) such as ozone and methane,
44 and other trace gases (Labrador et al., 2005; Schumann and Huntrieser, 2007; Wild, 2007; Liaskos et al., 2015; Finney et al.,
45 2016a; Murray, 2016; Tost, 2017; He et al., 2022b).

46

47 Reportedly, the lightning flash rate (LFR) is related to the stage of convective cloud development (Williams et al., 1989),
48 Convective Available Potential Energy (CAPE) (Romps et al., 2014), cloud liquid–ice water content (Saunders et al., 1991;
49 Finney et al., 2014) and even to the convective precipitation volume (Goodman et al., 1990; McCaul et al., 2009; Romps et
50 al., 2014). Long-term global warming is associated with changes in the overall temperature and relative humidity profiles in
51 the atmosphere and global convective adjustment (Manabe and Wetherald, 1975; Del Genio et al., 2007), which can strongly
52 affect the lightning-related factors described above. Consequently, long-term global warming can be a fundamentally
53 important factor affecting long-term variations in global lightning activity. Findings from many earlier numerical simulation
54 studies manifest that global lightning activities are sensitive to long-term global warming, with most studies showing 5–16%
55 (average around 10%) increases in global lightning activities per 1 K global warming (Price and Rind, 1994; Zeng et al.,
56 2008; Hui and Hong, 2013; Banerjee et al., 2014; Krause et al., 2014; Romps et al., 2014; Clark et al., 2017). However, other
57 numerical simulation studies such as those using an ice-based lightning scheme or convective mass flux as a proxy to
58 parameterize lightning have yielded opposite results, suggesting that global lightning activity will decrease under long-term
59 global warming (Clark et al., 2017; Finney et al., 2018).

60

61 Aside from long-term global warming, changes in aerosol loading can also be responsible for long-term global lightning
62 activity variations. Aerosols influence lightning activity through aerosol radiative and microphysical effects, but the degree
63 to which the two distinct effects influence regional or global scale lightning activities remains unclear (Yuan et al., 2011;

64 Yang et al., 2013; Tan et al., 2016; Altaratz et al., 2017; Wang et al., 2018; Liu et al., 2020). Further research is needed. It is
65 urgently necessary to elucidate the effects of aerosol radiative and microphysical effects on lightning on a global scale. The
66 aerosol radiative effects indicate that aerosols can heat the atmospheric layer and can cool the Earth's surface by absorbing
67 and scattering solar radiation (Kaufman et al., 2002; Koren et al., 2004, 2008; Li et al., 2017). Thereby, convection and
68 electrical activities are likely to be inhibited (Koren et al., 2004; Yang et al., 2013; Tan et al., 2016). The microphysical
69 effects suggest that by acting as cloud condensation nuclei (CCN) or as ice nuclei, aerosols can reduce the mean size of
70 cloud droplets, consequently suppressing the coalescence of cloud droplets into raindrops. As a result, more liquid water
71 particles are uplifted to higher mixed-phase regions of the troposphere, where they invigorate lightning (Wang et al., 2018;
72 Liu et al., 2020).

73

74 The Earth has experienced a considerable degree of global warming and changes in AeroPEs since the 1960s (Hoesly et al.,
75 2018; Climate at a Glance | National Centers for Environmental Information (NCEI), 2022). However, how historical
76 lightning has trended and how lightning has responded to historical global warming and changes in AeroPEs are not well
77 examined. This topic is worth exploring because historical lightning densities are indicators for all lightning-related
78 phenomena (Price and Rind, 1994). Exploring the historical global LNO_x emission trend is also meaningful because it can
79 indicate the effects of LNO_x emissions on atmospheric chemistry and global climate. Furthermore, investigating the effects
80 of historical global warming and increases in AeroPEs on historical lightning– LNO_x trends can provide a basis for assessing
81 future lightning– LNO_x trends.

82

83 Large-scale volcanic eruptions such as the 1991 Pinatubo eruption inject tremendous amounts of sulfuric gas into the
84 stratosphere, where it converts to H_2SO_4 aerosols. Consequently, the stratospheric aerosols have increased in abundance after
85 the volcanic eruptions. The enhanced stratospheric aerosol layer can cool the Earth's surface heterogeneously and can
86 decrease the total amount of water in the atmosphere (Soden et al., 2002; Boucher, 2015, p.63). The near-global
87 perturbations in the radiative energy balance and meteorological fields caused by such strong volcanic eruptions might
88 influence global lightning activities. If so, there might be ramifications for all lightning-related phenomena. Nevertheless,
89 they remain poorly understood.

90

91 In our earlier work, we developed a new process and ice-based lightning scheme called the ECMWF-McCAUL scheme (He
92 et al., 2022b). This lightning scheme was developed by combining benefits of the lightning scheme used in the European
93 Centre for Medium-Range Weather Forecasts (ECMWF) forecasting system (Lopez, 2016) and those presented in reports by
94 McCaul et al. (McCaul et al., 2009). The ECMWF-McCAUL scheme simulated the best lightning density spatial
95 distributions among four existing lightning schemes when compared against satellite lightning observations (Lightning
96 Imaging Sensor (LIS) and Optical Transient Detector (OTD)) during 2007–2011. The sensitivity of global lightning activity

97 to changes in surface temperature on a decadal timescale was estimated as $10.13\% \text{ K}^{-1}$ using the ECMWF-McCAUL scheme
98 (He et al., 2022b), which is close to most past estimates (average around $10\% \text{ K}^{-1}$).
99

100 Using a chemistry–climate model CHASER (MIROC) with two lightning schemes (the widely used cloud top height scheme
101 and the ice-based ECMWF-McCAUL scheme), we investigated historical lightning– LNO_x trends quantitatively and
102 ascertained how global warming, increases in AeroPEs, and the Pinatubo eruption respectively influenced them. Using two
103 lightning schemes, we demonstrated the sensitivities of different lightning schemes to historical global warming, increases in
104 AeroPEs, and the Pinatubo eruption.
105

106 Research methods including the model description and experiment setup, are described in Sect. 2. In Sect. 3.1, the simulated
107 historical lightning distributions and trends are validated using LIS/OTD lightning observations. Section 3.2 presents the
108 effects of global warming and increases in AeroPEs on historical lightning– LNO_x trends. In Sect. 3.3, the Pinatubo volcanic
109 eruption effects on historical lightning– LNO_x trends are discussed. Section 3.4 elucidated model intercomparisons of LFR
110 trends and variation between our study (CHASER) and other CMIP6 model outputs. Section 4 presents relevant discussions
111 and conclusions based on these study findings.

112 **2 Method**

113 **2.1 Chemistry–climate model**

114 We used the CHASER (MIROC) global chemistry–climate model (Sudo et al., 2002; Sudo and Akimoto, 2007; Watanabe et
115 al., 2011; Ha et al., 2021) for this study, which incorporated consideration of detailed chemical and physical processes in the
116 troposphere and stratosphere. The CHASER version adopted for this study simulates the distributions of 94 chemical species
117 while reflecting the effects of 269 chemical reactions (58 photolytic, 190 kinetic, and 21 heterogeneous). As processes
118 associated with tropospheric chemistry, Non-Methane Hydrocarbons (NMHC) oxidation and the fundamental chemical cycle
119 of $\text{O}_x\text{--NO}_x\text{--HO}_x\text{--CH}_4\text{--CO}$ are considered. CHASER simulates stratospheric chemistry involving the Chapman mechanisms
120 and catalytic reactions associated with HO_x , NO_x , ClO_x , and BrO_x . Moreover, it simulates the formation of polar
121 stratospheric clouds (PSCs) and heterogeneous reactions occurring on their surfaces. CHASER is on-line-coupled to MIROC
122 AGCM ver. 5.0 (Watanabe et al., 2011), which simulates cumulus convection (Arakawa–Schubert scheme) and grid-scale
123 large-scale condensation to represent cloud and precipitation processes. The radiation flux is calculated using a two-stream k
124 distribution radiation scheme, which considers absorption, scattering, and emissions by aerosol and cloud particles as well as
125 by gaseous species (Sekiguchi and Nakajima, 2008; Goto et al., 2015). The aerosol component in CHASER is coupled with
126 the SPRINTARS aerosol model (Takemura et al., 2009), particularly for simulating primary organic carbon, sea-salt, and
127 dust, which is also based on MIROC. The aerosol radiation effects are considered in both large-scale condensation and

128 cumulus convection schemes, although the aerosol microphysical effects are only reflected in the large-scale condensation
129 scheme.

130

131 This study used a horizontal resolution of T42 ($2.8^\circ \times 2.8^\circ$), with vertical resolution of 36 σ -p hybrid levels from the surface
132 to approximately 50 km. Anthropogenic and biomass burning emissions were obtained from the CMIP6 forcing datasets
133 (van Marle et al., 2017; Hoesly et al., 2018) for 1959–2014 (<https://esgf-node.llnl.gov/search/input4mips/>, last access: 19
134 September 2022). Interannual variation in biogenic emissions for isoprene, monoterpene, acetone, and methanol were
135 considered using an off-line simulation by the Vegetation Integrative Simulator for Trace Gases (VISIT) terrestrial
136 ecosystem model (Ito and Inatomi, 2012). The residual biogenic emissions (ethane, propane, ethylene, propene) used are
137 climatological values derived from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) modeling system
138 (Guenther et al., 2012).

139

140 The CHASER (MIROC) global chemistry–climate model originally parameterizes lightning with the widely used cloud top
141 height scheme (Price and Rind, 1992). A newly developed ice-based lightning scheme called the ECMWF-McCAUL here
142 had been implemented into CHASER (MIROC) (He et al., 2022b). The ECMWF-McCAUL scheme computes LFRs as a
143 function of CAPE and Q_{Ra} (Q_{Ra} represents the total volumetric amount of cloud ice, graupel, and snow in the charge
144 separation region). Compared with the cloud top height, a salient advantage of the ECMWF-McCAUL scheme is that it has a
145 direct physical link with the charging mechanism.

146 **2.2 Lightning NO_x emission parameterizations**

147 We tested two lightning schemes for this study. The first lightning scheme is the widely used cloud top height (CTH) scheme
148 (Price and Rind, 1992), which was used originally in CHASER (MIROC). This lightning scheme uses the following
149 equations to calculate LFR.

$$150 \quad F_l = 3.44 \times 10^{-5} H^{4.9} \quad (1)$$

$$151 \quad F_o = 6.2 \times 10^{-4} H^{1.73} \quad (2)$$

152 Therein, F represents the total flash frequency (fl. min⁻¹), H stands for the cloud-top height (km), and subscripts l and o
153 respectively denote the land and ocean (Price and Rind, 1992). Actually, we realize the CTH scheme in CHASER using the
154 following equations (Eq. (3) and Eq. (4)) (Sudo et al., 2002). Each model layer's cumulus cloud fractions are used to weight
155 the calculated lightning densities from that layer in the CTH scheme.

$$156 \quad F_l = \sum_{i=1}^{n=36} adj_factor \times Cu_CF_i \times (H_i - H_{surface})^{4.9} \quad (3)$$

$$157 \quad F_o = \sum_{i=1}^{n=36} adj_factor \times Cu_CF_i \times (H_i - H_{surface})^{1.73} \quad (4)$$

158 In those equations, i represents the model layer index. In addition, adj_factor represents adjustment factors that differ for
159 different model layers and model grids. Cu_CF_i symbolizes the cumulus cloud fraction at model layer i . H_i and $H_{surface}$
160 respectively denote the altitude of model layer i and the altitude of the model's surface layer.

161

162 The second lightning scheme used for this study is a newly developed one named the ECMWF-McCAUL scheme (He et al.,
163 2022b), which is based on the original ECMWF scheme and findings reported by McCaul et al. (2009). The ECMWF-
164 McCaul scheme calculates LFRs as a function of $CAPE$ ($\text{m}^2 \text{ s}^{-2}$) and Q_{Ra} (Q_{Ra} symbolizes the total volumetric amount of
165 cloud ice, graupel, and snow in the charge separation region) as

166
$$f_l = \alpha_l Q_{Ra} CAPE^{1.3} \quad (5)$$

167
$$f_o = \alpha_o Q_{Ra} CAPE^{1.3} \quad (6)$$

168 where f_l and f_o respectively symbolize the total flash density ($\text{fl. m}^{-2} \text{ s}^{-1}$) over land and ocean. In addition, α_l and α_o are
169 constants ($\text{fl. s}^{1.6} \text{ kg}^{-1} \text{ m}^{-2.6}$) determined after calibration against LIS/OTD climatology, respectively, for land and ocean.

170 For this study, α_l and α_o are set respectively as 2.67×10^{-16} and 1.68×10^{-17} . In the charge separation region (from 0° to
171 -25°C isotherm), Q_{Ra} (kg m^{-2}) is expressed as a proxy for the charging rate because of collisions between graupel and
172 hydrometeors of other types (McCaul et al., 2009). Moreover, Q_{Ra} represents the total volumetric amount of hydrometeors of
173 three kinds (graupel, snow, and cloud ice) within the charge separation region, calculated as

174
$$Q_{Ra} = \int_{z_0}^{Z-25} (q_{graup} + q_{snow} + q_{ice}) \bar{\rho} dz, \quad (7)$$

175 where q_{graup} , q_{snow} , and q_{ice} respectively represent the mass mixing ratios (kg kg^{-1}) of graupel, snow, and cloud ice. In
176 addition, q_{ice} was diagnosed using Arakawa–Schubert cumulus parameterization. Then, q_{graup} and q_{snow} were computed at
177 each vertical level of the model using the following equations.

178
$$q_{graup} = \beta \frac{P_f}{\bar{\rho} V_{graup}} \quad (8)$$

179
$$q_{snow} = (1 - \beta) \frac{P_f}{\bar{\rho} V_{snow}} \quad (9)$$

180 In those equations, P_f represents the vertical profile of the frozen precipitation convective flux ($\text{kg m}^{-2} \text{ s}^{-1}$), $\bar{\rho}$ denotes the
181 air density (kg m^{-3}), and V_{graup} and V_{snow} respectively express the typical fall speeds for graupel and snow set to 3.1 and 0.5
182 m s^{-1} for this study. For land, the dimensionless coefficient β is set as 0.7, whereas it is set to 0.45 for oceans to consider
183 the observed lower graupel content over the oceans.

184

185 Based on the cold cloud depth, a fourth-order polynomial (equation 10) is used to calculate the proportion of total flashes
186 that are cloud-to-ground (p). An earlier report of the literature describes the method (Price and Rind, 1993).

187
$$p = \frac{1}{64.9 - 36.54D + 7.493D^2 - 0.648D^3 + 0.021D^4} \quad (10)$$

188 The depth of the cloud above the 0°C isotherms is represented by D (km) in that equation.

189

190 According to recent studies, the intra-cloud (IC) lightning flashes are as efficient as cloud-to-ground (CG) lightning flashes
 191 at producing NO_x. The lightning NO_x production efficiency is estimated as 100–400 mol per flash (Ridley et al., 2005;
 192 Cooray et al., 2009; Ott et al., 2010; Allen et al., 2019). The LNO_x production efficiencies for IC and CG are therefore set to
 193 the same value (250 mol per flash) in CHASER, which is the median of the commonly cited range of 100–400 mol per flash.
 194 Therefore, in this study, the distinctions between IC and CG do not affect the distribution or magnitude of LNO_x emissions.
 195 It is noteworthy that marked uncertainties are involved in ascertaining the LNO_x production efficiency (Allen et al., 2019;
 196 Bucsela et al., 2019). The choice of a different LNO_x production efficiency might affect the simulation of LNO_x emissions.
 197 Further research must be undertaken to implement and validate a more sophisticated parameterization of LNO_x production
 198 efficiency in chemistry–climate models. The calculated total column LNO_x for each grid was distributed into each model
 199 layer based on a prescribed “backward C-shaped” LNO_x vertical profile (Ott et al., 2010).

200 **2.3 Lightning observation data for model evaluation**

201 We used LIS/OTD gridded climatology datasets for this study, consisting of climatologies of total LFRs observed using the
 202 Lightning Imaging Sensor (LIS) and Optical Transient Detector (OTD). The OTD aboard the MicroLab-1 satellite and LIS
 203 aboard the Tropical Rainfall Measuring Mission (TRMM) satellite (Cecil et al., 2014). Both sensors detected lightning by
 204 monitoring pulses of illumination produced by lightning in the 777.4 nm atomic oxygen multiplet above background levels.
 205 In low Earth orbit, both sensors viewed Earth locations for approximately 3 min during the pass of the OTD or 1.5 min
 206 during passing of the LIS. Each day, OTD and LIS respectively orbited the globe 14 times and 16 times. OTD observed data
 207 between +75 and -75° latitude during May 1995 – March 2000, whereas LIS collected data between +38 and -38° latitude
 208 during January 1998 – April 2015. This study uses the LIS/OTD 2.5 Degree Low Resolution Time Series (LRTS), which
 209 provides daily LFRs on a 2.5° regular latitude–longitude grid for May 1995 – April 2015.

210 **2.4 CMIP6 model outputs for model comparison**

211 For the comparison of different model outputs from our study (CHASER) and other Earth system models or chemistry–
 212 climate models, we used LFR and surface temperature data from the CMIP6 CMIP Historical experiments from CESM2-
 213 WACCM (Danabasoglu, 2019), GISS-E2-1-G (Kelley et al., 2020), and UKESM1-0-LL (Tang et al., 2019). CESM2-
 214 WACCM uses the Community Earth System Model ver. 2 (Danabasoglu et al., 2020). The CESM2 is an open-source fully
 215 coupled Earth system model. The Whole Atmosphere Community Climate Model ver. 6 (WACCM6) is the atmospheric
 216 component coupled to the other components in CESM2. The GISS-E2-1-G is the NASA Goddard Institute for Space Studies
 217 (GISS) chemistry–climate model version E2.1 based on the GISS Ocean v1 (G01) model (Miller et al., 2014; Kelley et al.,
 218 2020). The UKESM1-0-LL is the UK's Earth system model, details of which were described by Sellar et al. (2019). We used
 219 3 ensembles from CESM2-WACCM, 9 ensembles from GISS-E2-1-G, and 18 ensembles from UKESM1-0-LL. Table S1
 220 presents all the ensemble members used for this study.

221 **2.5 Experiment setup**

222 We have conducted six sets of experiments with each set of experiments conducted using both the ECMWF-McCAUL
 223 (abbreviated as F1) and CTH (abbreviated as F2) schemes. Table 1 presents the major settings of all experiments with the
 224 relative explanations of those settings. STD-F1/F2 are standard experiments with the simulation period of 1959–2014. They
 225 are intended to reproduce the historical trends of lightning and LNO_x . Climate1959-F1/F2 are experiments that keep the
 226 climate simulations fixed to 1959 to derive the effects of global warming on historical lightning trends. ClimateAero1959-
 227 F1/F2 are intended to reflect the conditions with climate simulations and aerosol and aerosol precursor (BC, OC, NO_x , SO_2)
 228 emissions fixed to 1959. The Aero1959-F1/F2 experiments are the same as the STD-F1/F2 experiments, except for the
 229 AeroPEs fixed to 1959. The fifth set of experiments (Volca-off-F1/F2) was intended to exclude the influences of the
 230 Pinatubo volcanic eruption to compare to the STD-F1/F2 and to evaluate the Pinatubo eruption effects on historical
 231 lightning– LNO_x trends and variation.

232

233 We simulate volcanic aerosol forcing by considering the prescribed stratospheric aerosol extinction in the radiation scheme.
 234 We used the NASA Goddard Institute for Space Studies (GISS) (Sato et al., 1993) and Chemistry–Climate Model Initiative
 235 (CCMI) (Arfeuille et al., 2013) stratospheric aerosol dataset as the stratospheric aerosol climate data. The NASA GISS
 236 dataset includes monthly zonal-mean stratospheric aerosol optical thickness in four spectral bands. The CCMI dataset for
 237 CHASER includes monthly zonal-mean stratospheric aerosol extinction coefficients in 20 spectral bands. To remove the
 238 volcanic perturbation while maintaining the stratospheric background aerosol in the Volca-off-F1/F2, we used the following
 239 equation to process the Stratospheric Aerosol Climatology (SAC) during June 1991 – May 1996.

$$240 \quad \mathbf{SAC}_{\text{no_pinatubo}} = \begin{cases} \mathbf{SAC}_{\text{background}}, & |\mathbf{SAC}_{\text{raw}} - \mathbf{SAC}_{\text{background}}| > 1.96\sigma, \\ \mathbf{SAC}_{\text{raw}}, & |\mathbf{SAC}_{\text{raw}} - \mathbf{SAC}_{\text{background}}| \leq 1.96\sigma \end{cases} \quad (11)$$

241 In that equation, $\mathbf{SAC}_{\text{no_pinatubo}}$ denotes the stratospheric aerosol climatological data as input data for Volca-off-F1/F2
 242 experiments, $\mathbf{SAC}_{\text{background}}$ represents the stratospheric background aerosol climatological data (For this study,
 243 $\mathbf{SAC}_{\text{background}}$ is the corresponding temporal averaged values of the NASA GISS and CCMI stratospheric aerosol
 244 dataset during June 1986 – May 1991 and June 1996 – May 2001, when the time is close to the eruption and the
 245 stratosphere was less affected by volcanic eruptions). $\mathbf{SAC}_{\text{raw}}$ stands for the original values of NASA GISS and CCMI
 246 stratospheric aerosol dataset during June 1991 – May 1996. Moreover, σ symbolizes the standard deviations of
 247 stratospheric background aerosol climate data (For this study, σ are the corresponding standard deviations of NASA
 248 GISS and CCMI stratospheric aerosol dataset during June 1986 – May 1991 and June 1996 – May 2001). As displayed
 249 in Eq. (11), when the absolute differences between $\mathbf{SAC}_{\text{raw}}$ and $\mathbf{SAC}_{\text{background}}$ are larger than 1.96σ , we replace the
 250 original values (June 1991 – May 1996) of the SAC with the temporal averaged values of the NASA GISS and CCMI
 251 dataset during June 1986 – May 1991 and June 1996 – May 2001. When the absolute differences between $\mathbf{SAC}_{\text{raw}}$ and

252 **SAC_{background}** are equal to or smaller than 1.96σ , we still use the original values (June 1991 – May 1996) of the SAC
 253 for the Volca-off experiments. The value of 1.96σ corresponds to the 95% confidence interval, which can remove the
 254 Pinatubo perturbation sufficiently but which can maintain the background level of stratospheric aerosol during June
 255 1991 – May 1996. Furthermore, the influences of the Pinatubo eruption affected the HadISST SSTs/sea ice fields. To
 256 remove the Pinatubo eruption's influences on the SSTs/sea ice fields from the Volca-off experiments also, we replaced
 257 the 1991-06 – 1995-05 SSTs/sea ice data with HadISST SSTs/sea ice climatological data during 1985–1990 when
 258 conducting the Volca-off experiments. The 1985–1990 period was chosen because it is approximately the period of
 259 1991-06 – 1995-05 and because the SSTs/sea ice fields were less affected by volcanic activity during 1985–1990.
 260

261 All the experiments calculate the LNO_x emissions rates interactively by LNO_x emission parameterizations except STD-
 262 rVolcaoff experiments. The STD-rVolcaoff experiments are the same as the STD experiments except for reading the
 263 daily LNO_x emission rates calculated from the Volca-off experiments. The STD-rVolcaoff experiments are conducted
 264 for comparison with STD experiments to elucidate the effects of LNO_x emissions changes caused by the Pinatubo
 265 eruption on atmospheric chemistry (typically methane lifetime).
 266

Table 1: All experiments conducted for this study

Name of experiment	Period	Climate (SSTs, sea ice, GHGs) ^a	Anthropogenic and biomass burning emissions	Biogenic emissions	Stratospheric aerosol climatology
STD-F1/F2 ^b	1959–2014	1959–2014	CMIP6 1959–2014		NASA GISS and CCMI stratospheric aerosol dataset ^c
Climate1959-F1/F2	1959–2014	Fixed to 1959 ^d	CMIP6 1959–2014	VISIT and MEGAN ^f	As above
ClimateAero1959-F1/F2	1959–2014	Fixed to 1959	AeroPEs fixed to 1959 ^e		As above
Aero1959-F1/F2	1959–2014	1959–2014	AeroPEs fixed to 1959		As above
Volca-off-F1/F2	1990–1999	1990–1999 ^g	CMIP6 1990–1999		Same dataset with volcanic perturbation removed
STD-rVolcaoff-F1/F2	1990–1999	All settings are the same as those used for STD experiment except for reading of the daily LNO _x emission rates calculated from the Volca-off experiments			

267 ^a For the model simulations, the climate is simulated by the prescribed SSTs/sea ice fields and the prescribed varying
 268 concentrations of GHGs (CO₂, N₂O, methane, chlorofluorocarbons – CFCs – and hydrochlorofluorocarbons – HCFCs) used

269 only in the radiation scheme. The SSTs/sea ice fields are obtained from the HadISST dataset (Rayner et al., 2003). The
270 prescribed GHGs concentrations are derived from CMIP6 forcing datasets (Meinshausen et al., 2017).

271 ^bWe use “F1” to stand for the ECMWF-McCAUL scheme; “F2” represents the CTH scheme.

272 ^cStratospheric aerosol radiative forcing is simulated using the prescribed stratospheric aerosol extinction, which is obtained
273 from the NASA GISS (Sato et al., 1993) and CCM1 (Arfeuille et al., 2013) stratospheric aerosol dataset.

274 ^dThe climate is fixed to 1959 for the whole simulation period using the 1959 SSTs/sea ice field and GHG concentrations
275 during the simulation period.

276 ^eAerosol (BC, OC) and aerosol precursor (NO_x, SO₂) emissions (anthropogenic + biomass burning) are fixed to 1959
277 throughout the simulation period.

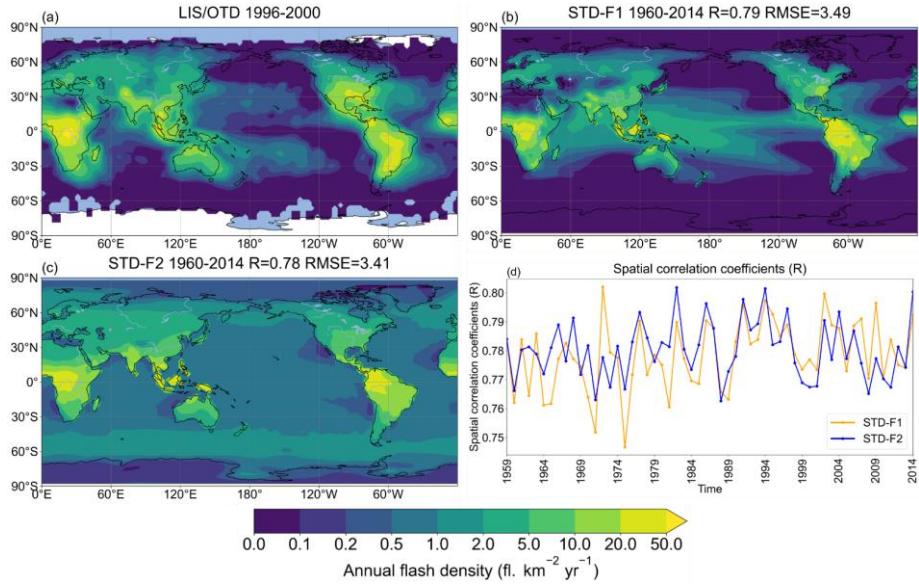
278 ^fSeveral biogenic emissions are interannually varying, including isoprene, monoterpenes, acetone, and methanol, which
279 were calculated using an off-line simulation using the Vegetation Integrative Simulator for Trace Gases (VISIT) terrestrial
280 ecosystem model (Ito and Inatomi, 2012). Some other reactive biogenic VOCs (ethane, propane, ethylene, propene) used are
281 climatological data derived from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) modeling system
282 (Guenther et al., 2012).

283 ^gHere the 1991-06 – 1995-05 SSTs/sea ice data were replaced with HadISST SSTs/sea ice climatological data during
284 1985–1990.

285 3 Results and Discussion

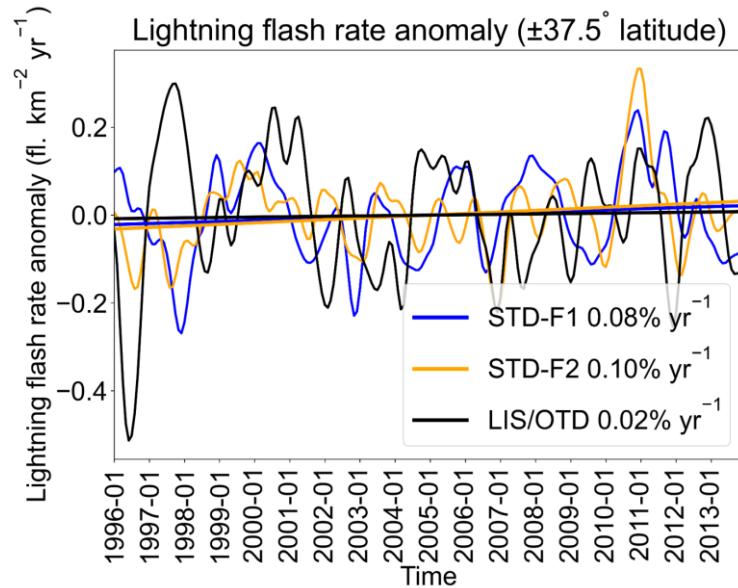
286 3.1 Validation of the simulated historical lightning distribution and trend

287 To increase the credibility of the conclusions obtained based only on the numerical simulations, the model calculations must
288 be evaluated using observational data. We used the LIS/OTD observations to evaluate the spatial and temporal distribution
289 and historical lightning trends simulated by CHASER (MIROC). Figures 1a–1c show the annual mean spatial distributions
290 of lightning observed by LIS/OTD and from model simulations using the ECMWF-McCAUL and CTH schemes. Both the
291 ECMWF-McCAUL and CTH schemes generally captured the hotspots of lightning (Central Africa, Maritime Continent,
292 South America), with strong spatial correlations between observations and model simulations ($R > 0.75$), even the lightning
293 distributions were not well captured over the ocean. Figure 1d exhibits strong spatial correlation between observations and
294 simulation results maintained throughout the simulation period (1959–2014).



295

296 **Figure 1:** Annual mean lightning flash densities from (a) LIS/OTD satellite observations spanning 1996–2000, (b) the STD
 297 experiment (1960–2014) with the ECMWF-McCAUL scheme used, (c) the STD experiment (1960–2014) with the CTH scheme
 298 used. R and RMSE shown in the titles of panels (b) and (c) are calculated between panels (b)–(c) and (a). Panel (d) presents the
 299 spatial correlation coefficients between modeled spatial lightning distribution of each year and LIS/OTD lightning climatologies
 300 during 1996–2000.



301

302 **Figure 2:** LFR anomalies of 1996–2013 within $\pm 37.5^\circ$ latitude obtained from two numerical experiments (STD-F1/F2) and
 303 LIS/OTD satellite observations. Curves represent the monthly time-series data of the $\pm 37.5^\circ$ latitude mean LFR anomalies with the

304 **1-D Gaussian (Denoising) filter applied. Lines are the fitting curves of the monthly time-series data of the $\pm 37.5^\circ$ latitude mean**
305 **LFR anomalies. Trends of the LFR anomalies in $\% \text{ yr}^{-1}$ are also presented in the legends.**

306

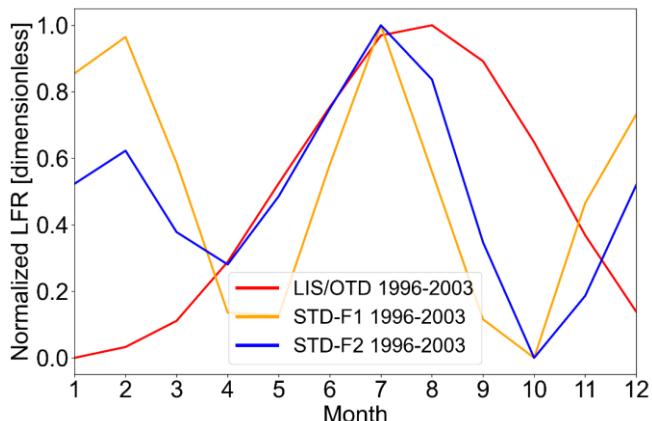
307 **Table 2: A statistical summary of the trends shown in Fig. 2 by Mann–Kendall rank statistic and Sen’s slope estimator. The**
308 **monthly time-series data of the $\pm 37.5^\circ$ latitude mean LFR anomalies were estimated by Mann–Kendall rank statistic and Sen’s**
309 **slope estimator. The column “Trend” shows whether these are significant trends with the significance set as 5%, as well as the**
310 **percentage trends in $\% \text{ yr}^{-1}$ estimated by linear regression. The “*p*-value” is calculated during Mann–Kendall trend test. “Slope”**
311 **shows Sen’s slope of trend. Q_{\min} and Q_{\max} respectively denote the lower and upper limits of the 95% confidence interval of Sen’s**
312 **slope.**

Experiment/dataset	Trend	<i>p</i> -value	Slope	Q_{\min}	Q_{\max}
STD-F1	No trend, $0.08 \% \text{ yr}^{-1}$	$p > 0.05$	0.0001	-0.0003	0.0005
STD-F2	No trend, $0.10 \% \text{ yr}^{-1}$	$p > 0.05$	0.0003	0.0	0.0006
LIS/OTD	No trend, $0.02 \% \text{ yr}^{-1}$	$p > 0.05$	-0.0001	-0.0006	0.0004

313

314 The LIS/OTD observations are also used to evaluate historical lightning trends simulated by CHASER (MIROC). We
315 examined the $\pm 37.5^\circ$ latitude mean LFR anomaly (1996–2013) calculated from LIS/OTD observations and STD-F1/F2
316 numerical experiments (Fig. 2 and Table 2). We also note some missing values within the $\pm 37.5^\circ$ latitude in LIS/OTD
317 observations. To constrain the comparisons between observations and simulations as like-for-like, when we encounter a
318 missing value in the LIS/OTD observations during spatial averaging, we also treat the CHASER simulated value at the same
319 location as a missing value. As displayed in Fig. 2, we would not necessarily expect that interannual variations of LFR
320 anomaly can be captured, because meteorological nudging was not applied and the simulated LFRs were only controlled by
321 the prescribed SSTs/sea ice data. Nevertheless, the overall trends of LFR anomaly simulated using both schemes well-
322 matched the LIS/OTD observations, as portrayed in Fig. 2. We further investigated the trends shown in Fig. 2 by Mann–
323 Kendall rank statistic and Sen’s slope estimator and the statistical summary is displayed in Table 2 (Salmi et al., 2002;
324 Hussain and Mahmud, 2019). Neither the LFR anomaly (within $\pm 37.5^\circ$ latitude) derived from LIS/OTD observations nor
325 simulations show a significant trend for 1996–2013 using the Mann–Kendall rank statistic test (significance inferred for 5%).
326 The global LFR anomaly during 1993–2013 obtained from simulations (STD-F1/F2) also shows no significant trend, which
327 is consistent with the Schuman Resonance (SR) intensity observations (1993–2013) at Rhode Island, USA (Earle Williams,
328 2022). However, the SR observations in Rhode Island (USA) exclude consideration of the influences of solar cycles, which
329 makes it less appropriate for lightning trend evaluation.

Mean annual cycle in area average lightning flash rate



330

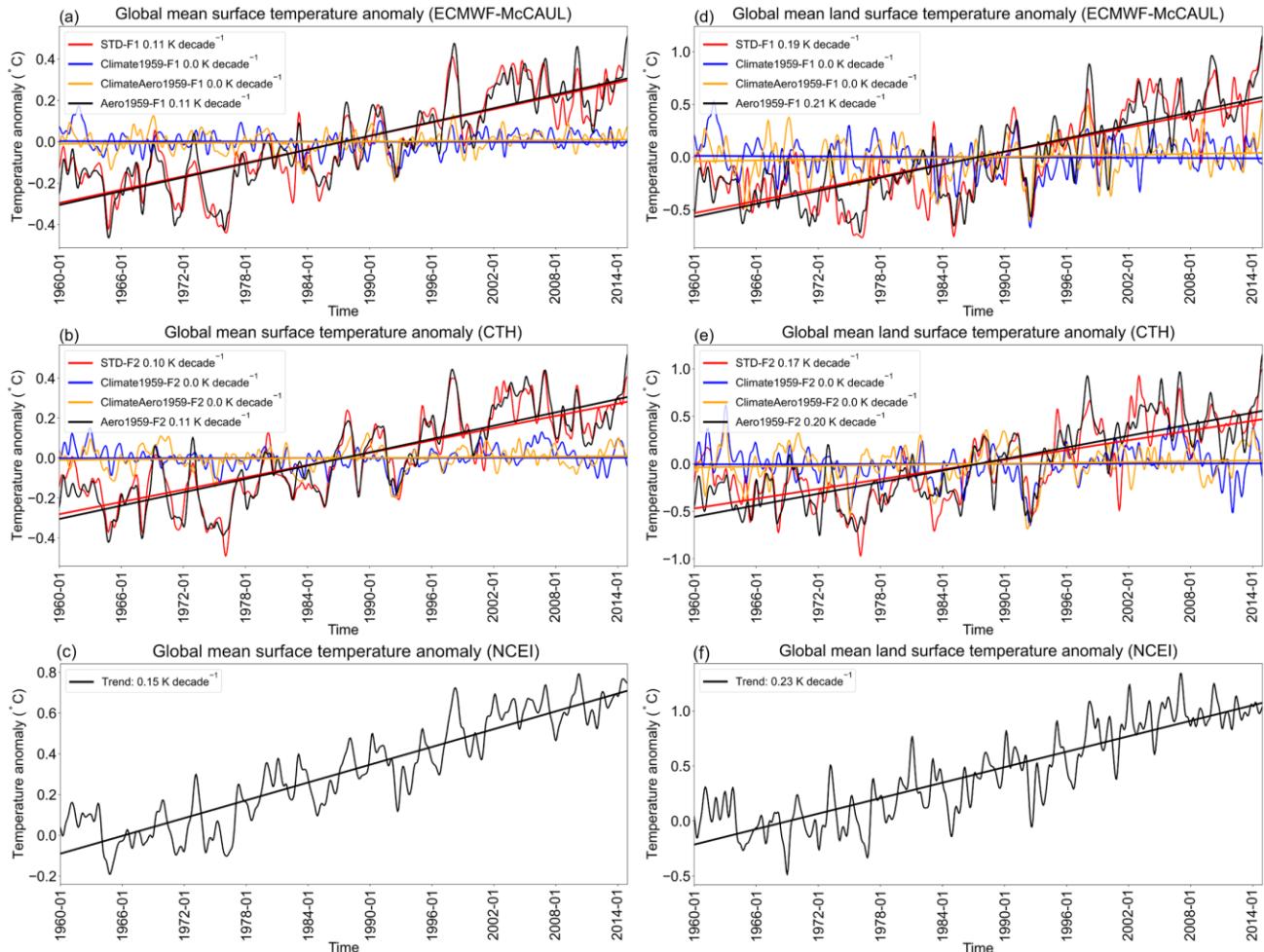
331 **Figure 3: Mean annual cycle in area average LFR during 1996–2003. The area average was taken over the grid cells where valid**
 332 **LIS/OTD lightning observations exist. LFR is normalized by min-max normalization.**

333

334 We further investigated the seasonal variabilities of simulated LFR and compared them against LIS/OTD observations. The
 335 results are depicted in Fig. 3. Both the CTH and ECMWF-McCAUL schemes captured the peak during JJA, but the
 336 overestimation of LFR by F1/F2 during DJF is also noticeable. Figure S1 presents comparison of the LFR global distribution
 337 in different seasons during 1996–2003 from LIS/OTD lightning observations and STD experiment outputs. Generally,
 338 CHASER well-captured the spatial distribution of LFR in all four seasons when compared against LIS/OTD observations.
 339 The spatial correlation coefficients (R) between observations and simulations are highest ($R=0.80$ for both lightning
 340 schemes) in DJF, indicating CHASER's considerable capability to reproduce the LFR spatial distribution in DJF. As
 341 displayed in the first row of Fig. S1, the overestimation of LFR by F1/F2 during DJF is primarily attributable to the
 342 overestimation of LFR within the Maritime Continent and South America, but this might also be attributable to the
 343 underestimation of LFR by LIS/OTD within these two regions. It is believed that the LIS/OTD lightning detection efficiency
 344 is highly sensitive to the characteristic of convective clouds (cloud albedo, cloud optical thickness, etc.) (Boccippio et al.,
 345 2002; Cecil et al., 2014). High cloud albedo and cloud optical thickness might engender the underestimation of LFR by
 346 LIS/OTD. It is also noteworthy that the seasonal variation and long-term trend of global lightning are strongly influenced by
 347 distinct different factors. The seasonal variation of global lightning activities is most strongly affected by the 23° obliquity of
 348 Earth's orbit and the asymmetric distribution of the continent between the Northern and Southern hemispheres. However, the
 349 long-term global lightning trend we investigated for this study is controlled mainly by climate forcers such as aerosols and
 350 GHGs. To minimize the effects of LFR seasonal variation on our study's results, we deseasonalized the results shown in all
 351 figures and tables by calculating their anomaly based on raw data. The validation described above and the deseasonalization
 352 of our study's results justified that the LFR seasonal variation (and the uncertainties in the simulation of LFR seasonal
 353 variation) in our study has a limited effect on these study results.

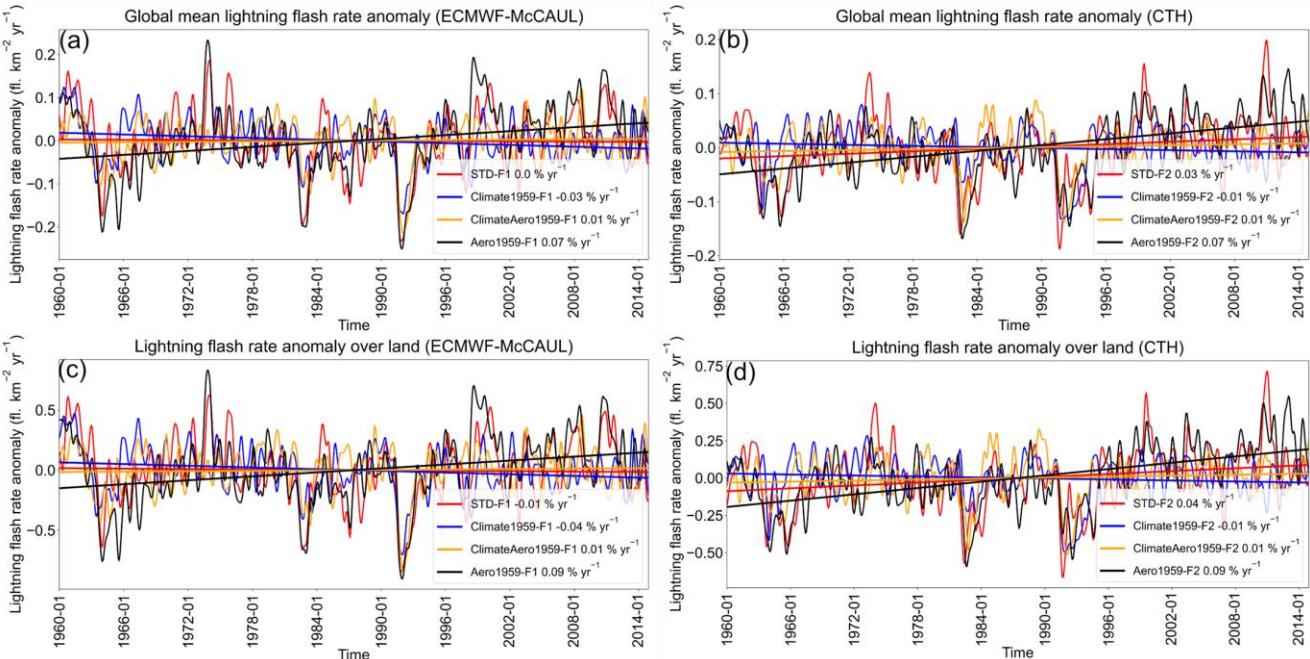
354 **3.2 Effects of global warming and increases in AeroPEs on historical lightning–LNO_x trends**

355 As introduced in Sect. 1, global warming and changes in AeroPEs are the two main factors which influence long-term
356 (1960–2014) historical lightning trends (Hereinafter, historical lightning trends represent lightning trends of 1960–2014.).
357 Evidence shows that the Pacific Decadal Oscillation (PDO) can also affect lightning trends over decadal time scales (Macias
358 Fauria and Johnson, 2006; Mallick et al., 2022), and further research is anticipated to verify it. To analyze the effects of
359 global warming on historical lightning trends, we designed and conducted two sets of experiments: one set of experiments
360 including “global warming” (STD-F1/F2) and another set of experiments excluding “global warming” (Climate1959-F1/F2).
361 Figures 4a and 4b respectively depict the global surface temperature anomalies calculated using the ECMWF-McCAUL and
362 CTH schemes. The STD and Aero1959 experiments show an increasing trend (around $0.11 \text{ K decade}^{-1}$) of global mean
363 surface temperature anomalies, which closely approximates the trend (around $0.15 \text{ K decade}^{-1}$) obtained from NOAA’s
364 National Centers for Environmental Information (NCEI) (Figs. 4c, 4f). Global temperature change data from 1880 to the
365 present are available from the NCEI, which tracks variations of the Earth’s temperature based on thousands of stations’
366 observation data around the globe (Climate at a Glance | National Centers for Environmental Information (NCEI), 2022).
367 When the prescribed SSTs/sea ice fields and GHGs concentrations were fixed to 1959 throughout the simulation period, the
368 simulated trends of global mean surface temperature anomalies turned out to be flat (Climate1959 and ClimateAero1959).
369 To elucidate the effects of increases in AeroPEs on averaged surface temperature to the greatest extent possible, we also
370 show the averaged surface temperature anomaly only over land regions (Figs. 4d–4f). The simulated global mean land
371 surface temperature anomalies are also well-matched with the NCEI observational data. The aerosol cooling effect can be
372 more evident when only examining surface temperature trends averaged over land (Figs. 4d–4e).



373

374 **Figure 4: Monthly time-series data of global mean surface temperature anomalies with 1-D Gaussian (Denoising) filter applied and**
 375 **their fitting curves calculated from the outputs of numerical experiments (a-b) and obtained from NCEI (c). Panels (d)-(f) are the**
 376 **same as panels (a)-(c), but the averaged surface temperature anomalies are only calculated within the global land regions. The**
 377 **trends of the fitting curves in K decade⁻¹ are also presented in the legends.**



378

379 **Figure 5:** Panels (a) and (b) show monthly time-series data of global mean LFR anomalies with 1-D Gaussian (Denoising) Filter
 380 applied and their fitting curves of different experiments simulated respectively using the ECMWF-McCAUL scheme and CTH
 381 scheme. Panels (c) and (d) are the same as panels (a) and (b), except that the averaged LFR anomalies are calculated only within
 382 global land regions. Trends of the fitting curves ($\% \text{ yr}^{-1}$) are also shown in the legends.

383

384 **Table 3:** A statistical summary of the trends shown in Fig. 5 by Mann-Kendall rank statistic and Sen's slope estimator. The
 385 monthly time-series data of global or land mean LFR anomalies were estimated by Mann-Kendall rank statistic and Sen's slope
 386 estimator. The column "Trend" shows whether these are significant trends with the significance set as 5%, as well as the
 387 percentage trends in $\% \text{ yr}^{-1}$ estimated by linear regression. The "p-value" is calculated during Mann-Kendall trend test. "Slope"
 388 shows Sen's slope of trend. Q_{\min} and Q_{\max} respectively denote the lower and upper limits of the 95% confidence interval of Sen's
 389 slope.

Experiment	Trend	p-value	Slope	Q_{\min}	Q_{\max}
STD-F1 (global)	No trend, 0.0 $\% \text{ yr}^{-1}$	$p > 0.05$	0.0	-0.0001	0.0
Climate1959-F1 (global)	Decreasing, -0.03 $\% \text{ yr}^{-1}$	$p < 0.01$	-0.0001	-0.0001	0.0
ClimateAero1959-F1 (global)	No trend, 0.01 $\% \text{ yr}^{-1}$	$p > 0.05$	0.0	0.0	0.0001
Aero1959-F1 (global)	Increasing, 0.07 $\% \text{ yr}^{-1}$	$p < 0.01$	0.0001	0.0001	0.0002
STD-F1 – Climate1959-F1 (global)	No trend, 0.03 $\% \text{ yr}^{-1}$	$p > 0.05$	0.0001	0.0	0.0001
STD-F1 – Aero1959-F1 (global)	Decreasing, -0.07 $\% \text{ yr}^{-1}$	$p < 0.01$	-0.0001	-0.0002	-0.0001
STD-F1 (land)	No trend, -0.01 $\% \text{ yr}^{-1}$	$p > 0.05$	0.0	-0.0002	0.0001
Climate1959-F1 (land)	Decreasing, -0.04 $\% \text{ yr}^{-1}$	$p < 0.01$	-0.0002	-0.0004	-0.0001

ClimateAero1959-F1 (land)	No trend, 0.01 % yr^{-1}	$p > 0.05$	0.0001	-0.0001	0.0002
Aero1959-F1 (land)	Increasing, 0.09 % yr^{-1}	$p < 0.01$	0.0005	0.0003	0.0006
STD-F1 – Climate1959-F1 (land)	No trend, 0.03 % yr^{-1}	$p > 0.05$	0.0002	-0.0001	0.0004
STD-F1 – Aero1959-F1 (land)	Decreasing, -0.10 % yr^{-1}	$p < 0.01$	-0.0005	-0.0007	-0.0003
STD-F2 (global)	Increasing, 0.03 % yr^{-1}	$p < 0.01$	0.0001	0.0	0.0001
Climate1959-F2 (global)	No trend, -0.01 % yr^{-1}	$p > 0.05$	0.0	-0.0001	0.0
ClimateAero1959-F2 (global)	No trend, 0.01 % yr^{-1}	$p > 0.05$	0.0	0.0	0.0001
Aero1959-F2 (global)	Increasing, 0.07 % yr^{-1}	$p < 0.01$	0.0001	0.0001	0.0002
STD-F2 – Climate1959-F2 (global)	Increasing, 0.04 % yr^{-1}	$p < 0.01$	0.0001	0.0	0.0001
STD-F2 – Aero1959-F2 (global)	Decreasing, -0.04 % yr^{-1}	$p < 0.01$	-0.0001	-0.0001	0.0
STD-F2 (land)	Increasing, 0.04 % yr^{-1}	$p < 0.01$	0.0003	0.0001	0.0004
Climate1959-F2 (land)	No trend, -0.01 % yr^{-1}	$p > 0.05$	-0.0001	-0.0002	0.0
ClimateAero1959-F2 (land)	No trend, 0.01 % yr^{-1}	$p > 0.05$	0.0001	0.0	0.0002
Aero1959-F2 (land)	Increasing, 0.09 % yr^{-1}	$p < 0.01$	0.0006	0.0004	0.0007
STD-F2 – Climate1959-F2 (land)	Increasing, 0.05 % yr^{-1}	$p < 0.01$	0.0003	0.0001	0.0005
STD-F2 – Aero1959-F2 (land)	Decreasing, -0.05 % yr^{-1}	$p < 0.01$	-0.0003	-0.0005	-0.0001

390

391 Figure 5, panels (a) and (b) respectively portray the global mean LFR anomalies and their fitting curves obtained from the
 392 outputs of the ECMWF-McCAUL scheme and CTH scheme. Besides, we displayed in Table 3 the statistical summary of the
 393 trends in Fig. 5 utilizing Mann–Kendall rank statistic and Sen’s slope estimator. The global lightning trend obtained from the
 394 STD-F1 experiment turned out to be statistically flat (0.0% yr^{-1}), whereas the outputs of the STD-F2 experiment exhibit a
 395 significant increasing global lightning trend (0.03% yr^{-1}) determined using the Mann–Kendall rank statistic (significance
 396 inferred for 5%).

397

398 Comparison of the lightning trends calculated from the STD and Climate1959 experiments showed that both lightning
 399 schemes demonstrated that historical global warming (1960–2014) enhances the global lightning trends toward positive
 400 trends (around 0.03% yr^{-1} or 3% K^{-1}). Global warming effects on historical lightning trends were evaluated as significant
 401 using the Mann–Kendall rank statistic, with significance inferred for 5%, when using the CTH scheme, but not in the case of
 402 the ECMWF-McCAUL scheme (see rows “STD-F1 – Climate1959-F1 (global)” and “STD-F2 – Climate1959-F2 (global)”
 403 ” in Table 3). As shown in Table 3, the differences in global lightning trends simulated by the STD-F1/F2 and Aero1959-
 404 F1/F2 experiments indicate that the increases in AeroPEs during 1960–2014 significantly suppress the global lightning
 405 trends (-0.07% yr^{-1} – -0.04% yr^{-1}). It is noteworthy that this suppression of lightning trends is only attributable to aerosol
 406 radiative effects. Further research must be conducted to elucidate the long-term effects of aerosols on lightning through
 407 aerosol microphysical effects. We also investigated lightning trends only over land regions (Figs. 5c–5d and Table 3) to

ascertain the effects of changes in AeroPEs to the greatest extent possible. When observing the lightning trends over land only, the degree of suppression of lightning trends attributable to increases in AeroPEs expands to $-0.10\% \text{ yr}^{-1} - -0.05\% \text{ yr}^{-1}$, which is attributable to most AeroPEs and their growth coming from land regions. It is noteworthy that we used the same SSTs/sea ice data in the Aero1959 as those used for STD experiments. The SSTs/sea ice data also reflected the effects of increases in AeroPEs. Therefore, we might underestimate the effects of increases in AeroPEs on lightning trends by comparing the results of STD and Aero1959 experiments.

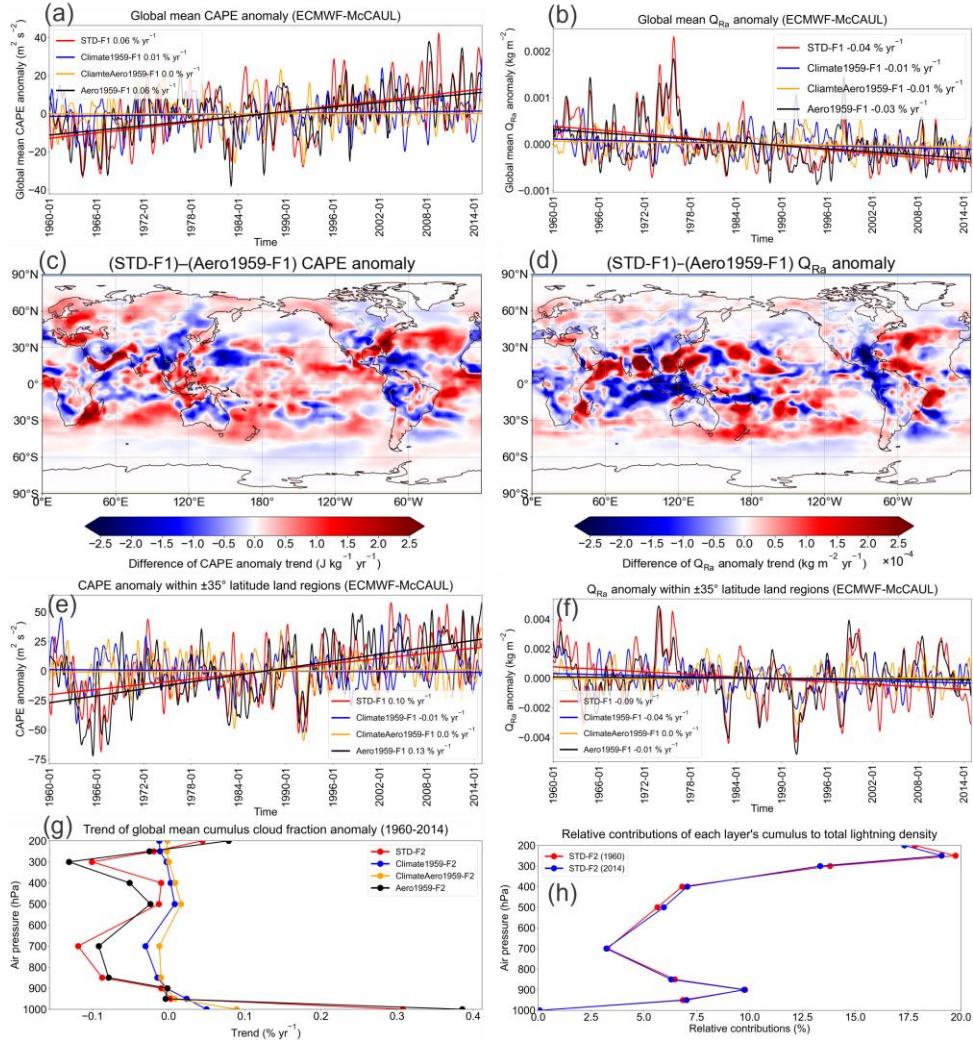


Figure 6: Panels (a) and (b) respectively show monthly time-series data of global mean CAPE and Q_{Ra} anomalies with 1-D Gaussian (Denoising) filter applied and their fitting curves simulated using the ECMWF-McCAUL scheme. Panels (c) and (d) respectively show differences in the CAPE anomaly trend ($\text{J kg}^{-1} \text{ yr}^{-1}$) and Q_{Ra} anomaly trend ($\text{kg m}^{-2} \text{ yr}^{-1}$) of the STD-F1 and Aero1959-F1 experiments in the global map. Panels (e) and (f) respectively show monthly time-series data of $\pm 35^\circ$ latitude land region mean CAPE and Q_{Ra} anomalies with 1-D Gaussian (Denoising) filter applied and their fitting curves simulated using the

420 **ECMWF-McCAUL scheme. Figure 6(g) portrays the vertical profiles of the trend of global mean cumulus cloud fraction anomaly**
421 **simulated by the CTH scheme. Panel (h) depicts the relative contributions of each layer's cumulus to total lightning density in 1960**
422 **and 2014, as calculated from the outputs of the STD-F2 experiment.**

423

424 For the ECMWF-McCAUL scheme, model outputs affirm that global warming can enhance the global mean CAPE anomaly
425 slightly and suppress the global mean Q_{Ra} anomaly (Figs. 6a–6b). Earlier studies have also indicated that the total solid
426 (cloud ice, snow, and graupel) mass mixing ratio within charge separation regions is lower under global warming. Moreover,
427 possible explanations are given in those studies (Finney et al., 2018; Romps, 2019). Because global warming enhances
428 global convection activities, and because lightning formation is highly related to convection activity, global warming
429 enhances the historical global lightning trend simulated using the ECMWF-McCAUL scheme, mainly as a result of the
430 simulated CAPE trend, which is enhanced by global warming.

431

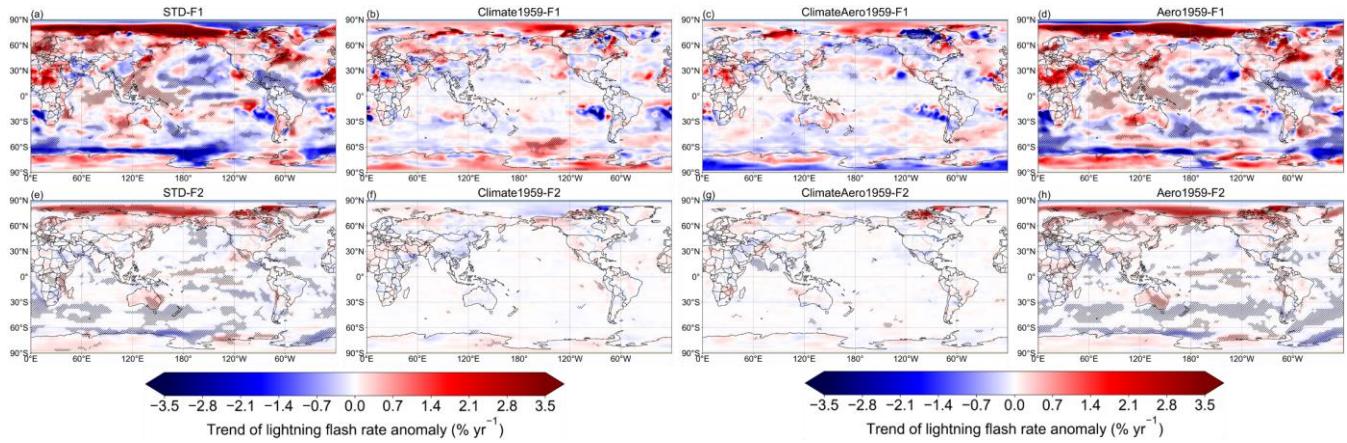
432 The past increases in AeroPEs exert negligible effects on the trends of global mean CAPE and Q_{Ra} anomalies, as displayed
433 in Figs. 6a–6b. However, as also demonstrated in our study (see Fig. 1), most lightning flashes occur over tropical and
434 subtropical land regions. It is displayed in Figs. 6c–6d that the past increases in AeroPEs mostly suppress the CAPE and Q_{Ra}
435 absolute trends within regions with high lightning densities. We further investigated the trends of $\pm 35^\circ$ latitude land region
436 mean CAPE and Q_{Ra} anomalies, and the results are portrayed in Figs. 6e–6f. Figs. 6e–6f show that past increases in
437 AeroPEs significantly suppress the Q_{Ra} trend ($-0.08\% \text{ yr}^{-1}$) and slightly suppress the CAPE trend ($-0.03\% \text{ yr}^{-1}$) within $\pm 35^\circ$
438 latitude land regions. Weaker convection activities (smaller CAPE) and fewer hydrometeors (cloud ice, graupel, snow) in the
439 charge separation regions ($0^\circ\text{C} - -25^\circ\text{C}$ isotherm) engender less lightning. In the case of the ECMWF-McCAUL scheme,
440 CAPE and Q_{Ra} trends were suppressed within $\pm 35^\circ$ latitude terrestrial regions. This constitutes the main reason for the
441 suppression of the historical global lightning trends induced by increases in AeroPEs through aerosol radiative effects. It is
442 noteworthy that, because the aerosol microphysical effects are only considered in the grid-scale large-scale condensation
443 scheme, our study might underestimate the aerosol microphysical effects which can enhance the trends of Q_{Ra} and LFR
444 toward the positive direction.

445

446 To explain the results simulated by the CTH scheme, we investigated the vertical profiles of the trend of the global mean
447 cumulus cloud fraction anomaly (Fig. 6g). Investigating cumulus cloud fraction is reasonable because each model layer's
448 cumulus cloud fractions are used to weight the calculated lightning densities from that layer in the CTH scheme, as
449 introduced in equations (3) and (4). Figure 6h shows the relative contributions of each model layer's cumulus to the
450 calculated global total lightning densities in 1960 and 2014 obtained using the CTH scheme. As Fig. 6h displayed, the
451 vertical profiles of relative contribution in 1960 and 2014 are almost identical. Cumulus convection is positively correlated
452 with lightning formation, which is the scientific basis of parameterizing lightning densities using the cumulus cloud top
453 height: the CTH scheme. Historical global warming enhances the lightning trend simulated by the CTH scheme mainly

454 because the simulated historical global warming increases the cumulus reaching 200 hPa, which contributes greatly to the
 455 simulated global total lightning density (Figs. 6g–6h). The increases in the deep convective cloud are regarded as related to
 456 the increases in tropopause height attributable to global warming, as shown in Fig. S2. The past increases in AeroPEs
 457 suppress the lightning trend simulated by the CTH scheme because increases in AeroPEs decrease the cumulus reaching 200
 458 hPa as well as the cumulus within the lower to middle troposphere by aerosol radiative effects (Fig. 6g). In addition, in the
 459 supplement, we present a figure (Fig. S3) resembling Fig. 6, but which includes only consideration of land regions. The
 460 mechanisms of global warming and increases in AeroPEs affecting lightning trends over land regions are similar to those
 461 described above on a global scale. We do not discuss details of them here.

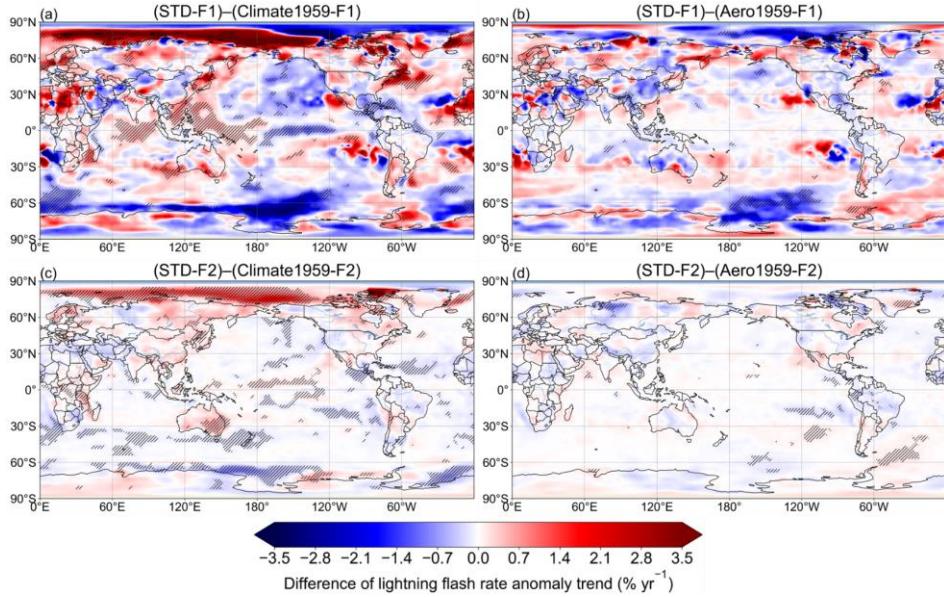
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463

464 **Figure 7: Trends of LFR anomaly ($\% \text{ yr}^{-1}$) during 1960–2014 on the two-dimensional map. The trend at every point was calculated**
 465 **from the function of approximating curve for the 1960–2014 time-series data (LFR anomaly) at each grid cell. The area in which**
 466 **the trend was found to be significant by the Mann–Kendall rank statistic test (significance inferred for 5%) is marked with**
 467 **hatched lines.**

468



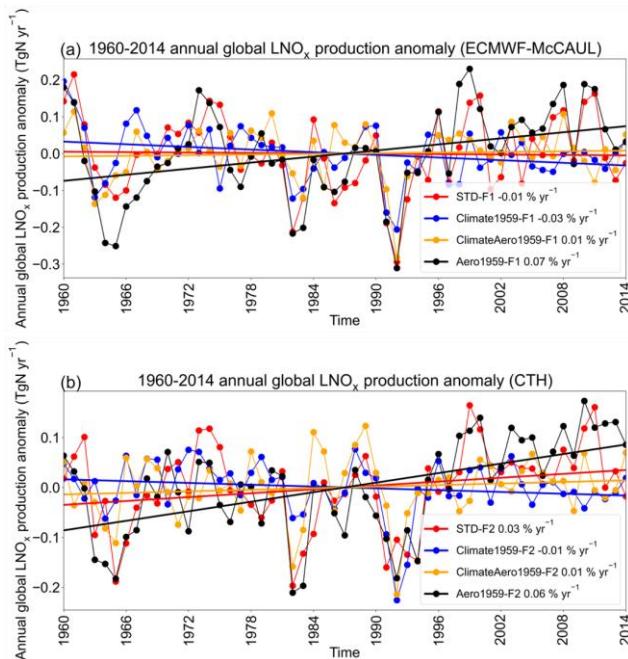
469

470 **Figure 8: Differences in trends of LFR anomaly during 1960–2014 on the global map. The area in which the trend of the**
 471 **differences of LFR anomaly time-series data was found to be significant by the Mann–Kendall rank statistic test (significance**
 472 **inferred for 5%) is marked with hatched lines.**

473

474 We also investigated lightning trends simulated in different experiments with the global map (Fig. 7). Both the ECMWF–
 475 McCaul and the CTH schemes show that lightning increased significantly in most parts of the Arctic region and decreased
 476 in some parts of the Southern Ocean during 1960–2014 (Figs. 7a, 7e). The significant lightning trends presented in Figs. 7a
 477 became nearly nonexistent when the climate simulations were fixed to 1959 (Figs. 7b, 7f), indicating the considerable effects
 478 of global warming on the trend of global lightning activities. Furthermore, the effects of past global warming and increases
 479 in AeroPEs on the lightning trends on the global map are displayed in Fig. 8. Figures 8a and 8c show that past global
 480 warming enhances lightning activities within the Arctic region and Japan, which is consistent with findings of an earlier
 481 study from which Japan thunder day data were reported (Fujibe, 2017). Figures 8a and 8c also show that historical global
 482 warming suppresses lightning activities around New Zealand and some parts of the Southern Ocean. Both lightning schemes
 483 demonstrated that the historical increases in AeroPEs suppress lightning activities in some parts of the Southern Ocean and
 484 South America. The ECMWF-McCAUL scheme also suggests that historical increases in AeroPEs suppress lightning
 485 activities by aerosol radiative effects in some parts of India and China, where AeroPEs increased dramatically during 1960–
 486 2014 because of rapid economic development and energy consumption. Many observation-based studies indicate that
 487 aerosols can invigorate lightning activities in some regions of China and India, typically under relatively clean conditions
 488 (e.g., $AOD < 1.0$), which is attributable to the aerosol microphysical effects (Wang et al., 2011; Zhao et al., 2017; Lal et al.,
 489 2018; Liu et al., 2020; Shi et al., 2020; Zhao et al., 2020). Therefore, a total positive effect of aerosol on historical lightning
 490 trends in China and India cannot be ruled out. We further provided the same figures as Figs. 7 and 8, but using different units

491 (fl. $\text{km}^{-2} \text{ yr}^{-2}$) in the supplementary information (Figs. S4 and S5). Figures S4 and S5 show that the absolute lightning trends
 492 (fl. $\text{km}^{-2} \text{ yr}^{-2}$) and the effects of global warming and increases in AeroPEs on the absolute lightning trends are slight in high-
 493 latitude regions but prominent in tropical areas.



494
 495 **Figure 9: Time-series data of 1960–2014 annual global LNO_x production anomalies (TgN yr^{-1}) and their fitting curves simulated
 496 using the ECMWF-McCAUL scheme (a) and the CTH scheme (b). Trends of the fitting curves in percent per year are presented in
 497 the legends.**
 498

499 **Table 4: A statistical summary of the trends shown in Fig. 9 by Mann–Kendall rank statistic and Sen’s slope estimator. The time-
 500 series data of annual global LNO_x production anomalies were estimated by Mann–Kendall rank statistic and Sen’s slope estimator.
 501 The column “Trend” shows whether these are significant trends with the significance set as 5%, as well as the percentage trends
 502 in yr^{-1} estimated by linear regression. The “*p*-value” is calculated during Mann-Kendall trend test. “Slope” shows Sen’s slope of
 503 trend. Q_{\min} and Q_{\max} respectively denote the lower and upper limits of the 95% confidence interval of Sen’s slope.**
 504

Experiment	Trend	<i>p</i> -value	Slope	Q_{\min}	Q_{\max}
STD-F1	No trend, -0.01 % yr^{-1}	$p > 0.05$	-0.0001	-0.002	0.0018
Climate1959-F1	Decreasing, -0.03 % yr^{-1}	$p < 0.05$	-0.0011	-0.0024	-0.0001
ClimateAero1959-F1	No trend, 0.01 % yr^{-1}	$p > 0.05$	0.0003	-0.0008	0.0013
Aero1959-F1	Increasing, 0.07 % yr^{-1}	$p < 0.01$	0.003	0.0011	0.0048
STD-F1 – Climate1959-F1	No trend, 0.02 % yr^{-1}	$p > 0.05$	0.0009	-0.0009	0.0025
STD-F1 – Aero1959-F1	Decreasing, -0.08 % yr^{-1}	$p < 0.01$	-0.003	-0.004	-0.0021

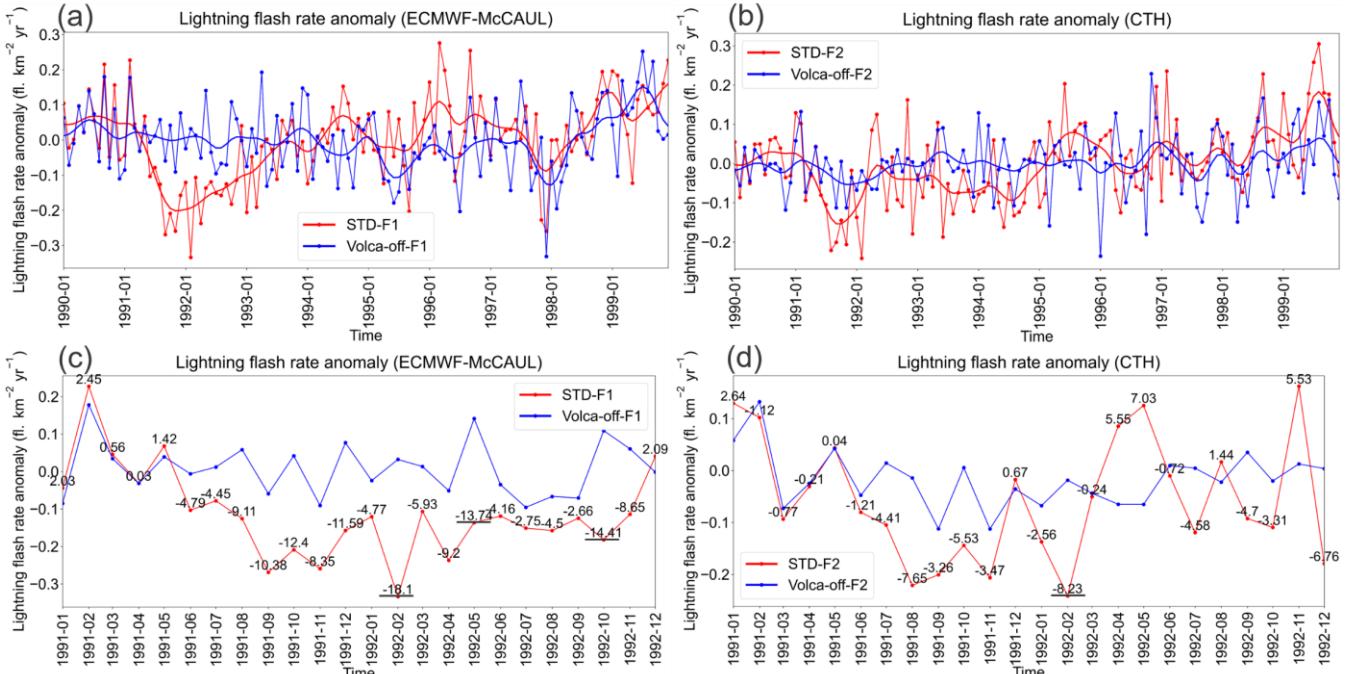
STD-F2	Increasing, 0.03 % yr ⁻¹	p < 0.05	0.0013	0.0001	0.0024
Climate1959-F2	No trend, -0.01 % yr ⁻¹	p > 0.05	-0.0007	-0.0014	0.0001
ClimateAero1959-F2	No trend, 0.01 % yr ⁻¹	p > 0.05	0.0005	-0.0004	0.0015
Aero1959-F2	Increasing, 0.06 % yr ⁻¹	p < 0.01	0.0033	0.0019	0.0046
STD-F2 – Climate1959-F2	Increasing, 0.04 % yr ⁻¹	p < 0.01	0.0021	0.0006	0.0033
STD-F2 – Aero1959-F2	Decreasing, -0.03 % yr ⁻¹	p < 0.01	-0.0019	-0.0029	-0.001

505

506 Trends in historical annual global LNO_x emissions for different scenarios are generally consistent with trends in historical
 507 global mean LFRs, as shown in Figs. 5a–5b and Fig. 9. This finding is not surprising because, as the lightning NO_x emission
 508 parameterizations introduced in Sect. 2.2 show, the simulated LFRs are linearly related to the simulated LNO_x emissions in
 509 our study. Comparison of the LNO_x trends calculated from the STD and Climate1959 experiments showed that both
 510 lightning schemes demonstrated that historical global warming (1960–2014) enhances the global LNO_x trends toward
 511 positive trends (0.02% yr⁻¹ – 0.04% yr⁻¹). Global warming effects on historical LNO_x trends were evaluated as significant
 512 using the Mann–Kendall rank statistic, with significance inferred for 5%, when using the CTH scheme, but not in the case of
 513 the ECMWF-McCAUL scheme (see rows “STD-F1 – Climate1959-F1” and “STD-F2 – Climate1959-F2” in Table 4). As
 514 shown in Table 4, the differences in global LNO_x trends simulated by the STD and Aero1959 experiments indicate that the
 515 increases in AeroPEs during 1960–2014 significantly suppress the global LNO_x trends (-0.08% yr⁻¹ – -0.03% yr⁻¹). The
 516 results presented in Fig. 9 and Table 4 imply that historical global warming and increases in AeroPEs can affect atmospheric
 517 chemistry and can engender feedback by influencing LNO_x emissions.

518 3.3 Pinatubo volcanic eruption effects on historical lightning–LNO_x trends

519 We estimate the Pinatubo eruption effects on historical lightning–LNO_x trends and variation by comparing the simulation
 520 results of STD and Volca-off experiments. The simulated global mean LFRs by STD and Volca-off experiments are the
 521 same until April 1991. They then begin to show differences from May 1991 (The time series of global mean LFRs is not
 522 shown.). This result is reasonable because the Pinatubo volcanic perturbations are removed from SAC during June 1991 –
 523 May 1996 in the Volca-off experiments by equation (11), and because the SAC of May 1991 used in CHASER is
 524 interpolated between the SAC of April 1991 and June 1991.



525

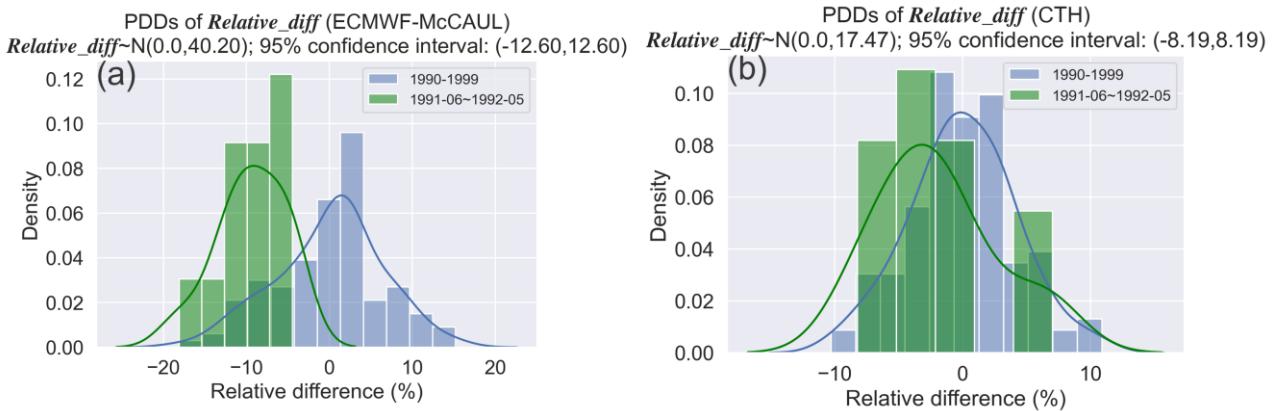
526 **Figure 10: Time series of LFR anomalies during 1990–1999 or during 1991–1992. Panels (a) and (b) show the time series of LFR**
 527 **anomalies and their smoothed curves by 1-D Gaussian (Denoising) filter for 1990–1999. Panels (c) and (d) present the time series of**
 528 **LFR anomalies during 1991–1992. Values shown over the red lines in panels (c) and (d) are *Relative_diff* calculated using**
 529 **equation 12.**

530

531 Figures 10c–10d portray the time series of LFR anomalies and *Relative_diff* (values over the red lines) during 1991–
 532 1992. ***Relative_diff*** are relative differences of the global mean LFR anomalies between STD and Volca-off experiments
 533 calculated using the following equation.

534
$$\text{Relative_diff} = 100\% \times \frac{LFRA_{STD} - LFRA_{Volca-off}}{LFRA_{Volca-off}}$$
 (12)

535 In the equation, ***LFRA_{STD}*** represents global mean LFR anomalies simulated by STD-F1/F2 experiments. ***LFRA_{Volca-off}***
 536 denotes global mean LFR anomalies simulated by Volca-off-F1/F2 experiments. ***LFRA_{Volca-off}*** symbolizes global mean
 537 LFRs simulated by Volca-off-F1/F2 experiments.

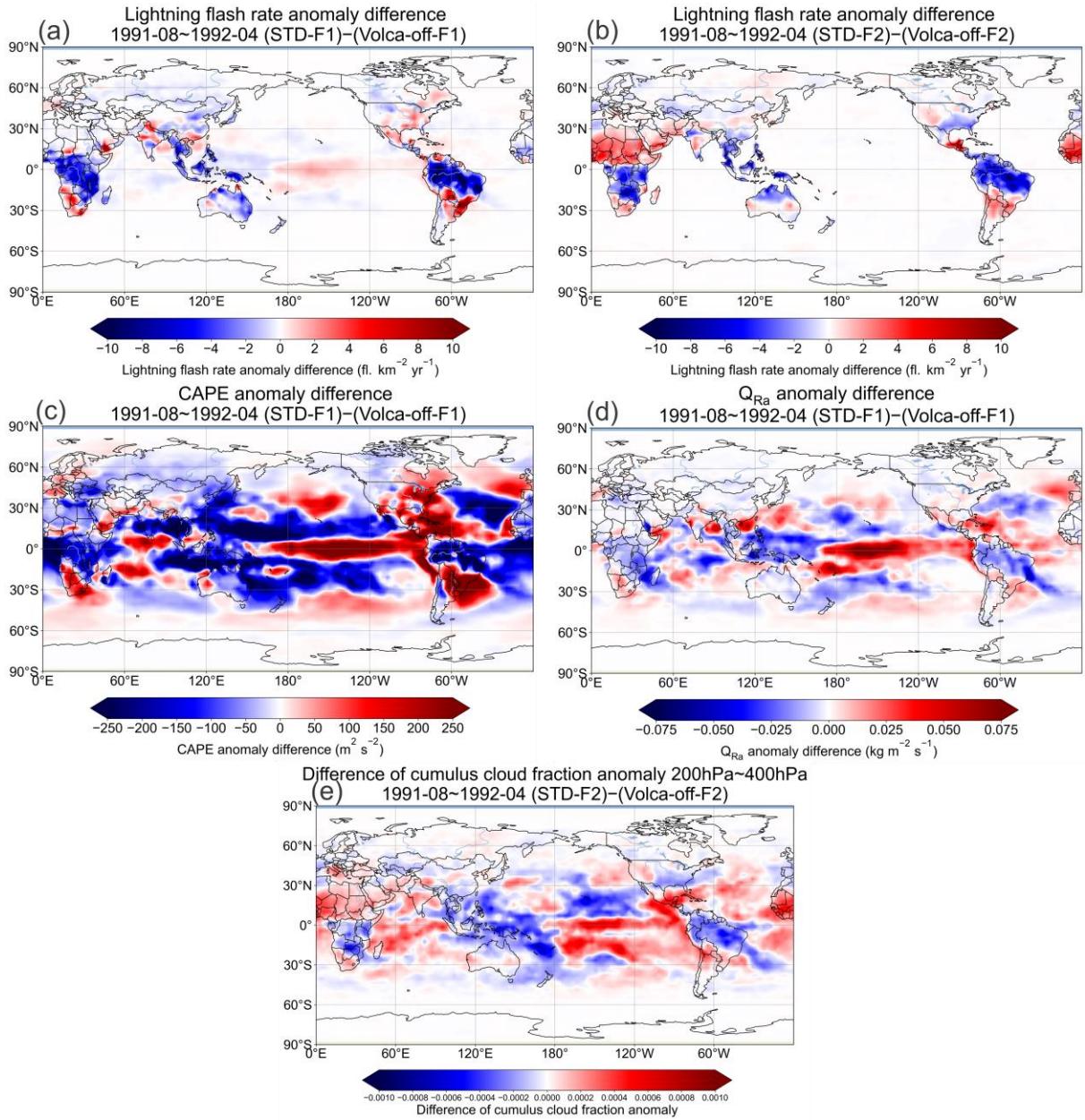


538

539 **Figure 11: Probability Density Distributions (PDDs) of *Relative_diff* obtained from monthly time-series data of *Relative_diff***
 540 **during 1990–1999 and 1991-06 – 1992-05 (a year after the Pinatubo eruption). The 1990–1999 *Relative_diff* for both lightning**
 541 **schemes are normally distributed with $N(\mu, \sigma^2)$ displayed in the titles of this figure. The 95% confidence interval of 1990–1999**
 542 ***Relative_diff* is also shown in the titles of this figure.**

543

544 The monthly time-series data of ***Relative_diff*** for 1990–1999 for both lightning schemes are calculated. The Probability
 545 Density Distributions (PDDs) of ***Relative_diff*** spanning 1990–1999 and 1991-06 – 1992-05 are displayed in Fig. 11. The
 546 1990–1999 ***Relative_diff*** presented in Fig. 11 (colored blue) are all normally distributed as determined by the
 547 Kolmogorov–Smirnov test. The 95% confidence interval of 1990–1999 ***Relative_diff*** is calculated and shown in the titles
 548 of Fig. 11. As displayed in Figs. 10c–10d, the underlined values (***Relative_diff***) exceeded the 95% confidence interval,
 549 indicating significant differences in the calculated global mean LFR anomalies by STD and Volca-off experiments. In other
 550 words, global lightning activities were suppressed significantly by the Pinatubo eruption during the first year after the
 551 eruption. The PDDs of 1991-06 – 1992-05 ***Relative_diff*** (colored green in Fig. 11) shifted to the left compared to the
 552 1990–1999 PDDs, indicating that global lightning activities were suppressed in the first year after the eruption.



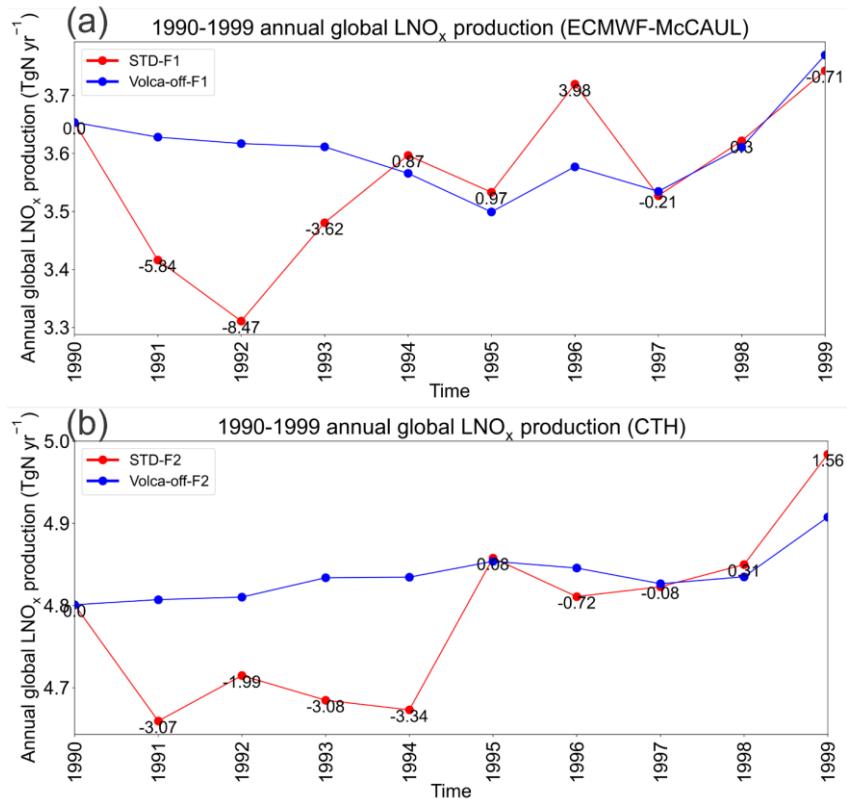
553

554 **Figure 12: 1991-08 – 1992-04 averaged LFR anomaly differences (a–b), CAPE anomaly differences (c), Q_{Ra} anomaly differences (d), and differences of 200 hPa – 400 hPa averaged cumulus cloud fraction anomaly between STD-F2 and Volca-off-F2 experiments (e) on the global map.**

557

558 Figures 12a–12b show 1991-08 – 1992-04 averaged LFR anomaly differences between STD and Volca-off experiments on 559 the global map. We found from Figs. 12a–12b that lightning activities are suppressed significantly within the three hotspots 560 of lightning activities (Central Africa, Maritime Continent, and South America) during 1991-08 – 1992-04, when the global

561 mean LFRs are found to be suppressed. To elucidate the potential reasons for the suppressed global lightning activities
 562 during the first year after the Pinatubo eruption, we first investigated the 1991-08 – 1992-04 averaged differences in CAPE
 563 and Q_{Ra} anomaly between STD-F1 and Volca-off-F1 (Figs. 12c–12d) because lightning densities are computed with CAPE
 564 and Q_{Ra} by the ECMWF-McCAUL scheme. Results showed that the Pinatubo eruption can engender apparent reductions of
 565 CAPE and Q_{Ra} within tropical and subtropical terrestrial regions (typically three hotspots of lightning activities) where
 566 lightning occurrence is frequent. These reductions constitute the main reason for the suppressed global lightning activities
 567 during the first year after the Pinatubo eruption simulated by the ECMWF-McCAUL scheme. We also examined the 1991-
 568 08 – 1992-04 averaged differences of 200 hPa – 400 hPa averaged cumulus cloud fraction anomaly between STD-F2 and
 569 Volca-off-F2 on the global map (Fig. 12e). The cumulus cloud fractions of each model layer are used to weight the
 570 calculated lightning densities from that layer by the CTH scheme, as explained in Sect. 2.2. As depicted in Fig. 12e and Fig.
 571 S6, the Pinatubo eruption led to marked reductions in the middle to upper tropospheric cumulus cloud fractions during 1991-
 572 08 – 1992-04 over three hotspots of lightning activities (Central Africa, Maritime Continent, and South America). As
 573 displayed in Fig. 6h, the cumulus that reached the middle to upper troposphere is related closely to lightning formation.
 574 Consequently, the simulated global lightning activities by the CTH scheme were also suppressed considerably during the
 575 first year after the Pinatubo eruption.



576

577 **Figure 13: 1990–1999 annual global LNO_x emissions calculated from the STD and Volca-off experiments' outputs simulated using**
578 **the ECMWF-McCAUL scheme (a) and the CTH scheme (b). Values over the red lines represent the relative differences (%)**
579 **between the red lines and blue lines, calculated with respect to the blue lines.**

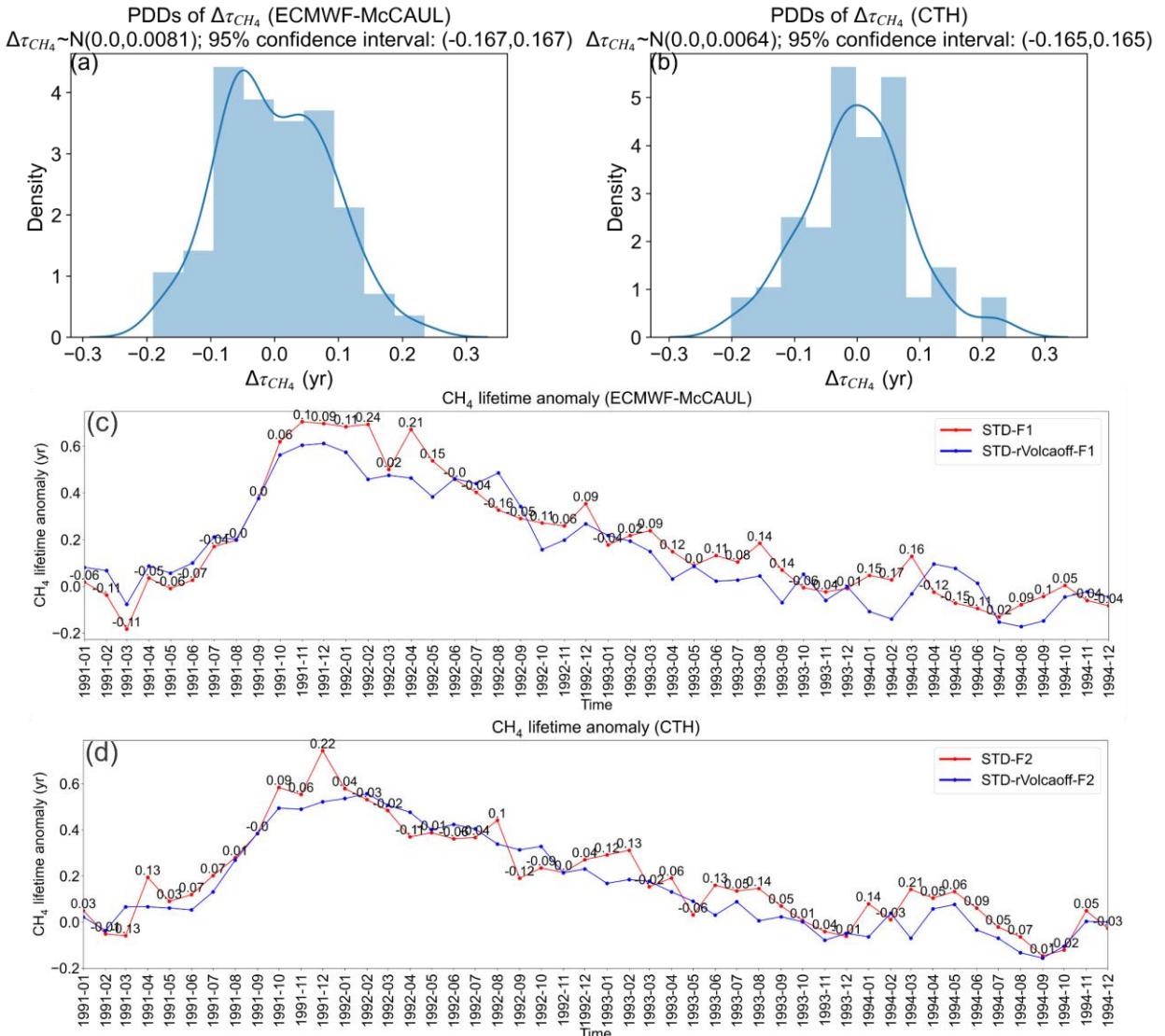
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581 Aside from the global lightning activity suppression described earlier, the production of LNO_x might also decrease after the
582 Pinatubo eruption. To explore this conjecture, we compared the LNO_x emissions in STD and Volca-off experiments (Fig. 13).
583 In the case of the ECMWF-McCAUL scheme, the reduction of LNO_x emissions caused by the Pinatubo eruption started in
584 1991 (5.84%) and continued until 1993, with the highest percentage reduction occurring in 1992 (8.47%) (Fig. 13a).
585 However, the CTH scheme showed a slightly different scenario of LNO_x emissions reduction after the Pinatubo eruption.
586 The LNO_x emissions are almost evenly reduced during 1991–1994 in the case of the CTH scheme (Fig. 13b). In conclusion,
587 our study indicates that the Pinatubo eruption can engender reductions in global LNO_x emissions, which last 2–3 years.
588 However, there exists some uncertainty in evaluating the magnitude of the reductions: from 1.99% to 8.47% for the annual
589 percentage reduction found from our study.

590

591 The simulated reduced global LNO_x emissions caused by the Pinatubo eruption might influence atmospheric chemistry
592 significantly. Most importantly, the reduced global LNO_x emissions might reduce OH radical production and extend the
593 global mean tropospheric lifetime of methane against tropospheric OH radical, abbreviated hereinafter as the methane
594 lifetime. We investigated this point further by comparing the methane lifetime anomaly simulated by STD and STD-
595 rVolcaoff experiments. As introduced in Sect. 2.5, the settings of STD-rVolcaoff experiments are the same as those use for
596 STD experiments, except that they use the daily LNO_x emission rates calculated from the Volca-off experiments. We
597 calculated the monthly CH₄ lifetime anomalies during 1990–1999 and $\Delta\tau_{CH_4}$ (the difference of CH₄ lifetime anomaly
598 between STD and STD-rVolcaoff experiments), which are shown in Figs. 14c–14d. Figures 14a–14b display the PDDs of
599 $\Delta\tau_{CH_4}$ monthly time series during 1990–1999. The $\Delta\tau_{CH_4}$ shown in Figs. 14a–14b are all normally distributed, as determined
600 using the Kolmogorov–Smirnov test. The 95% confidence interval of $\Delta\tau_{CH_4}$ is calculated and shown in the titles of Figs.
601 14a–14b. The annual global LNO_x production averaged during 1990–1999 is 3.56 TgN yr⁻¹ for STD-F1 and 4.79 TgN yr⁻¹
602 for STD-F2. At this level of annual global LNO_x production, we found that within the first two years after the Pinatubo
603 eruption, the $\Delta\tau_{CH_4}$ exceeded the 95% confidence interval simulated by both lighting schemes (1992-02 and 1992-04 in the
604 case of the ECMWF-McCAUL scheme; 1991-12 in the case of the CTH scheme). However, the widely cited range of annual
605 global LNO_x production is 2–8 TgN yr⁻¹ (Schumann and Huntrieser, 2007). Presuming that $\Delta\tau_{CH_4}$ responds linearly to the
606 LNO_x emission level, and that the annual global LNO_x production is 8 TgN yr⁻¹, then the extension of the CH₄ lifetime
607 because of the reduced LNO_x emissions can reach around 0.54 years for the ECMWF-McCAUL scheme. As a comparison,
608 ultraviolet shielding effects caused by stratospheric aerosols after the Pinatubo eruption led to the maximum increase of the
609 methane lifetime by about 0.6 years (Figs. 14c–14d).

610



611

612 **Figure 14:** Panels (a) and (b) show the Probability Density Distributions (PDDs) of $\Delta\tau_{CH_4}$ obtained from the monthly time series
 613 data of $\Delta\tau_{CH_4}$ during 1990–1999. $\Delta\tau_{CH_4}$ represents the difference in CH₄ lifetime anomaly between STD and STD-rVolcaoff
 614 experiments. The 95% confidence interval of $\Delta\tau_{CH_4}$ is also presented in the titles of panels (a)–(b). Panels (c) and (d) show monthly
 615 time series of CH₄ lifetime anomalies simulated by STD-F1/F2 and STD-rVolcaoff-F1/F2 experiments. Values over the red lines
 616 represent $\Delta\tau_{CH_4}$.

617 3.4 Model intercomparisons of LFR trends with CMIP6 model outputs

618 The historical lightning trends demonstrated in our study are undoubtedly worth comparing with the results of other
 619 chemistry–climate models or Earth system models. As introduced in Sect. 2.4, for comparison of the simulated LFR trends

620 and variations in our study with those of other CMIP6 models' outputs, we used all available LFR data from the CMIP6
621 CMIP Historical experiments from CESM2-WACCM (3 ensembles) (Danabasoglu, 2019), GISS-E2-1-G (9 ensembles)
622 (Kelley et al., 2020), and UKESM1-0-LL (18 ensembles) (Tang et al., 2019). Table S1 presents a complete list of the
623 ensemble members we used. It is noteworthy that the LFR data obtained from the three CMIP6 models described earlier are
624 calculated using the CTH scheme. The results of model intercomparisons of LFR trends and variations are displayed in Fig.
625 15.

626 As displayed in Figs. 15a–15b and Table 6, both the ECMWF-McCAUL and the CTH schemes (STD-F1/F2) simulated
627 almost flat global lightning trends (even the trend is estimated to be significant in the case of the CTH scheme (0.03 \% yr^{-1})),
628 but the ensemble mean obtained from another three CMIP6 models exhibit much larger significant increasing global
629 lightning trends (trends from 0.11 \% yr^{-1} to 0.25 \% yr^{-1}). Many reasons underlie the differences in global lightning trends
630 simulated by CHASER in our study and by the three CMIP6 models, including the use of different methods to determine
631 SSTs/sea ice fields. Instead of using a coupled Atmosphere–Ocean general circulation model to calculate SSTs/sea ice fields
632 dynamically in the three CMIP6 models, CHASER uses the prescribed HadISST data (Rayner et al., 2003), which are based
633 on plenty of observational data. Changes in the global mean sea surface temperature anomaly during 1960–2014 (ΔSST)
634 obtained from STD-F1/F2 and CMIP6 model outputs are presented in Table 5. We also used the observation-based Extended
635 Reconstructed SST (ERSST) dataset (Huang et al., 2017) constructed by NOAA to evaluate the ΔSST obtained from
636 different models. The ΔSST calculated from ERSST during 1960–2014 is 0.549°C , which most closely approximates the
637 ΔSST obtained from STD-F1/F2. Considered from the perspective of SSTs/sea ice fields alone, the results (global lightning
638 trends) of our study are expected to be closer to the actual situation.

639

640 Actually, the three CMIP6 models simulated stronger global warming during 1960–2014 than CHASER in our study, as
641 displayed in Fig. S7 and Table 5. The CTH scheme is reported to respond positively to simulated global warming (Price and
642 Rind, 1994; Zeng et al., 2008; Hui and Hong, 2013; Banerjee et al., 2014; Krause et al., 2014; Clark et al., 2017). The
643 simulated stronger global warming by the three CMIP6 models is regarded as responsible for differences in simulated global
644 lightning trends between our study and the three CMIP6 models (Figs. 15a–15b and Table 6). We further investigated the
645 sensitivities of the global mean LFR anomaly change to the global mean surface temperature anomaly increase ($\text{ \% }^\circ\text{C}^{-1}$)
646 obtained from CHASER and the three CMIP6 models. The sensitivities in percentage per degree Celsius are presented in
647 Table 5. Overall, even when using the same CTH scheme, the sensitivities ($\Delta\text{LFR}/\Delta\text{TS}$) simulated by the three CMIP6
648 models are higher than that simulated by CHASER in our study. This different sensitivity might be partially attributable to
649 the nonlinear relation between lightning response and climate change (Pinto, 2013; Krause et al., 2014). Compared to the
650 CTH scheme, the ECMWF-McCAUL scheme simulated a statistically non-significant negative sensitivity ($\Delta\text{LFR}/\Delta\text{TS}$),
651 which is attributable to the stronger suppression of positive global lightning trends caused by increases in AeroPEs simulated
652 using the ECMWF-McCAUL scheme.

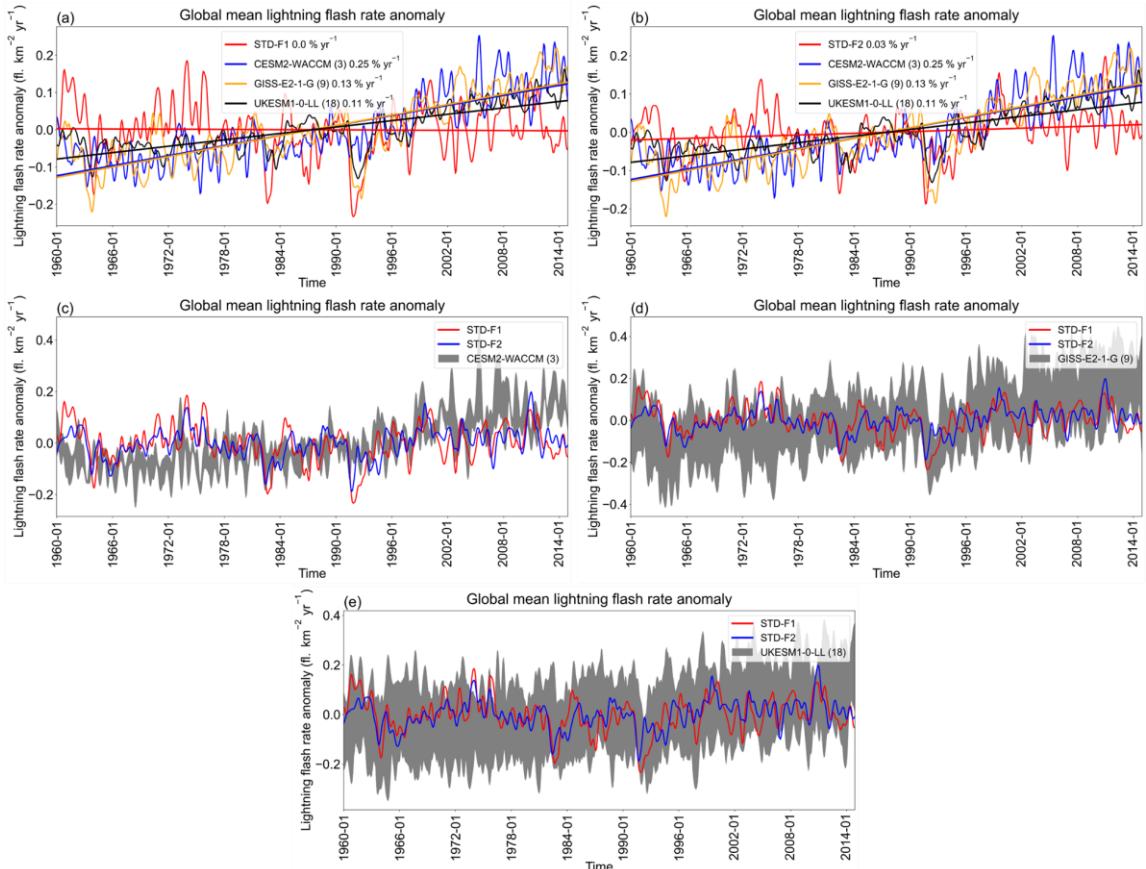
653

654 **Table 5: Changes in global mean surface temperature anomaly (ΔTS), global mean sea surface temperature anomaly (ΔSST),
655 global mean lightning flash rate anomaly (ΔLFR), and the rate of change of LFR anomaly corresponding to each degree-Celsius
656 increase in global mean surface temperature anomaly ($\Delta LFR/\Delta TS$) obtained from STD-F1/F2 and CMIP6 model outputs. The
657 change of ΔSST obtained from the ERSST dataset is also shown in this Table. Changes were obtained by calculating the difference
658 between the rightmost and leftmost points of the approximating curve for the 1960–2014 time-series data.**

Model/experiment/dataset	ΔTS (°C)	ΔSST (°C)	ΔLFR (%)	$\Delta LFR/\Delta TS$ (% °C ⁻¹)
STD-F1	0.593	0.428	-0.272	-0.46
STD-F2	0.563	0.432	1.497	2.66
CESM2-WACCM	1.245	1.077	13.758	11.05
GISS-E2-1-G	0.810	0.677	7.248	8.95
UKESM1-0-LL	1.141	0.999	5.942	5.21
ERSST	—	0.549	—	—

659

660 Figures 15d–15e affirm that the global lightning variation simulated by our study is basically within the full ensemble range
661 of GISS-E2-1-G and UKESM1-0-LL. After the Pinatubo eruption, as described in Sect. 3.3 of this report, the GISS-E2-1-G
662 and UKESM1-0-LL models also manifest significant suppression of global lightning activities, but the CESM2-WACCM
663 model shows no such phenomenon. The commonalities and differences in global lightning trends and variations found in the
664 model intercomparisons imply that great uncertainties existed in past (1960–2014) global lightning trend simulations. Such
665 uncertainties deserve to be investigated further.



666

667 **Figure 15: Comparisons of simulated global mean LFR anomalies found in our study (CHASER) and found using other CMIP6
668 models. All the figures are created based on the monthly time-series data of global mean LFR anomalies with a 1-D Gaussian
669 (Denoising) filter applied. For CMIP6 models, the ensemble mean is shown as the solid line, and the full ensemble range is shown
670 as grey shading (c–e). Fitting curves and the trends of fitting curves ($\% \text{ yr}^{-1}$) are also given in (a–b).**

671

672 **Table 6: A statistical summary of the trends shown in Figs. 15a–15b by Mann–Kendall rank statistic and Sen’s slope estimator.**
673 The time-series data of global mean LFR anomalies were estimated by Mann–Kendall rank statistic and Sen’s slope estimator. The
674 column “Trend” shows whether these are significant trends with the significance set as 5%, as well as the percentage trends in %
675 yr^{-1} estimated by linear regression. The “ p -value” is calculated during Mann-Kendall trend test. “Slope” shows Sen’s slope of
676 trend. Q_{\min} and Q_{\max} respectively denote the lower and upper limits of the 95% confidence interval of Sen’s slope.

677

Experiment/model	Trend	p -value	Slope	Q_{\min}	Q_{\max}
STD-F1	No trend, 0.0 % yr^{-1}	$p > 0.05$	0.0	-0.0001	0.0
STD-F2	Increasing, 0.03 % yr^{-1}	$p < 0.01$	0.0001	0.0	0.0001
CESM2-WACCM	Increasing, 0.25 % yr^{-1}	$p < 0.01$	0.0004	0.0003	0.0004

GISS-E2-1-G	Increasing, 0.13 % yr ⁻¹	<i>p</i> < 0.01	0.0004	0.0004	0.0004
UKESM1-0-LL	Increasing, 0.11 % yr ⁻¹	<i>p</i> < 0.01	0.0002	0.0002	0.0003

678 4 Discussion and Conclusions

679 We used two lightning schemes (the CTH and ECMWF-McCAUL schemes) to study historical (1960–2014) lightning–
 680 LNO_x trends and variations and their influencing factors (global warming, increases in AeroPEs, and Pinatubo eruption)
 681 within the CHASER (MIROC) chemistry–climate model. The CTH scheme, which is the most widely used lightning scheme,
 682 nevertheless lacks a direct physical link with the charging mechanism. The ECMWF-McCAUL scheme is a newly
 683 developed process-based/ice-based lightning scheme with a direct physical link to the charging mechanism.

684

685 With only the aerosol radiative effects considered in the lightning–aerosols interaction, both lightning schemes simulated
 686 almost flat trends of global mean LFR during 1960–2014 (no trend is detected in the case of the ECMWF-McCAUL scheme,
 687 but a slightly significant increasing trend is detected in the case of the CTH scheme). Reportedly, because the aerosol
 688 microphysical effects can enhance lightning activities (Yuan et al., 2011; Wang et al., 2018; Liu et al., 2020), our study
 689 might underestimate the increasing trend of global mean LFR (our study only considered the aerosol radiative effects in
 690 aerosol–lightning interactions). Further research is anticipated, with consideration of the effects of aerosol microphysical
 691 effects on long-term lightning trends. Moreover, both lightning schemes manifest that past global warming enhances the
 692 historical trend of global mean lightning density toward the positive direction (around 0.03% yr⁻¹ or 3% K⁻¹). However, past
 693 increases in AeroPEs exert the opposite effect to the lightning trend (-0.07% yr⁻¹ – -0.04% yr⁻¹). The effects of the increased
 694 AeroPEs on the lightning trend only over land regions expand to -0.10% yr⁻¹ – -0.05% yr⁻¹, which implies that the effects are
 695 more significant over land regions. We obtained similar results for the historical global LNO_x emissions trend, which
 696 indicates that historical global warming and increases in AeroPEs can affect atmospheric chemistry and engender feedback
 697 by influencing LNO_x emissions. Although the CTH and ECMWF-McCAUL schemes use different parameters to simulate
 698 lightning, both lightning schemes indicate that the enhanced global convective activity under global warming is the main
 699 reason for the increase in lightning–LNO_x emissions. In contrast, the increases in AeroPEs have decreased lightning–LNO_x
 700 emissions by weakening the convective activity in the lightning hotspots. By analyzing the simulation results on the global
 701 map, we also found that the effects of historical global warming and increases in AeroPEs on lightning trends are
 702 heterogeneous across different regions. Our results indicate that historical global warming enhances lightning activities
 703 within the Arctic region and Japan but suppresses lightning activities around New Zealand and some parts of the Southern
 704 Ocean. Both lightning schemes demonstrated that the historical increases in AeroPEs suppress lightning activities in some
 705 parts of the Southern Ocean and South America. The ECMWF-McCAUL scheme also suggests that historical increases in
 706 AeroPEs suppress lightning activities in some parts of India and China when only the aerosol radiative effects are considered.

707 This finding is plausible because both countries experienced dramatic increases in AeroPEs during 1960–2014 because of
708 rapid economic growth.

709

710 Furthermore, this report is the first describing significant suppression of global lightning activity during the first year after
711 the Pinatubo eruption, which is indicated in both lightning schemes (global lightning activities decreased by up to 18.10%
712 simulated by the ECMWF-McCAUL scheme). This finding is mainly attributable to the Pinatubo eruption weakening of the
713 convective activities within the hotspots of lightning, which in turn decreased Q_{Ra} and middle-level to high-level cumulus
714 cloud fractions in these regions. The simulation results also indicate that the Pinatubo eruption can engender reductions in
715 global LNO_x emissions, which last 2–3 years. However, some uncertainty exists in evaluating magnitude of these reductions
716 (from 1.99% to 8.47% for the annual percentage reduction in our study). The case study of the Pinatubo eruption in our
717 research indicates that other large-scale volcanic eruptions can also engender significant reduction of global lightning
718 activities and global-scale LNO_x emissions.

719

720 Lastly, we compared the global lightning trends demonstrated in our study with the outputs of three CMIP6 models:
721 CESM2-WACCM, GISS-E2-1-G, and UKESM1-0-LL. We used all available LFR data from the CMIP6 CMIP historical
722 experiments from the three models described above. The three CMIP6 models suggest significant increasing trends in
723 historical global lightning activities, which differs from the findings of our study in the magnitude of lightning trends. Unlike
724 the three CMIP6 models which use a coupled Atmosphere–Ocean general circulation model to calculate SSTs/sea ice fields
725 dynamically, our study (CHASER) uses the prescribed HadISST SSTs/sea ice data, which more closely reflect the actual
726 situation. Therefore, we believe that the results (the historical global lightning trends) obtained from our study (CHASER)
727 more closely approximate the actual situation. However, model intercomparisons of global lightning trends still indicate that
728 considerable uncertainties exist in historical (1960–2014) global lightning trend simulations, and that such uncertainties
729 deserve further investigation.

730 **Code availability**

731 The source code for CHASER to reproduce results obtained from this work is obtainable from the repository at
732 <https://doi.org/10.5281/zenodo.5835796> (He et al., 2022a).

733 **Data availability**

734 The LIS/OTD data used for this study are available from <https://ghrc.nsstc.nasa.gov/hydro/?q=LRTS> (last access: 11 January
735 2022). The CMIP6 model outputs (LFR and surface temperature) used for this study are available from

736 <https://aims2.llnl.gov/search> (last access: 1 February 2023). The Extended Reconstructed SST data used for this study are
737 available from <https://www.ncei.noaa.gov/products/extended-reconstructed-sst> (last access: 27 March 27 2023).

738 **Author contributions**

739 YFH conducted all simulations, interpreted the results, and wrote the manuscript. KS developed the CHASER (MIROC)
740 model code, conceived the presented idea, and supervised the findings of this work and the manuscript preparation.

741 **Competing interests**

742 The authors declare that they have no conflict of interest.

743 **Acknowledgments**

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