

1 **Introducing new lightning schemes into the CHASER (MIROC)**

2 **chemistry climate model**

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7 **Abstract.** The formation of nitrogen oxides (NO_x) associated with lightning activities (hereinafter designated as LNO_x) is a
8 major source of NO_x. In fact, it is regarded as the dominant NO_x source in the middle to upper troposphere. Therefore,
9 improving the prediction accuracy of lightning and LNO_x in chemical climate models is crucially important. This study
10 implemented three new lightning schemes with the CHASER (MIROC) global chemical transport/climate model. The first
11 lightning scheme is based on upward cloud ice flux (ICEFLUX scheme). The second one (the original ECMWF scheme), also
12 adopted in the European Centre for Medium-Range Weather Forecasts (ECMWF) forecasting system, calculates lightning
13 flash rates as a function of Q_R (a quantity intended to represent the charging rate of collisions between graupel and other types
14 of hydrometeors inside the charge separation region), convective available potential energy (CAPE), and convective cloud-
15 base height. For the original ECMWF scheme, by tuning the equations and adjustment factors for land and ocean, a new
16 lightning scheme named ECMWF-McCAUL scheme was also tested in CHASER. The ECMWF-McCAUL scheme calculates
17 lightning flash rates as a function of CAPE and column precipitating ice. In the original version of CHASER (MIROC),
18 lightning is initially parameterized with the widely used cloud top height scheme (CTH scheme). Model evaluations with
19 lightning observations conducted using the Lightning Imaging Sensor (LIS) and Optical Transient Detector (OTD) indicate
20 that both the ICEFLUX and ECMWF schemes simulate the spatial distribution of lightning more accurately on a global scale
21 than the CTH scheme does. The ECMWF-McCAUL scheme showed the highest prediction accuracy for the global distribution
22 of lightning. Evaluation by atmospheric tomography (ATom) aircraft observations (NO) and tropospheric monitoring
23 instrument (TROPOMI) satellite observations (NO₂) shows that the newly implemented lightning schemes partially facilitated
24 the reduction of model biases (NO and NO₂) typically within the regions where LNO_x is the major source of NO_x when
25 compared using the CTH scheme. Although the newly implemented lightning schemes have a minor effect on the tropospheric
26 mean oxidation capacity compared to the CTH scheme, they led to marked changes of oxidation capacity in different regions
27 of the troposphere. Historical trend analyses of flash and surface temperatures predicted using CHASER (2001–2020) show
28 that lightning schemes predicted an increasing trend of lightning or no significant trends, except for one case of the ICEFLUX
29 scheme, which predicted a decreasing trend of lightning. The global lightning rates of increase during 2001–2020 predicted
30 by the CTH scheme were 17.69%/°C and 2.50%/°C, respectively, with and without meteorological nudging. The un-nudged
31 runs also included the short-term surface warming but without the application of meteorological nudging. Furthermore, the
32 ECMWF schemes predicted a larger increasing trend of lightning flash rates under the short-term surface warming by a factor
33 of 4 (ECMWF-McCAUL scheme) and 5 (original ECMWF scheme) compared to the CTH scheme without nudging. In
34 conclusion, the three new lightning schemes improved global lightning prediction in the CHASER model. However, further
35 research is needed to assess the reproducibility of trends of lightning over longer periods.

36 **Keywords**

37 lightning, lightning scheme, lightning NO_x, chemistry-climate model, lightning under climate change

38 1 Introduction

39 Nitric oxide (NO) can be formed during lightning activities. Also, NO can be oxidized quickly to nitrogen dioxide (NO₂). An
40 equilibrium between NO and NO₂ can be reached during daytime. Those gases are known collectively as NO_x (Finney et al.,
41 2014). Actually, LNO_x is estimated as contributing approximately 10% of the global NO_x source. Regarded as the dominant
42 NO_x source in the middle to upper troposphere (Schumann and Huntrieser, 2007; Finney et al., 2016a), NO_x is associated
43 with many chemical reactions in the atmosphere. Most importantly, NO reacts with peroxy radical to reproduce OH radical.
44 Photochemical dissociation of NO₂ engenders the production of ozone (Isaksen and Hov, 1987; Grewe, 2007). The primary
45 oxidants in the atmosphere, which are OH radical and ozone, control the oxidation capacity of the atmosphere. Results of
46 several studies have indicated that global-scale LNO_x emissions are an important contributor to ozone and other trace gases,
47 especially in the upper troposphere (Labrador et al., 2005; Wild, 2007; Liaskos et al., 2015). Consequently, LNO_x influences
48 atmospheric chemistry and global climate to a considerable degree (Schumann and Huntrieser, 2007; Murray, 2016; Finney
49 et al., 2016b; Tost, 2017). However, large uncertainties remain in predicting lightning and LNO_x in chemical climate models
50 (Tost et al., 2007). Therefore, improving lightning prediction accuracy and quantifying LNO_x in chemical climate models is
51 crucially important for future atmospheric research.

52

53 Global chemical climate models (CCMs) such as CHASER (MIROC) (Sudo et al., 2002; Sudo and Akimoto, 2007;
54 Watanabe et al., 2011) most often use the convective cloud-top height to parameterize the lightning flash rate (Price and
55 Rind, 1992; Lamarque et al., 2013). The Earth System Models (ESMs) recently used in the sixth Coupled Model
56 Intercomparison Project (CMIP6) all used the convective cloud-top height to calculate the lightning flash rates (Thornhill et
57 al., 2021). Not only in global CCMs but the studies of LNO_x in regional-scale models have also made significant progress in
58 recent years (Heath et al., 2016; Kang et al., 2019a; Kang et al., 2019b; Kang et al., 2020).

59

60 The spaceborne Lightning Imaging Sensor (LIS) and Optical Transient Detector (OTD) lightning observation data (Cecil et
61 al., 2014) are often utilized to evaluate the performance of different lightning schemes. A new lightning scheme proposed by
62 Finney et al. (2014), which is based on upward cloud ice flux, has shown better spatial and temporal correlation coefficients
63 and root mean square errors (RMSEs) than the cloud top height scheme compared against the LIS/OTD lightning
64 observations. Another lightning scheme also showed more accurate lightning prediction than the cloud top height scheme,
65 which is also adopted in the ECMWF forecasting system (Lopez, 2016). This lightning scheme uses Q_R (a quantity intended
66 to represent the charging rate of collisions between graupel and other types of hydrometeors inside the charge separation
67 region), convective available potential energy (CAPE), and convective cloud-base height to compute the lightning flash rate
68 (Lopez, 2016). The two new lightning schemes (Finney et al., 2014; Lopez 2016) mentioned above have only been evaluated
69 in a few chemical transport and climate models. The new lightning schemes are expected to be evaluated and compared in
70 more chemical transport and climate models, such as CHASER. To achieve better prediction accuracy for lightning and
71 better quantification of LNO_x in chemical climate models, comparing and optimizing the existing lightning schemes and
72 evaluating them with various observation data are also important.

73

74 Lightning simulations are also fundamentally important in chemical climate model studies for predictions of atmospheric
75 chemical fields and climate. Nevertheless, different lightning schemes respond very differently on decadal to multi-decadal
76 time scales under global warming. Some lightning schemes such as those using cloud top height or CAPE \times precipitation
77 rate as a proxy for calculating lightning indicate that lightning increases concomitantly with increasing global average
78 temperature. By contrast, other lightning schemes, such as those using convective mass flux or upward cloud ice flux as a
79 proxy of lightning, indicate that lightning will decrease as the global average temperature increases (Clark et al., 2017;
80 Finney et al., 2018). Several studies (Price and Rind 1994; Zeng et al., 2008; Jiang and Liao 2013; Banerjee et al., 2014;

81 Krause et al., 2014; Clark et al., 2017) have found 5–16% increases in lightning flashes per degree of increase in global
82 mean surface temperatures with the lightning scheme based on cloud top height. Over the contiguous United States
83 (CONUS), the CAPE \times precipitation rate proxy predicted a $12 \pm 5\%$ increase in the CONUS lightning flash rate per degree
84 of global mean temperature increase (Romps et al., 2014). Compared to the findings reported by Romps et al. (2014), Finney
85 et al. (2020) found a relatively small response of lightning to climate change ($2\% \text{ K}^{-1}$) over Africa using a cloud-ice-based
86 parametrisation for lightning. By contrast, Finney et al. (2018) found a 15% global mean lightning flash rate decrease with
87 the lightning scheme based on upward cloud ice flux in 2100 under a strong global warming scenario. Furthermore, a 2.0%
88 decrease in global mean lightning flashes per degree of increase in the global mean surface temperature with the lightning
89 scheme based on convective mass flux has been reported by Clark et al. (2017). Although it remains unclear which lightning
90 scheme is best suited to predicting future lightning (Romps, 2019), comparing different lightning schemes in different
91 chemical climate models is valuable for consideration of the sensitivity of lightning to global warming.

92
93 This study introduced three new lightning schemes into CHASER (MIROC). The first lightning scheme (Finney et al., 2014)
94 is based on upward cloud ice flux. The second one (Lopez, 2016), also adopted in the ECMWF forecasting system,
95 calculates lightning flash rates as a function of Q_R (defined in Sect. 2.2), CAPE, and convective cloud-base height. In the
96 case of the second lightning scheme, by tuning the equations and adjustment factors based on a study reported by McCaul et
97 al. (2009), a new lightning scheme named ECMWF-McCAUL scheme was also tested for CHASER (MIROC). The
98 ECMWF-McCAUL scheme calculates lightning flash rates as a function of CAPE and column precipitating ice. Our study
99 conducted detailed evaluation of lightning and LNO_x by LIS/OTD lightning observations, NASA/ATom aircraft
100 observations, and TROPOMI satellite observations. The effects of different lightning schemes on the atmospheric chemical
101 fields were evaluated. Also, 20-year (2001–2020) historical trend analyses of lightning densities and LNO_x emissions
102 simulated by different lightning schemes were conducted. Based on the results, the effects of LNO_x emissions during 2001–
103 2020 on tropospheric NO_x and O₃ column trends were estimated and discussed.

104
105 Research methods, including the model description and experiment setup, are described in Sect. 2. In Sect. 3.1, the
106 evaluation of lightning schemes using LIS/OTD lightning observations is explained. In Sect. 3.2, LNO_x emission simulation
107 by different lightning schemes is evaluated with aircraft and satellite measurements. Section 3.3 presents a discussion of the
108 effects of different lightning schemes on the atmospheric chemical fields. Historical trends of lightning simulated by
109 different lightning schemes are analyzed and discussed in Sect. 3.4. Section 3.5 discussed how LNO_x emissions (2001–2020)
110 trends influence the tropospheric NO_x and O₃ column trends. Section 4 presents the discussions and conclusions of this
111 study.

112 **2 Method**

113 **2.1 Chemistry-climate model**

114 The model used for this study is the CHASER (MIROC) global chemical transport and climate model (Sudo et al., 2002;
115 Sudo and Akimoto, 2007; Watanabe et al. 2011; Ha et al., 2021), which incorporates consideration of detailed chemical and
116 transport processes in the troposphere and stratosphere. CHASER calculates the distributions of 94 chemical species and
117 reflects the effects of 269 chemical reactions (58 photolytic, 190 kinetic, 21 heterogeneous). Its tropospheric chemistry
118 incorporates consideration of Non-Methane Hydrocarbons (NMHC) oxidation and the fundamental chemical cycle of O_x–
119 NO_x–HO_x–CH₄–CO. Its stratospheric chemistry simulates chlorine-containing and bromine-containing compounds,
120 chlorofluorocarbons (CFCs), hydrofluorocarbons (HFCs), carbonyl sulfide (OCS), and N₂O. Furthermore, it simulates the
121 formation of polar stratospheric clouds (PSCs) and heterogeneous reactions on their surfaces. CHASER is coupled to the

122 MIROC AGCM ver. 5.0 (Watanabe et al., 2011). Grid-scale large-scale condensation and cumulus convection (Arakawa–
 123 Schubert scheme) are used to simulate cloud/precipitation processes. Aerosol chemistry is coupled with the SPRINTARS
 124 aerosol model (Takemura et al., 2009), which is also based on MIROC.
 125
 126 For this study, horizontal resolution used is T42 ($2.8^\circ \times 2.8^\circ$), with vertical resolution of 36 σ -p hybrid levels from the
 127 surface to approximately 50 km. The AGCM meteorological fields (u , v , T) simulated by MIROC were nudged towards the
 128 six-hourly NCEP FNL data (<https://rda.ucar.edu/datasets/ds083.2/>, last access: 6 December 2021). Anthropogenic precursor
 129 emissions such as NO_x , CO , O_3 , SO_2 , and VOCs were obtained from the HTAP-II inventory for 2008
 130 (https://edgar.jrc.ec.europa.eu/dataset_htap_v2, last access: 6 December 2021), with biomass burning emissions from
 131 MACC-GFAS (Inness et al., 2013). The monthly soil NO_x emissions used in CHASER (MIROC) are constant for each year
 132 and are derived from Yienger and Levy (1995).

133 2.2 Lightning NO_x emission scheme

134 The lightning flash rate in CHASER is originally parameterized by cloud-top height (Price and Rind, 1992, 1993), with a “C-
 135 shaped” NO_x vertical profile adopted (Pickering et al., 1998). The equations used to calculate the lightning flash rate by
 136 cloud-top height are
 137 $F_l = 3.44 \times 10^{-5} H^{4.9}$ (1)
 138 $F_o = 6.2 \times 10^{-4} H^{1.73}$ (2)
 139 where F represents the total flash frequency (fl. min^{-1}), H stands for the cloud-top height (km), and subscripts l and o
 140 respectively denote the land and ocean (Price and Rind, 1992).

141
 142 For this study, three new lightning schemes are implemented into CHASER (MIROC). One is based on upward cloud ice
 143 flux. It calculates the lightning flash rate by the following equations, as described by Finney et al. (2014).

$$144 f_l = 6.58 \times 10^{-7} \phi_{ice} \quad (3)$$

$$145 f_o = 9.08 \times 10^{-8} \phi_{ice} \quad (4)$$

146 Therein, f_l and f_o respectively represent the flash density (fl. $\text{m}^{-2} \text{s}^{-1}$) over land and ocean. Also, ϕ_{ice} is the upward cloud ice
 147 flux at 440 hPa ($\text{kg}_{ice} \text{m}_{cloud}^{-2} \text{s}^{-1}$) as calculated using

$$148 \phi_{ice} = \frac{q \times \Phi_{mass}}{c}, \quad (5)$$

149 where q denotes the specific cloud ice water content at 440 hPa ($\text{kg}_{ice} \text{kg}_{air}^{-1}$), Φ_{mass} stands for the updraught mass flux at 440
 150 hPa ($\text{kg}_{air} \text{m}_{cell}^{-2} \text{s}^{-1}$), and c represents the fractional cloud cover at 440 hPa ($\text{m}_{cloud}^2 \text{m}_{cell}^{-2}$). The 440 hPa pressure level is chosen
 151 because it is a representative pressure level of fluxes in deep convective clouds (Finney et al., 2014). Moreover, Romps
 152 (2019) has proposed an alternative approach to applying the ICEFLUX scheme by using the upward cloud ice flux at 260-K
 153 isotherms instead of at 440 hPa isobars. As suggested by Romps (2019), the isotherm-alternative is more appropriate for
 154 climate change simulations because the charge separation zone will follow the isotherms instead of the isobars with climate
 155 change. The 260-K isotherm is chosen because it is close to the 440 hPa isobar based on a present-day tropical sounding and
 156 it lies within the mixed-phase regions of clouds (Romps, 2019). To distinguish the two different approaches to applying the
 157 ICEFLUX scheme, the isobar approach is abbreviated as ICEFLUX_P and the isotherm-alternative is abbreviated as
 158 ICEFLUX_T.

159
 160 Another new lightning scheme, also adopted in the ECMWF forecasting system, calculates lightning flash rates as a function
 161 of the Q_R (defined in equation 8), $CAPE$, and convective cloud-base height (Lopez, 2016) as

$$162 f_T = \alpha Q_R \sqrt{CAPE} \min(z_{base}, 1800)^2, \quad (6)$$

163 where f_T is the total lightning flash density (fl. $\text{m}^{-2} \text{s}^{-1}$), z_{base} is the convective cloud-base height in m, α (fl. $\text{kg}^{-1} \text{m}^{-3}$) is a
 164 constant obtained after calibration against the LIS/OTD climatology, which is set to 1.11×10^{-15} in this study. As explained by
 165 Lopez (2016), the number 1800 used in equation (6) is a constraint to let the term z_{base} remains constant after it exceeds
 166 1800 m. Note that the equation (6) is standardized on base SI units. $CAPE$ ($\text{m}^2 \text{s}^{-2}$) is diagnosed from the following
 167 equation.

$$168 \quad CAPE = \int_{z_{LFC}}^{z_{w=0}} \max \left(g \frac{T_v^u - \bar{T}_v}{\bar{T}_v}, 0 \right) dz \quad (7)$$

169 In that equation, g is the constant of gravity. Also, T_v^u and \bar{T}_v respectively denote the virtual temperatures in the updraft and
 170 the environment. The integral in equation (7) starts at the level of free convection z_{LFC} and stops at the level at which
 171 negative buoyancy is found ($w = 0$). Quantity Q_R (kg m^{-2}) is intended to represent the charging rate of collisions between
 172 graupel and other types of hydrometeors inside the charge separation region. It is empirically calculated as

$$173 \quad Q_R = \int_{z_0}^{z_{-25}} q_{graup} (q_{cond} + q_{snow}) \bar{\rho} dz, \quad (8)$$

174 where z_0 and z_{-25} signify the heights (m) of the 0° and -25°C isotherms, and q_{cond} denotes the mass mixing ratio of cumulus
 175 cloud liquid water (kg kg^{-1}). The respective amounts of graupel (q_{graup} ; kg kg^{-1}) and snow (q_{snow} ; kg kg^{-1}) are computed
 176 from the following equations for each vertical level of the model.

$$177 \quad q_{graup} = \beta \frac{P_f}{\bar{\rho} V_{graup}} \quad (9)$$

$$178 \quad q_{snow} = (1 - \beta) \frac{P_f}{\bar{\rho} V_{snow}} \quad (10)$$

179 In those equations, P_f denotes the vertical profile of the frozen precipitation convective flux ($\text{kg m}^{-2} \text{s}^{-1}$), $\bar{\rho}$ stands for the
 180 environmental air density (kg m^{-3}), and V_{graup} and V_{snow} respectively express the typical fall speeds for graupel and snow set
 181 to 3.0 and 0.5 m s^{-1} . The dimensionless coefficient β is set as 0.7 for land and 0.45 for ocean to account for the observed
 182 lower graupel contents over oceans.

183

184 For the original ECMWF scheme, by tuning the calculation equations based on findings reported by McCaul et al. (2009),
 185 and the adjustment factors for land and ocean, the lightning prediction accuracy is improved further, as explained in Sect.
 186 3.1. We named the new lightning scheme as ECMWF-McCAUL scheme, and it simulates the lightning flash rate by the
 187 following equations.

$$188 \quad f_l = \alpha_l Q_{Ra} CAPE^{1.3} \quad (11)$$

$$189 \quad f_o = \alpha_o Q_{Ra} CAPE^{1.3} \quad (12)$$

190 Therein, f_l and f_o respectively denote the total flash density (fl. $\text{m}^{-2} \text{s}^{-1}$) over land and ocean. Also, α_l and α_o are constants
 191 (fl. $\text{s}^{1.6} \text{kg}^{-1} \text{m}^{-2.6}$) obtained after calibration against LIS/OTD climatology, respectively, for land and ocean. For this
 192 study, α_l and α_o are set respectively to 2.67×10^{-16} and 1.68×10^{-17} . Then $CAPE$ is computed in the same way as the
 193 original ECMWF scheme. In addition, Q_{Ra} (kg m^{-2}) is a proxy for the charging rate resulting from the collisions between
 194 graupel and hydrometeors of other types inside the charge separation region (from 0° to -25°C isotherm), as reported by
 195 McCaul et al. (2009). Also, Q_{Ra} represents the total volumetric amount of precipitating ice in the charge separation region,
 196 calculated as

$$197 \quad Q_{Ra} = \int_{z_0}^{z_{-25}} (q_{graup} + q_{snow} + q_{ice}) \bar{\rho} dz, \quad (13)$$

198 where q_{graup} , q_{snow} , and q_{ice} respectively represent the mass mixing ratios (kg kg^{-1}) of graupel, snow, and cloud ice. In
 199 this study, q_{graup} and q_{snow} were computed respectively by equations (9) and (10). For the ECMWF-McCAUL scheme,
 200 V_{graup} and V_{snow} are set respectively to 3.1 and 0.5 m s^{-1} . Then q_{ice} was diagnosed using Arakawa-Schubert cumulus
 201 parameterization.

202

Table 1: Basic information of all lightning schemes assessed for this study

Abbreviation	Parameter	Remark
CTH (Price, C., & Rind, D., 1994)	Cloud top height	Originally used in CHASER (MIROC)
ICEFLUX (Finney et al., 2014)	Upward cloud ice flux at 440 hPa isobar (ICEFLUX P) or at 260-K isotherm (ICEFLUX T)	The 440 hPa level is used as a pressure level representative of fluxes in deep convective clouds
ECMWF-original (Lopez, 2016)	<ul style="list-style-type: none"> ● Q_R (Described in equation 8) ● CAPE ● Convective cloud-base height 	Also adopted in the ECMWF forecasting system
ECMWF-McCAUL	<ul style="list-style-type: none"> ● Column precipitating ice ● CAPE 	Equations and adjustment factors are modified from the original ECMWF scheme. Equations are modified based on findings reported by McCaul (McCaul et al., 2009)

204

205 Table 1 presents all the lightning schemes examined for this study. As described in this paper, the original ECMWF scheme
 206 and the ECMWF-McCAUL scheme are designated collectively as ECMWF schemes. Based on the recent studies, the intra-
 207 cloud (IC) lightning flashes are as efficient as the cloud-to-ground (CG) lightning flashes in NO_x generation and the
 208 lightning NO_x production efficiency (LNO_x PE) is reported to be 100–400 mol per flash (Ridley et al., 2005; Cooray et al.,
 209 2009; Ott et al., 2010; Allen et al., 2019). Therefore, the LNO_x PE values of IC and CG used in CHASER are set to the same
 210 value (250 mol per flash), which is the median of the commonly cited range of 100–400 mol per flash.

211

212 A fourth-order polynomial is used to calculate the proportion of total flashes that are cloud-to-ground (p) based on the cold
 213 cloud depth, as described in an earlier report (Price and Rind, 1993).

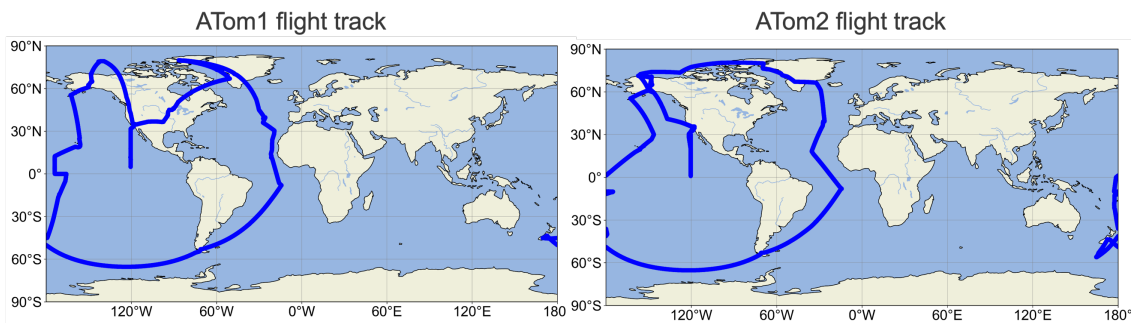
$$214 \quad p = \frac{1}{64.09 - 36.54D + 7.493D^2 - 0.648D^3 + 0.021D^4} \quad (14)$$

215 In that equation, D represents the depth of cloud above the 0°C isotherms in kilometres.

216 2.3 Observation data for model evaluation

217 2.3.1 Lightning observations

218 The LIS/OTD gridded climatology datasets are used for this study, consisting of climatologies of total lightning flash rates
 219 observed using the Lightning Imaging Sensor (LIS) and spaceborne Optical Transient Detector (OTD): OTD aboard the
 220 MicroLab-1 satellite and LIS aboard the Tropical Rainfall Measuring Mission (TRMM) satellite (Cecil et al., 2014). Both
 221 sensors detect lightning by monitoring pulses of illumination produced by lightning in the 777.4 nm atomic oxygen multiplet
 222 above background levels. Both sensors, in low Earth orbit, view an Earth location for about 3 min as OTD passes overhead
 223 or for 1.5 min as LIS passes overhead. Actually, OTD and LIS circle the globe 14 times a day and 16 times a day,
 224 respectively. OTD collected data between +75 and -75° latitude from May 1995 through March 2000, whereas LIS observed
 225 between +38 and -38° latitude from January 1998 through April 2015. The product used throughout this paper is the
 226 LIS/OTD 2.5 Degree Low Resolution Time Series (LRTS). The LRTS includes the daily lightning flash rate on a 2.5°
 227 regular latitude–longitude grid from May 1995 through April 2015.



228 **Figure 1: ATom1 and ATom2 flight tracks.**

230 2.3.2 Atmospheric tomography (ATom) aircraft observations

231 To evaluate the LNO_x emissions calculated by different lightning schemes, we used NO observation by the atmospheric
 232 tomography (ATom) aircraft missions (Wofsy et al., 2018). By deploying an extensive gas and aerosol payload on the

233 NASA DC-8 aircraft, ATom is designed to sample the atmosphere systematically on a global scale, performing continuous
234 profiling from 0.2 to 12 km altitude. Flights took place in each of the four seasons of 2016 through 2018. Since most of the
235 lightning occurs over land regions during summer, ATom1 (July–August 2016) and ATom2 (January–February 2017) were
236 used to evaluate LNO_x emissions (corresponding to summer in the northern and southern hemispheres, respectively). Both
237 ATom1 and ATom2 originate from the Armstrong Flight Research Center in Palmdale, California, USA, fly north to the
238 western Arctic, south to the South Pacific, east to the Atlantic, north to Greenland, and return to California across central
239 North America. Figure 1 exhibits the respective flight tracks of ATom1 and ATom2. To evaluate the model simulated NO
240 against the ATom observations, we have sampled the specific flight track and timings from the modelled data.

241 **2.3.3 TROPOMI satellite observations**

242 Tropospheric Monitoring Instrument (TROPOMI) is the payload on-board the Sentinel-5 Precursor (S5P) satellite of the
243 European Space Agency (ESA), which was launched in October 2017. TROPOMI has been providing observations of
244 important atmospheric pollutants (NO₂, O₃, CO, CH₄, SO₂, CH₂O) with an unprecedented horizontal resolution of approx. 7
245 × 3.5 km² since August 2017 (changed to 5.5 × 3.5 km² after August 2019). The data used in this study is the TROPOMI
246 level-2 offline (OFFL) tropospheric NO₂ columns in 2019. The product version is 1.0.0 from 2019-01-01 to 2019-03-20 and
247 updated to 1.1.0 from 2019-03-21 to 2019-12-31. For the direct comparisons between TROPOMI level-2 products with
248 CHASER results, the following procedures were conducted to pre-process the TROPOMI data and CHASER modelled
249 fields.

- 250 1. The TROPOMI retrievals with quality assurance (QA) values of ≥ 0.75 were selected.
- 251 2. Horizontally, the TROPOMI data (tropospheric NO₂ columns, temperatures, pressures, averaging kernels) were
252 interpolated to the CHASER 2.8° × 2.8° grid.
- 253 3. The modelled results were sampled based on the TROPOMI overpass time. The CHASER tropospheric NO₂ columns
254 were calculated by using the sampled modelled results, the averaging kernels retrieved from the TROPOMI retrievals, and
255 the temperature and pressure profiles provided by TROPOMI retrievals. The averaging kernels are applied to each layer of
256 the CHASER outputs following the equation (16).
- 257 4. The pre-processed data described above were used to produce the monthly averaged data.

258 **2.3.4 OMI satellite observations**

259 Ozone Monitoring Instrument (OMI) is a key instrument onboard NASA's Aura satellite for measuring criteria pollutants
260 such as O₃, NO₂, SO₂, and aerosols. OMI has been providing observations with spatial resolution varying from 13 km × 25
261 km to 26 km × 128 km since October 2004 (Goldberg et al., 2019). The NO₂ product used in this study is the level-3 daily
262 global gridded (0.25° × 0.25°) Nitrogen Dioxide product (OMNO2d) (Nickolay et al. 2019). The O₃ product used in this
263 study is the monthly mean tropospheric column O₃ product developed from OMI in combination with Aura Microwave
264 Limb Sounder (MLS) with the detailed method described by Ziemke et al. (2006).

265 **2.4 Experiment setup**

266 For this study, all the introduced lightning schemes were implemented into CHASER (MIROC). Six sets of experiments
267 were conducted for this study and the detailed settings of all experiments are presented in Table 2. For each set of
268 experiments, the same initial conditions and chemical emissions were used except for LNO_x emissions. The set of
269 experiments that applied meteorological nudging also has the same meteorological conditions. The monthly varying soil NO_x
270 emissions used are constant each year for all experiments derived from Yienger and Levy (1995). All experiments used the
271 “backward C-shaped” LNO_x vertical profile (Ott et al., 2010). The LNO_x PE values of IC and CG used in all experiments are
272 set to the same value (250 mol per flash), which is based on the recent literature (Ridley et al., 2005; Cooray et al., 2009; Ott

273 et al., 2010; Allen et al., 2019). It is noteworthy that there still exist large uncertainties in determining the LNO_x PE values
 274 (Allen et al., 2019; Bucselá et al., 2019) and the choice of different LNO_x PE values may influence the simulated LNO_x
 275 emissions and chemical fields. A more sophisticated parametrisation of LNO_x PE values needs to be implemented and
 276 verified in the chemistry-climate models in future research.

277

278 The first set of experiments was conducted for the years of 2001–2020. It was used to evaluate the distribution of the
 279 lightning flash rate against LIS/OTD lightning observations and to derive the historical lightning trend. The second set of
 280 experiments is the same as the first set of experiments, but uses daily mean LNO_x emission rates of 2001 calculated using
 281 lightning schemes for each year. This set of experiments is used to produce results for comparison with those of the first set
 282 of experiments to estimate the effects of LNO_x emission trends on tropospheric NO_x and O₃ column trends. The third set of
 283 experiments gives results for 2011–2020. These experiments are used to estimate the effects of different lightning schemes
 284 on atmospheric chemical fields. To normalize the different annual LNO_x emission amounts by different lightning schemes,
 285 temporally and spatially uniform adjustment factors were applied to adjust the mean LNO_x production (2011–2020) to 5.0
 286 TgN yr⁻¹. Note the 10-years (2011–2020) mean LNO_x production was adjusted to 5.0 TgN yr⁻¹ but the LNO_x production in
 287 each year is not exactly 5.0 TgN yr⁻¹. This adjustment was achieved by first conducting the simulations without any
 288 adjustment and the 2011–2020 mean LNO_x production (P_{LNO_x}) was calculated, then the corresponding adjustment factor
 289 (*adj_factor*) can be calculated by using the following equation.

$$290 \text{ adj_factor} = \frac{5.0}{P_{LNO_x}} \quad (15)$$

291 Similarly, we also adjusted the LNO_x emissions in the fourth to the sixth sets of experiments to 5.0 TgN yr⁻¹. The fourth set
 292 of experiments is for 2016, with the fifth set for 2017. These two sets of experiments were conducted to compare model
 293 results with ATom1 and ATom2 aircraft observations. The sixth set of experiments is for 2019. It is conducted to evaluate
 294 model results using TROPOMI satellite observations.

295

Table 2: All experiments in this study

Number	1st		2nd		3rd	4th	5th	6th
Period	2001–2020	2001–2020	2001–2020	2001–2020	2011–2020	2016	2017	2019
Nudging	On	Off ^a	On	Off	On	On	On	On
LNO _x emissions	Interactively calculated ^b	Interactively calculated	Fixed to 2001	Fixed to 2001	Interactively calculated	Interactively calculated	Interactively calculated	Interactively calculated
Adjusted to 5.0 TgN yr ⁻¹	No	No	No	No	Yes	Yes	Yes	Yes
Climate ^c	2001-2020 (RCP4.5)	2001-2020 (RCP4.5)	2001-2020 (RCP4.5)	2001-2020 (RCP4.5)	2011-2020 (RCP4.5)	2016 (RCP4.5)	2017 (RCP4.5)	2019 (RCP4.5)
Anthropogenic emissions	HTAP-II (2008) for all years							
Soil NO _x emissions	Monthly varying values but constant for each year derived from Yienger and Levy (1995)							
Biomass burning emissions	MACC (2001-2020)	MACC (2001-2020)	MACC (2001-2020)	MACC (2001-2020)	MACC (2011-2020)	MACC (2016)	MACC (2017)	MACC (2019)

296 ^aNudging off means the meteorological fields (u, v, T) are free-running instead of nudging towards the NCEP FNL data.

297 ^bLNO_x is interactively calculated by using different lightning schemes.

298 ^cThe climate change is simulated by prescribed SST/sea ice fields and prescribed varying concentrations of GHGs (CO₂,
 299 N₂O, methane, chlorofluorocarbons – CFCs – and hydrochlorofluorocarbons – HCFCs) utilized only in the radiation scheme.

300 The SST/sea ice fields are obtained from the HadISST dataset (Rayner et al., 2003).

301 **3 Results and Discussion**

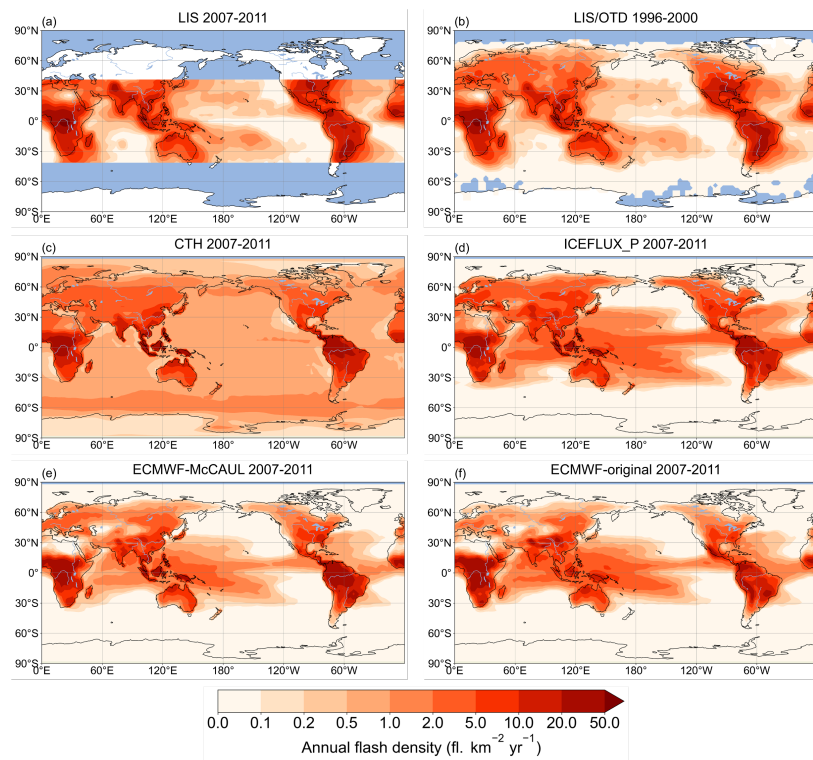
302 **3.1 Evaluation of the lightning schemes**

303 As investigated by Finney et al. (2014), 5 years data are necessary and appropriate to produce a lightning climatology.
304 Therefore, model results with nudging (2007-2011) were evaluated against the climatological lightning distributions of LIS
305 (2007-2011) within $\pm 38^\circ$ latitude and LIS/OTD (1996-2000) within a broader range of $\pm 75^\circ$ latitude. We have evaluated the
306 potential uncertainties associated with the inconsistency of the time period of simulated lightning and observed lightning
307 (2007-2011 and 1996-2000). The statistical analysis between LIS (2007-2011) and LIS/OTD (1996-2000) within $\pm 38^\circ$
308 latitude exhibits an extremely high spatial correlation coefficient ($R=0.99$) and relatively small relative bias (0.65%), which
309 supports the reasonability of comparing model results with the observation data within different time range.

310
311 The distribution of lightning observed by LIS/OTD and simulated by CHASER (MIROC) with different lightning schemes is
312 depicted in Fig. 2. Figure 2 shows that lightning over the ocean is not well reproduced by the original CTH scheme.

313 Actually, it is improved considerably by the new lightning schemes. Compared with the CTH scheme, the original ECMWF
314 scheme better represents the lightning distribution in South Asia including the Indian region. The ECMWF schemes and the
315 ICEFLUX_P scheme reduced negative biases in North America compared to the CTH scheme. In Australia, the ECMWF
316 schemes better simulate the horizontal distribution of lightning. All lightning schemes failed to capture the worldwide
317 maximum value found over the Congo Basin, although all lightning schemes captured the active region in central Africa.

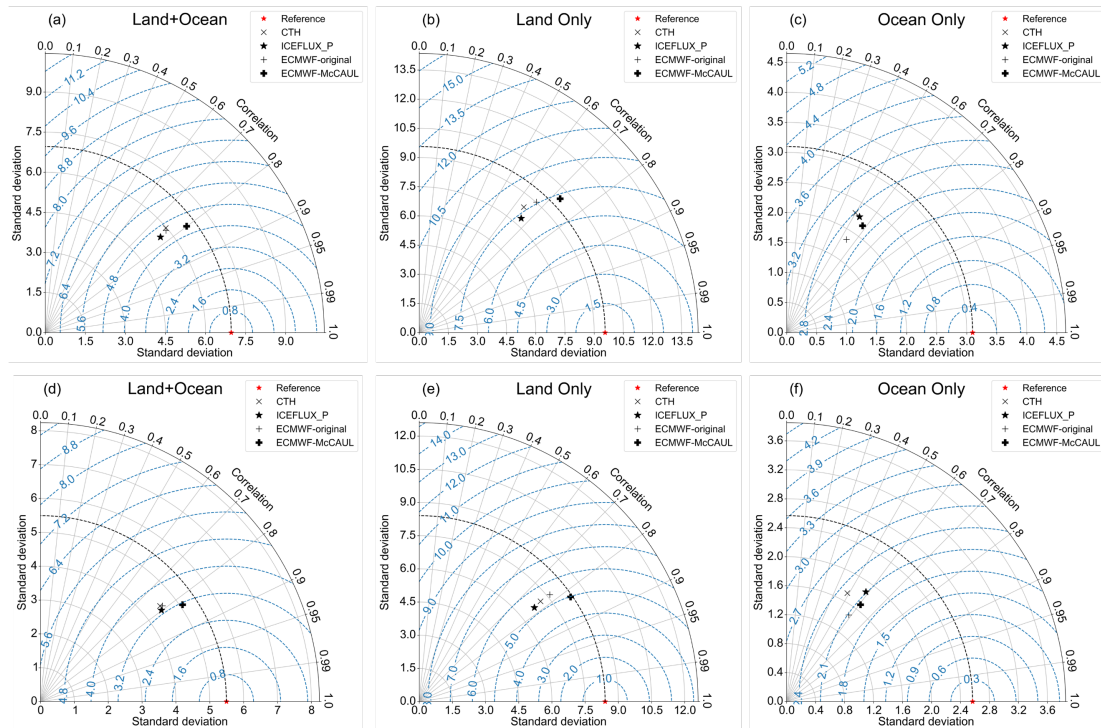
318



319
320 **Figure 2: Annual mean lightning flash densities from (a) LIS satellite observations spanning 2007–2011, (b) LIS/OTD satellite**
321 **observations spanning 1996–2000 but with a wider range, (c) the CTH scheme in 2007–2011, (d) the ICEFLUX_P scheme in 2007–**
322 **2011, (e) the ECMWF-McCAUL scheme in 2007–2011, and (f) the original ECMWF scheme in 2007–2011.**

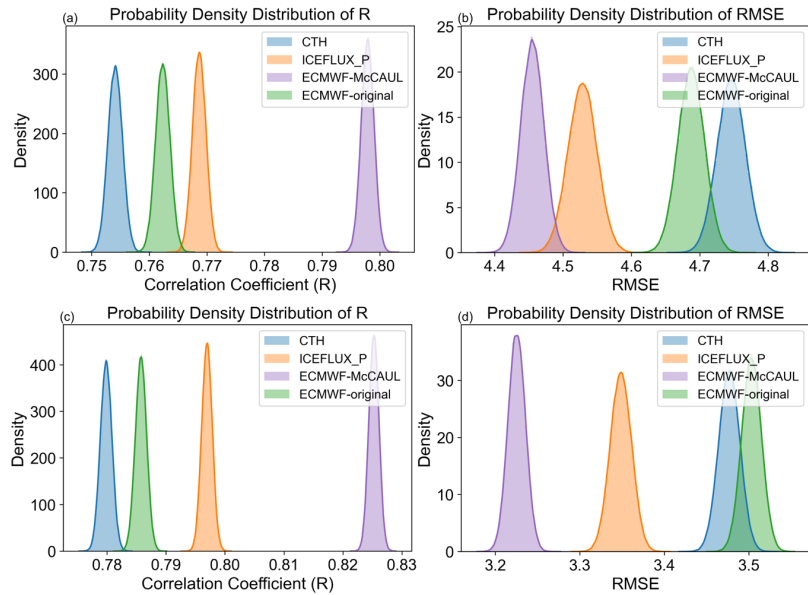
323
324 To directly estimate the prediction accuracy of all lightning schemes, the Taylor diagrams are displayed in Fig. 3. In Fig. 3a,
325 the overall prediction accuracy of the ICEFLUX_P and original ECMWF schemes evaluated against the LIS 2007-2011
326 lightning climatology is slightly improved compared to the CTH scheme. This improvement is more obvious when
327 considering land and ocean separately (Figs. 3b-c). In the case of Figs. 3a-c, the ECMWF-McCAUL scheme has shown the
328 best prediction accuracy among all lightning schemes. In Fig. 3d, comparison of the annual mean lightning flash rate of
329 LIS/OTD 1996–2000 and the CHASER calculation for 2007–2011 yields spatial correlation coefficients of 0.80 and 0.79 for

330 the ICEFLUX_P and original ECMWF schemes, respectively, which are slightly higher than that found for the CTH scheme
 331 (0.78). The overall RMSE of the ICEFLUX_P scheme is 3.32 fl. km⁻² yr⁻¹, which is slightly less than that of the CTH scheme
 332 of 3.44 fl. km⁻² yr⁻¹. Among all lightning schemes, the ECMWF-McCAUL scheme exhibits the highest spatial correlation
 333 coefficient (0.83) and the lowest RMSE (3.20 fl. km⁻² yr⁻¹) as depicted in Fig. 3d. As displayed in Fig. 2, the prediction
 334 accuracy of lightning over the ocean is significantly improved, which can also be verified in Fig. 3f.



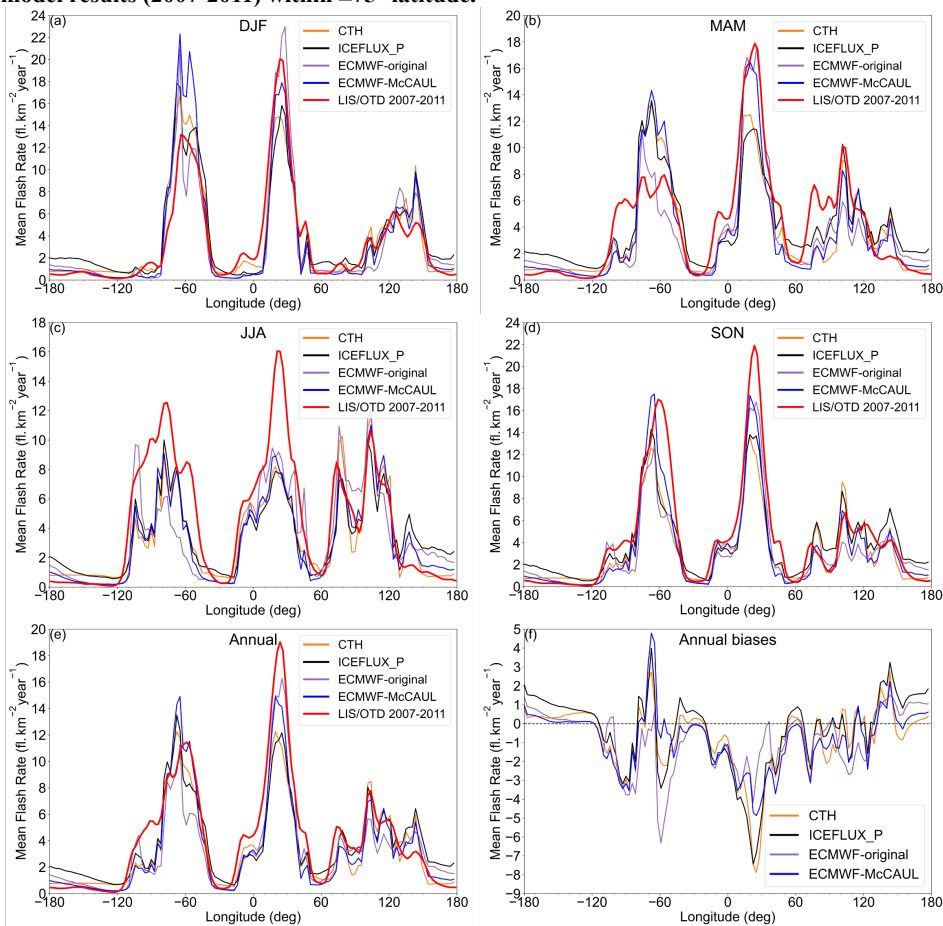
335
 336 **Figure 3: Taylor diagram showing the prediction accuracy of various lightning schemes in 2007–2011 simulations compared to the**
 337 **LIS 2007-2011 lightning climatology (a-c) and the LIS/OTD 1996–2000 lightning climatology (d-f).**
 338

339 To estimate whether the improvement of prediction accuracy discussed in Fig. 3 is significant, a significant test is conducted
 340 by considering the uncertainties in the LIS/OTD observations. Based on the uncertainties in the LIS/OTD observations, the
 341 probability density distributions (PDDs) of spatial correlation coefficients (R) and RMSE between the model and
 342 observations are derived by using a Monte Carlo method and displayed in Fig. 4. The uncertainties in the LIS/OTD
 343 observations are determined based on the uncertainties of the instrument bulk flash detection efficiency of LIS ($88 \pm 9\%$)
 344 and OTD ($54 \pm 8\%$) (Boccippio et al., 2002). The R and RMSE shown in Fig. 4 are all normally distributed which is
 345 determined by the Kolmogorov–Smirnov test. Based on the probability density functions of R and RMSE derived from Fig.
 346 4, the order of R between the model and observations is estimated to be ECMWF-McCAUL > ICEFLUX_P > ECMWF-
 347 original > CTH with a confidence limit larger than 99.9%. Moreover, the order of RMSE between the model and
 348 observations is estimated to be ECMWF-McCAUL < ICEFLUX_P < ECMWF-original and CTH with a confidence limit
 349 larger than 95%. According to the significant test described above, we can conclude that the newly implemented lightning
 350 schemes have improved the lightning prediction accuracy compared to the original CTH scheme.



351

352 **Figure 4: The probability density distributions (PDDs) of spatial correlation coefficients (R) and RMSE between the model and**
 353 **LIS/OTD lightning observations. Figures 4(a–b) show the PDDs obtained between LIS lightning climatology (2007–2011) and the**
 354 **model outputs (2007–2011) within $\pm 38^\circ$ latitude. Figures 4(c–d) show the PDDs obtained between LIS/OTD lightning climatology**
 355 **(1996–2000) and the model results (2007–2011) within $\pm 75^\circ$ latitude.**



356

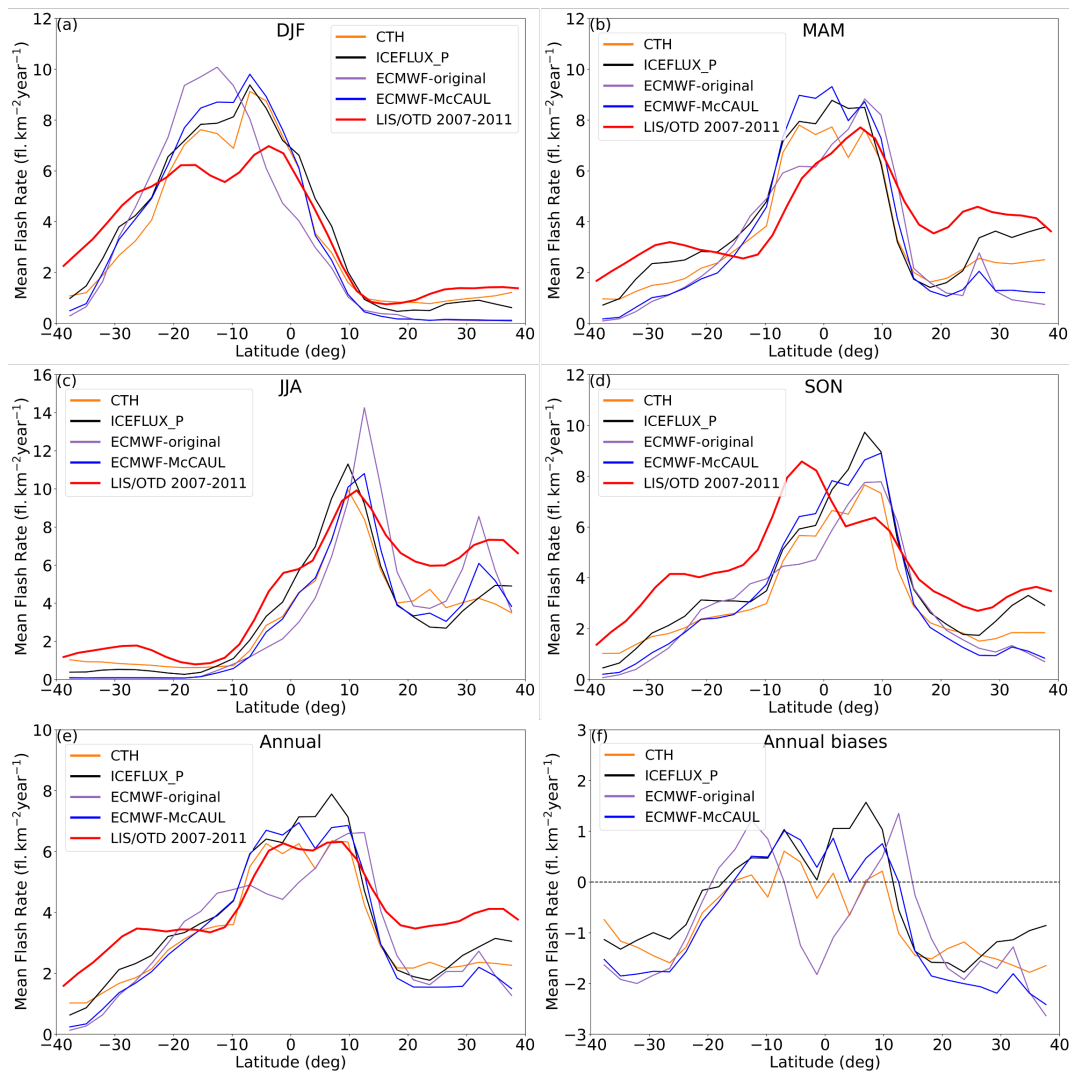
357 **Figure 5: Seasonal and annual meridional average lightning flash densities distribution from LIS 2007–2011 climatology (red line)**
 358 **and from simulation results (2007–2011) obtained using different lightning schemes. The meridional average is only taken within the**
 359 **LIS viewing region of $\pm 38^\circ$ latitude. The biases (model-obs.) in Fig. 5e are also portrayed in Fig. 5f.**
 360

361 Figure 5 displays a comparison of seasonal and annual meridional average lightning flash densities from simulations (2007–
 362 2011) and LIS satellite observations (2007–2011). As Fig. 5 shows, the pairs of curves are usually in good agreement, even
 363 though the annual plot (Fig. 5e) highlights the underestimation which occurs for Africa (from 0 degrees to 30 degrees east)
 364 and North America (from 80 degrees west to 120 degrees west). The ECMWF schemes have made improvements within
 365 Africa. Also, the ICEFLUX_P and the original ECMWF schemes have slightly reduced the biases over North America. A

366 noticeable underestimation over the Americas in JJA and overestimation in MAM can be observed respectively in Figs. 5c
 367 and 5b. Lightning densities over Africa are generally underestimated to varying degrees in different seasons, with the
 368 greatest underestimation occurring in JJA (Fig. 5c). Lightning densities over Asia (from 60 degrees east to 120 degrees east)
 369 are slightly underestimated in MAM (Fig. 5b). The ICEFLUX_P scheme has reduced the biases.

370

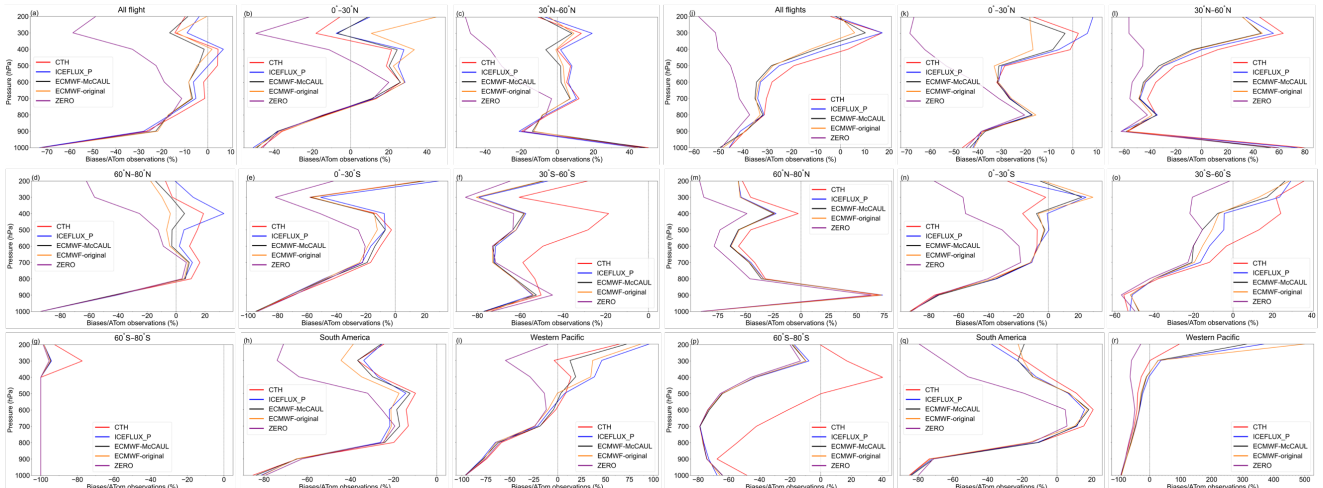
371 Figure 6 is the same as Fig. 5, but for the zonal mean distributions. The curves of the model results and the observation
 372 results in Fig. 6 generally show good agreement. Figure 6f shows that, overall, the ICEFLUX_P and the ECMWF-McCAUL
 373 schemes slightly overestimated the lightning densities near the equator (10°S – 10°N). All lightning schemes underestimated
 374 the lightning densities within 15°N – 38°N and 20°S – 38°S . Figure 6f also shows that the ICEFLUX_P scheme has reduced
 375 the biases within 10°N – 38°N and 15°S – 38°S compared to the CTH scheme. In DJF (Fig. 6a), all lightning schemes
 376 overestimated the flash densities over the low latitude regions but slightly underestimated the flash densities over the middle
 377 latitude regions in the Southern Hemisphere. In MAM (Fig. 6b), lightning densities are overestimated near the equator and
 378 underestimated over 15°N – 38°N and 15°S – 38°S by all lightning schemes to varying degrees. In JJA (Fig. 6c), noticeable
 379 overestimation around 10°N by the original ECMWF scheme is apparent. Moreover, the CTH and the original ECMWF
 380 schemes respectively facilitated reduction of model biases over 15°S – 38°S and 15°N – 38°N . As Fig. 6d shows, the model-
 381 predicted lightning maximum value is shifted approximately 15 degrees to the north in SON compared to the lightning
 382 observations. Figure 6d also shows that all lightning schemes underestimated the lightning densities over 15°N – 38°N and
 383 0° – 38°S . The ICEFLUX_P scheme has shown improvement over these regions.



384
 385 **Figure 6: Seasonal and annual zonal average lightning flash densities distribution from LIS 2007–2011 climatology (red line) and**
 386 **from the simulation results (2007–2011) obtained using different lightning schemes. The biases (model-obs.) in Fig. 6e are also**
 387 **presented in Fig. 6f.**

389 3.2.1 Evaluation of LNO_x emissions by ATom1 and ATom2 observations

390 To evaluate the LNO_x emissions of different lightning schemes, we used ATom1 and ATom2 aircraft measurements (NO)
 391 for comparison against model results. All lightning schemes, when implemented in CHASER, produce flash rates
 392 corresponding to global annual LNO_x emissions within the range estimated by Schumann and Huntrieser (2007) of 2–8 TgN
 393 yr⁻¹. To eliminate differences in annual total LNO_x emissions by different lightning schemes, we chose to adjust the annual
 394 LNO_x emissions of all lightning schemes to 5.0 TgN yr⁻¹ by applying adjustment factors. The “backward C-shaped” LNO_x
 395 vertical profile is applied to all lightning schemes.



396 **Figure 7: Vertical profile of biases/ATom1 observations (a–i) and the vertical profile of biases/ATom2 observations (j–r). The bias is**
 397 **the model bias (NO) against ATom observations (NO). Data for each pressure level P are calculated within the range of P±50 hPa.**
 398 **South America is the region of 0°–30°S, 0°–30°W. The Western Pacific is the region of 10°N–30°S, 160°E–160°W.**

401 **Table 3: Model biases (NO) when compared against ATom1 (upper panel) and ATom2 (lower panel). The unit is ppt. The biases**
 402 **within the South America region (0–30°S, 0–30°W) and Western Pacific region (10°N–30°S, 160°E–160°W) are also shown in this**
 403 **table.**

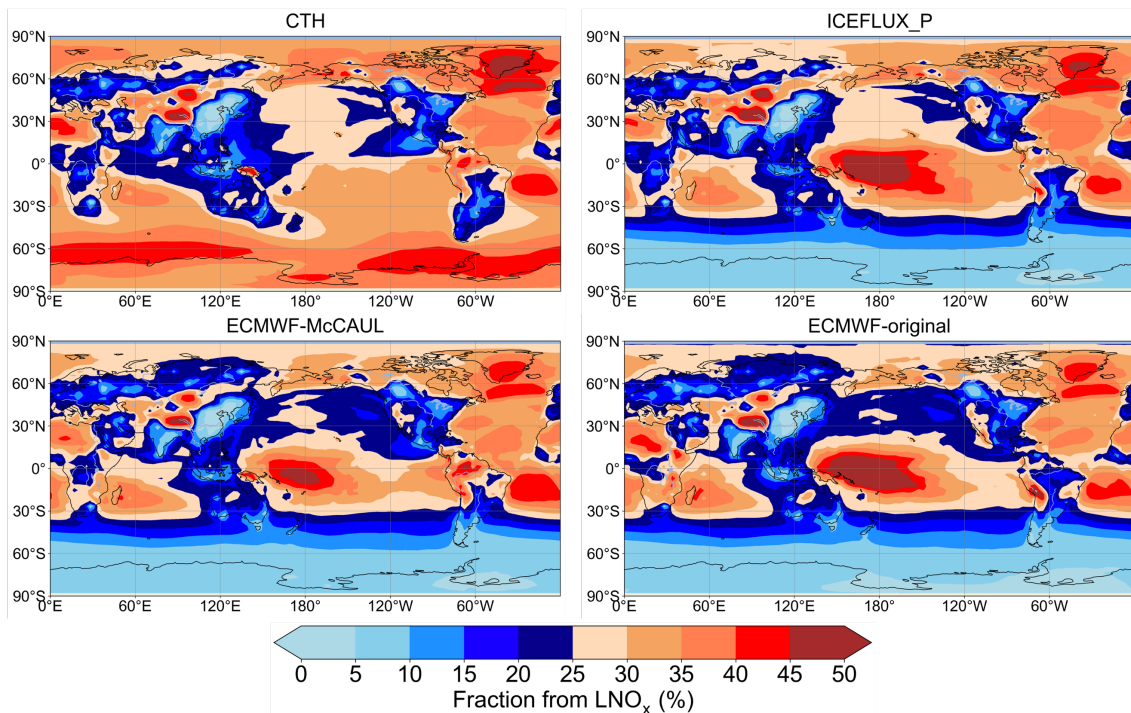
Lightning scheme	All flight	0°–30°N	30°N–60°N	60°N–80°N	0°–30°S	30°S–60°S	60°S–80°S	South America	Western Pacific
CTH	-6.54	-3.22	-0.50	-13.06	-9.33	-12.32	-7.55	-6.79	-3.03
ICEFLUX_P	-5.18	0.31	1.15	-9.16	-8.21	-16.21	-8.28	-7.00	0.08
ECMWF-McCAUL	-6.99	0.13	-1.05	-14.80	-9.43	-16.42	-8.29	-7.17	-2.24
ECMWF-original	-5.48	7.03	0.28	-16.66	-9.59	-16.38	-8.30	-8.71	-0.72
ZERO	-19.00	-11.02	-20.85	-32.98	-15.91	-17.35	-8.34	-13.77	-8.63
Lightning scheme	All flight	0°–30°N	30°N–60°N	60°N–80°N	0°–30°S	30°S–60°S	60°S–80°S	South America	Western Pacific
CTH	-0.91	-2.57	5.80	-6.18	-11.11	3.61	1.45	-19.16	-4.70
ICEFLUX_P	-1.04	-0.76	3.98	-6.81	-7.45	2.82	-4.88	-22.02	3.01
ECMWF-McCAUL	-1.73	-3.71	2.81	-6.89	-3.71	1.81	-5.33	-12.24	1.20
ECMWF-original	-1.95	-5.26	2.96	-6.87	-2.74	1.58	-5.23	-13.90	3.55
ZERO	-12.66	-15.51	-11.08	-9.77	-28.40	-4.18	-5.94	-47.68	-13.14

404
 405 Table 3 presents model biases of different lightning schemes against the ATom1 and ATom2 observations. Figure 7 displays
 406 the vertical profile of biases/ATom observations in percentage terms. In Table 3 and Fig. 7, case ZERO is the case with the
 407 lightning flash, with LNO_x emissions completely switched off. Comparisons between model results and ATom observations
 408 were conducted within two specific regions (South America region and Western Pacific region) in which LNO_x is the major
 409 source of NO_x (Fig. 8). As Table 3 and Fig. 7 show, the model generally tends to underestimate the NO concentrations. The
 410 model biases are reduced considerably by including lightning NO_x sources. For ATom1, overall, the ICEFLUX_P scheme
 411 has the smallest model bias. The original ECMWF scheme also reduced the model biases compared to the CTH scheme

412 (Table 3). The ICEFLUX_P and the ECMWF-McCAUL schemes reduced the model biases substantially within 0°–30°N
 413 latitude where the lightning activities are most dominant during the ATom1 observation period (2016-07-29 ~ 2016-08-23).
 414 In the range of 30°S to 80°N in ATom1, overall the ICEFLUX_P scheme reduced the model biases considerably and the
 415 ECMWF schemes slightly reduced or extended the model biases compared to the CTH scheme (Table 3, Figs. 7b–e).
 416 However, in the range of 30°S–80°S, the model biases were extended by the ICEFLUX_P and the ECMWF schemes
 417 compared to the CTH scheme (Table 3, Figs. 7f–g).

418

419 For ATom2, overall, the ECMWF schemes slightly reduced the model biases over the upper troposphere, compared to the
 420 CTH scheme (Fig. 7j). During the ATom2 observation period (2017-01-26 ~ 2017-02-21), the lightning activities are most
 421 dominant within the range of 0°–30°S, where the model biases were reduced significantly by newly implemented lightning
 422 schemes. A hotspot of lightning activities during the ATom2 observation period is the South America region, where the
 423 model biases were reduced dramatically by the ECMWF schemes. The model biases were mostly reduced by the newly
 424 implemented lightning schemes within the low latitude and middle latitude regions, but slightly extended within the high
 425 latitude regions. The model biases were mostly reduced or extended over the middle to upper troposphere (Fig. 7). This is
 426 true because most LNO_x was distributed over the middle to upper troposphere. Also, NO_x has a longer lifetime over the
 427 middle to upper troposphere. In the Western Pacific region, results obtained from comparisons with ATom1 and ATom2
 428 indicate that all lightning schemes overestimated LNO_x emissions in the upper troposphere; also, both the ICEFLUX_P
 429 scheme and ECMWF schemes reduced the total model biases considerably more than the CTH scheme did.



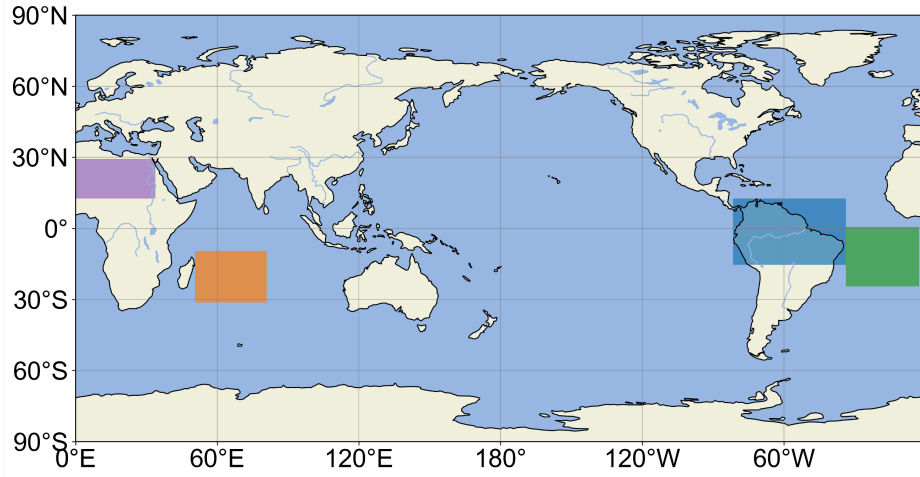
430
 431 **Figure 8: Sensitivity of simulated tropospheric NO₂ columns to LNO_x emissions using different lightning schemes in 2019. NO₂**
 432 **column because of LNO_x emissions was determined as the difference between the simulation with LNO_x emissions and a simulation**
 433 **that excludes LNO_x emissions.**

434 3.2.2 Evaluation of LNO_x emissions by TROPOMI satellite observations

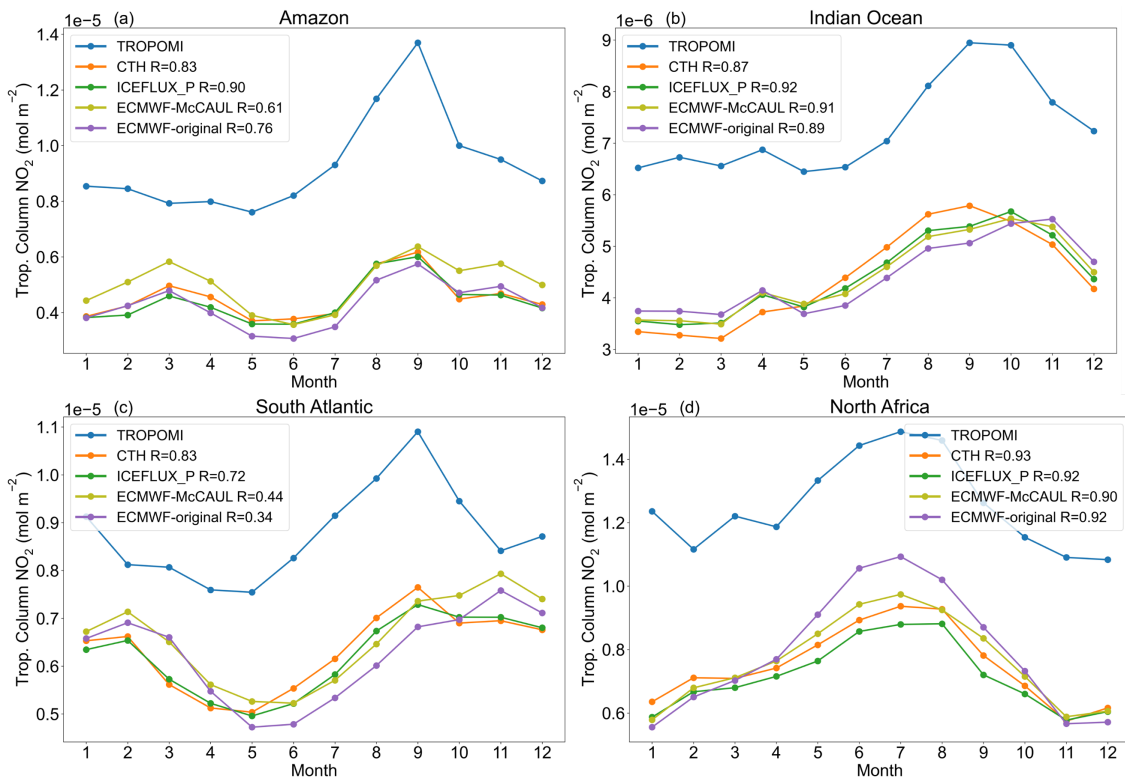
435 TROPOMI satellite observations of tropospheric NO₂ columns were used to evaluate LNO_x emission results obtained using
 436 the CHASER model. To eliminate differences in annual total LNO_x emissions attributable to the different lightning schemes,
 437 we adjusted the annual LNO_x emissions of all lightning schemes to 5.0 TgN yr⁻¹ using different adjustment factors. For direct
 438 comparison between CHASER and TROPOMI tropospheric NO₂ columns, the averaging kernel information from
 439 TROPOMI observations was used. The averaging kernels were applied to CHASER outputs following Eq. (16).

$$440 X_{chaser} = \sum_{i=1}^N A_{tropomi} x_{chaser} \quad (16)$$

441 In that equation, X_{chaser} represents the CHASER tropospheric NO_2 column after averaging kernels applied, $A_{tropomi}$
 442 denotes the TROPOMI averaging kernels, x_{chaser} denotes the CHASER NO_2 partial column at layer i , and N denotes the
 443 number of tropospheric layers.



444
 445 **Figure 9: Four target regions for which LNO_x is the major source of NO_x. The four target regions are North Africa (purple),**
 446 **Indian Ocean (orange), Amazon (blue), and South Atlantic (green).**
 447



448
 449 **Figure 10: Comparisons of smoothed CHASER and TROPOMI (blue) tropospheric NO_2 columns over four target regions in 2019.**
 450 **Legends show the temporal correlation coefficients.**
 451

452 Comparison between TROPOMI observations and CHASER outputs indicates that the CHASER model tends to
 453 underestimate tropospheric NO_2 columns. Overall, the newly implemented lightning schemes have not shown improvements
 454 of model biases of tropospheric NO_2 columns at an annual global scale. To minimize the uncertainties of model biases of
 455 tropospheric NO_2 columns caused by other factors, we chose to further evaluate the LNO_x emissions by TROPOMI
 456 observations over four specific regions (Fig. 9), where LNO_x is the major source of NO_x (as shown in Fig. 8).
 457 Figure 10 presents a comparison of smoothed CHASER and TROPOMI tropospheric NO_2 columns over four target regions
 458 in 2019. The spatial average values of each month in 2019 are shown in Fig. 10. That figure shows, generally, the model
 459 captured the temporal variation of tropospheric NO_2 columns in the four regions. Actually, the temporal variations of
 460 modelled tropospheric NO_2 columns are close to each other. For the Amazon region, lightning activities are most dominant

461 during MAM and SON, when the ECMWF-McCAUL scheme has shown noticeable improvements in model biases (Fig.
 462 10a). Figure 10b reveals that all the newly implemented schemes slightly reduced the model biases with the original
 463 ECMWF scheme showing the smallest model biases during the most prevailing season of lightning (DJF). Figure 10c is for
 464 the South Atlantic region where the most prevailing season of lightning is also DJF. Figure 10c shows that the ECMWF
 465 schemes slightly reduced the model biases compared to the CTH scheme. Referring to Fig. 10d, the dominant season of
 466 lightning is JJA, when the ECMWF-original scheme considerably reduced the model biases and the ECMWF-McCAUL
 467 scheme also slightly reduced the model biases.

468 3.3 Effects of different lightning schemes on tropospheric chemical fields

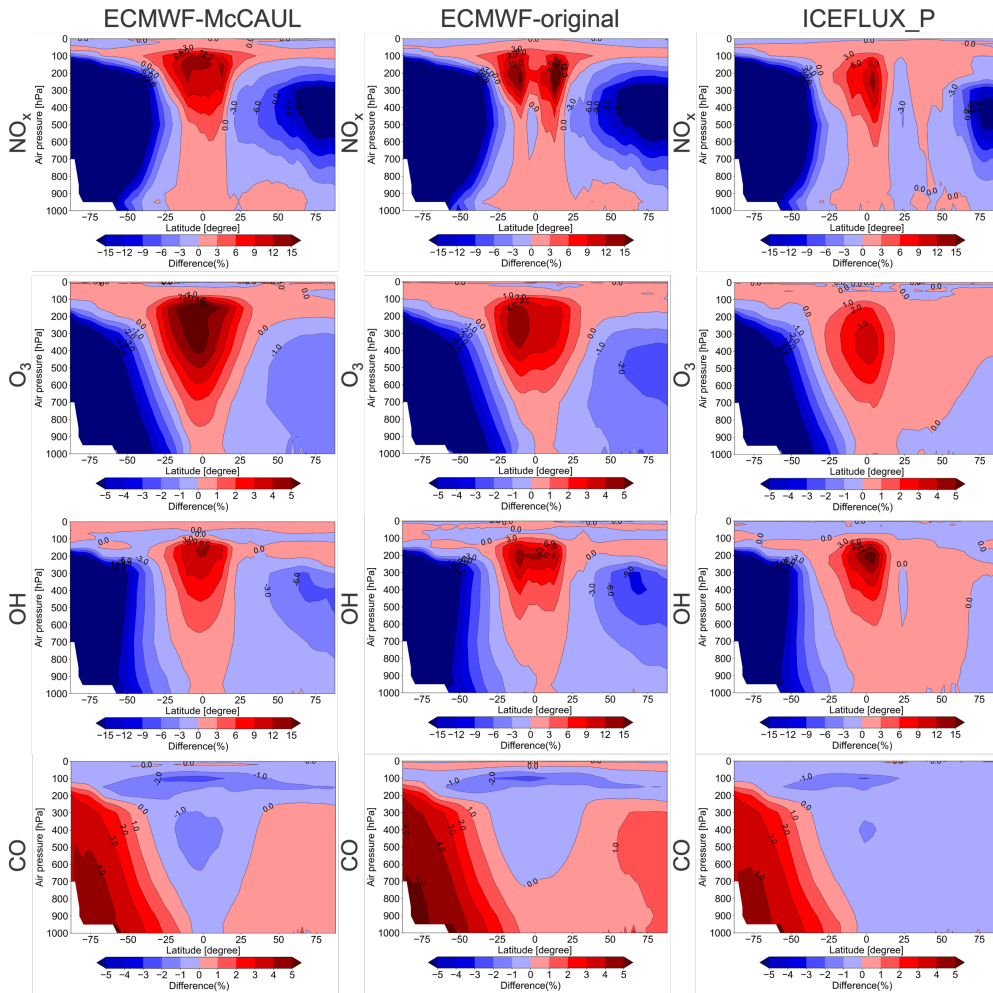
469 In the tropospheric chemical field, LNO_x has an important role. The LNO_x effects on the tropospheric chemical fields vary
 470 along with differences in the horizontal distribution of LNO_x in different lightning schemes. To evaluate the influences of
 471 different lightning schemes on the tropospheric chemical fields, several ten-year (2011–2020) experiments were conducted
 472 with the ten-year mean LNO_x production of all lightning schemes adjusted to 5.0 TgN yr⁻¹ (Sect. 2.4). CTH scheme with a
 473 “backward C-shaped” profile is regarded as the base scheme. The effects of different lightning schemes on the atmospheric
 474 chemistry are calculated as shown in Eq. (17).

$$475 \text{Impact}_{ij} = \frac{(LS_{ij} - \text{Base}_j)}{\text{Base}_j} \quad (17)$$

476 Therein, *Impact*_{ij} represents the effects of the *i*-th lightning scheme on the concentrations of target atmospheric component
 477 *j*. Also, *LS*_{ij} denotes the concentrations of target atmospheric component *j* simulated by the *i*-th lightning scheme. *Base*_{*j*}
 478 stands for the concentrations of target atmospheric component *j* as simulated using the base scheme.

479
 480 Figure 11 presents the respective effects of the ECMWF-McCAUL, original ECMWF, and ICEFLUX_P schemes on the
 481 atmospheric chemical fields (NO_x, O₃, OH, CO) relative to the base scheme CTH. The ECMWF-McCAUL scheme led to an
 482 increase (approximate maximum is 12%) in NO_x concentration at low latitude regions and a decrease (approximate
 483 maximum is 15%) at middle to high latitude regions. In the case of the ECMWF-McCAUL scheme, the concentration of
 484 ozone and OH radical mostly increased at low latitude regions and decreased at middle to high latitude regions in the
 485 Southern Hemisphere, which corresponds to the changing pattern of NO_x. The effects of the original ECMWF scheme on the
 486 atmospheric chemical fields are similar to that of the ECMWF-McCAUL scheme. However, the original ECMWF scheme
 487 led to a higher total tropospheric CO burden compared to the ECMWF-McCAUL scheme. As Fig. 11 shows, the three
 488 lightning schemes led to a marked decrease in NO_x, O₃, and OH radical concentrations over the South Pole region. This
 489 decrease occurred because the lightning densities and the LNO_x emissions simulated by the CTH scheme are markedly
 490 higher than those simulated using other lightning schemes at this latitude band (Fig. 2). Moreover, NO_x can engender the
 491 formation of ozone and OH radical. In the case of the ICEFLUX_P scheme, the concentrations of NO_x, ozone, and OH
 492 radical mostly increased in the tropics and decreased at middle to high latitude regions in the Southern Hemisphere.

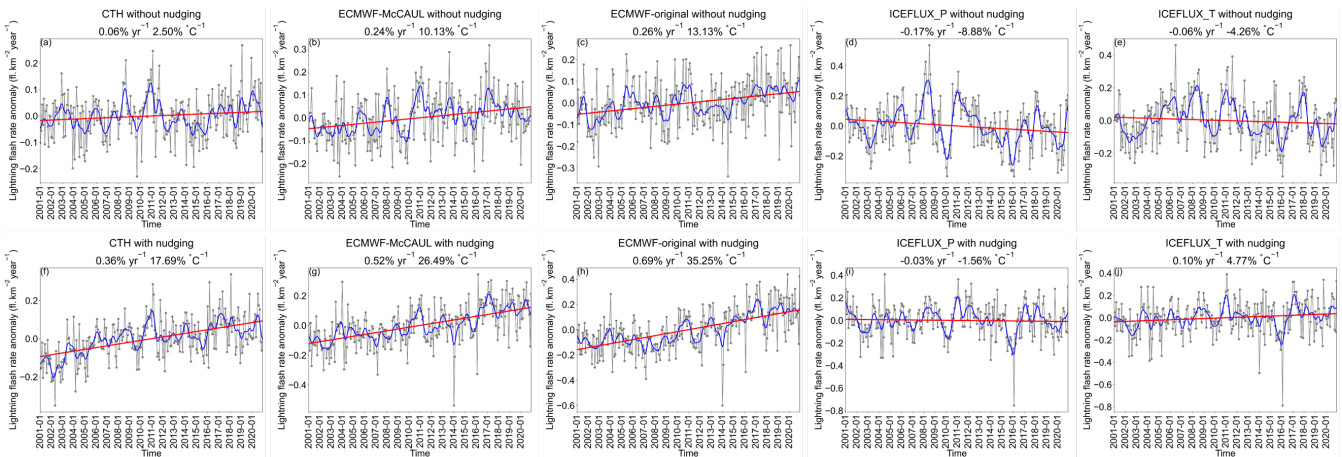
493
 494 Methane lifetime is an indicator reflecting the tropospheric oxidation capacity. The global mean tropospheric lifetime of
 495 methane against tropospheric OH radical spanning 2011–2020 with the CTH, original ECMWF, ECMWF-McCAUL, and
 496 ICEFLUX_P schemes are estimated respectively as 9.226 years, 9.299 years, 9.256 years, and 9.229 years. Compared to the
 497 CTH scheme, the ECMWF schemes led to a slight increase in methane's global mean tropospheric lifetime. In contrast, the
 498 methane's global mean tropospheric lifetime simulated by the ICEFLUX_P scheme is almost the same as that simulated by
 499 the CTH scheme. Although little difference exists in the total tropospheric oxidation capacity simulated by different
 500 lightning schemes, the ECMWF schemes and ICEFLUX_P scheme led to marked changes of oxidation capacity in different
 501 regions of the troposphere.



502
503 **Figure 11: Effects of ECMWF-McCAUL scheme, original ECMWF scheme, and ICEFLUX_P scheme on the atmospheric chemical**
504 **fields (NO_x, O₃, OH, CO) relative to the CTH scheme on the zonal mean (%).**

505 **3.4 Historical trend analysis of lightning density**

506 The accuracy of predicting the simulated lightning distribution under the current climate is only one aspect of lightning
507 scheme evaluation. The ability of the lightning scheme to reproduce the trend of lightning under climate change is also an
508 important factor. For this study, 20 years of (2001–2020) experiments were conducted to analyze the historical trends of
509 lightning flash rates simulated using different lightning schemes (Sect. 2.4).



510
511 **Figure 12: Global anomaly of lightning flash rates calculated from simulation results (2001–2020) using different lightning schemes.**
512 **Figures 12(a–e) present results without nudging; Figs. 12(f–j) present results with nudging. The grey lines with points represent the**
513 **monthly time-series data of the global mean lightning flash rate anomaly. The blue curves represent the monthly time-series data of**
514 **the global mean lightning flash rate anomaly with the 1-D Gaussian (Denosing) Filter applied. The red lines are the fitting curves**
515 **of the grey lines.**

518 **Table 4: Changes in global mean surface temperature (ΔTS), global mean lightning flash rate (ΔLFR), and the rate of change of**
519 **lightning flash rate corresponding to each degree Celsius increase in global mean surface temperature ($\Delta LFR/\Delta TS$). The upper panel**
520 **shows results obtained without nudging. The lower panel presents results obtained with nudging. Changes were obtained by**
521 **calculating the difference between the rightmost and leftmost points of the approximating curve for the 2001–2020 time-series data.**

Lightning scheme	ΔTS ($^{\circ}C$)	ΔLFR (%)	$\Delta LFR/\Delta TS$ ($\%/^{\circ}C$)
CTH	0.38	0.95	2.50
ECMWF-McCAUL	0.39	3.95	10.13
ECMWF-original	0.40	5.25	13.13
ICEFLUX_P	0.40	-3.55	-8.88
ICEFLUX_T	0.34	-1.45	-4.26
Lightning scheme	ΔTS ($^{\circ}C$)	ΔLFR (%)	$\Delta LFR/\Delta TS$ ($\%/^{\circ}C$)
CTH	0.39	6.90	17.69
ECMWF-McCAUL	0.39	10.33	26.49
ECMWF-original	0.39	13.74	35.23
ICEFLUX_P	0.39	-0.61	-1.56
ICEFLUX_T	0.39	1.86	4.77

522

523 Figure 12 shows the global anomaly of lightning flash rates calculated from the simulation results. Because nudging to
524 meteorological reanalysis data cannot be used when predicting lightning trends under future climate changes, we also
525 showed the results without nudging. The un-nudged runs also represented the short-term surface warming like the
526 experiments with nudging. The only differences between the un-nudged and nudged experiments are whether the
527 meteorological fields are nudged towards the six-hourly NCEP FNL data. We used the Mann–Kendall rank statistic to
528 ascertain whether the lightning trends in Fig. 12 are significant (Hussain et al., 2019). From results of the Mann–Kendall
529 rank statistic test (significance set as 5%), all the trends in Fig. 12 were inferred as significant except for the trends shown in
530 Figs. 12a, e, i. As Fig. 12 shows, all lightning schemes predicted increasing trends or no significant trends of lightning except
531 the ICEFLUX_P scheme without nudging, which predicted a decreasing lightning trend. The isotherms alternative
532 application of ICEFLUX (ICEFLUX_T) led to slightly enhanced lightning trends toward positive lighting trends compared
533 to the ICEFLUX_P scheme. As explained by Romps (2019), the ICEFLUX_P approach is based on a fixed isobar which
534 makes it less convenient for climate change studies. Therefore, at least the lightning trends simulated by the ICEFLUX_T
535 approach are expected closer to the real situation than the ICEFLUX_P approach.

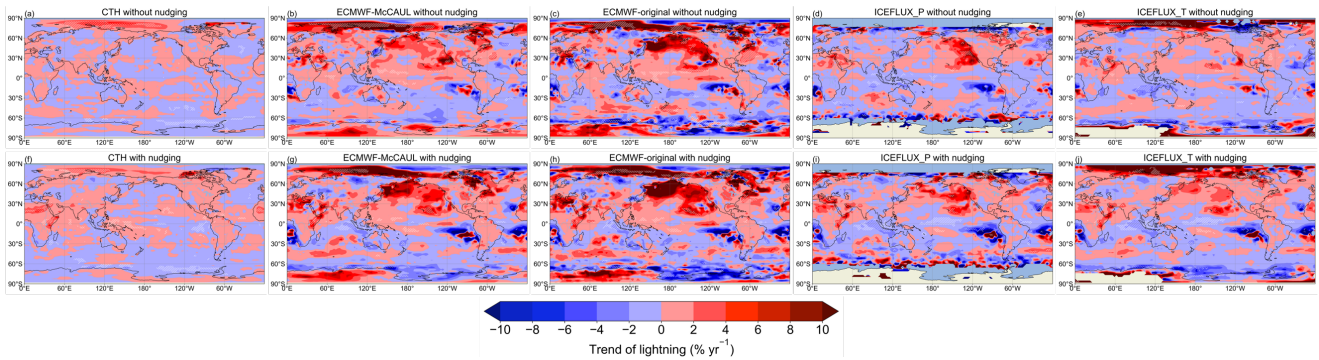
536

537 As displayed in Fig. 12, the positive lightning trends are generally enhanced by application of meteorological nudging. A
538 further investigation of the trends of CAPE during 2001–2020 discloses that the trends of global averaged CAPE are also
539 enhanced by application of meteorological nudging. Since higher CAPE means higher buoyancy in the updrafts, which led to
540 the higher lightning densities calculated by the lightning schemes used in this study. It is worth noting that even though the
541 meteorological fields (u , v , T) of nudged simulations are expected closer to the real situations, we cannot analogously deduce
542 that the lightning trends predicted by the nudged runs are also closer to the real situations. This is because the predicted
543 lightning trends are not only controlled by the meteorological fields but also controlled by many other physical processes
544 (e.g., cumulus parameterization).

545

546 Few studies have specifically examined the lightning trends predicted by the ECMWF schemes under the short-term surface
547 warming. When nudging was not applied, the ECMWF schemes predicted the increasing trends of lightning flash rates under

548 the short-term surface warming by factors of 4 (ECMWF-McCAUL scheme) and 5 (original ECMWF scheme) compared to
 549 the CTH scheme (Table 4).

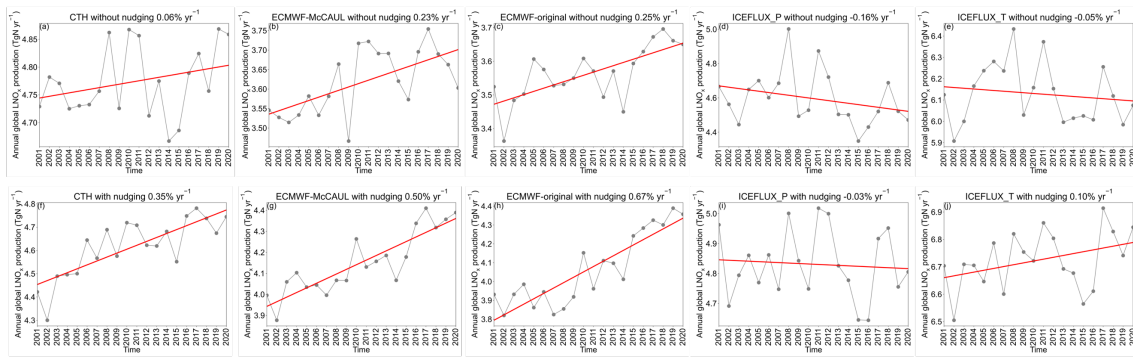


550
 551 **Figure 13: Changes in the lightning flash rate ($\% \text{ yr}^{-1}$) during 2001–2020 on the two-dimensional map. Changes at every point were**
 552 **calculated from the function of approximating curve for the 2001–2020 time-series data at each grid cell. Figures 13(a–e) show results**
 553 **without nudging; Figs. 13(f–j) show results with nudging. There are some missing values in the case of the ICEFLUX scheme because**
 554 **the upward cloud ice flux used is diagnosed as zero by the CHASER model typically within the high latitude regions.**
 555

556 Figure 13 shows a global map of changes in the lightning flash rate ($\% \text{ yr}^{-1}$) during 2001–2020. In Fig. 13, the area in which
 557 the trend was found to be significant by the Mann–Kendall rank statistic test (significance level inferred for 5%) is marked
 558 with hatched lines. As Fig. 13 shows, the distribution of trends simulated by the same lightning scheme is similar whether
 559 meteorological nudging was applied or not. As displayed in Fig. 13, in the Arctic region of the Eastern Hemisphere, both the
 560 CTH scheme and the ECMWF schemes showed an increasing trend of lightning. Earlier studies based on the World Wide
 561 Lightning Location Network (WWLLN) lightning observations have indicated that lightning densities in the Arctic increase
 562 concomitantly with increasing global mean surface temperature (Holzworth et al., 2021). Earlier studies indicate that the
 563 results of the CTH scheme and the ECMWF schemes are reasonable for the Arctic region of the Eastern Hemisphere. In the
 564 high latitude region of the Southern Hemisphere (60°S – 70°S), both the CTH scheme and the ECMWF schemes showed
 565 decreasing lightning trends. Lightning is rarely observed south of 60°S (Kelley et al., 2018). Moreover, the trends of
 566 lightning in this region expected to occur with the short-term surface warming remain highly uncertain. In some parts of the
 567 Northern Pacific Ocean, the ECMWF schemes and ICEFLUX scheme results showed increasing trends of lightning, which is
 568 consistent with results obtained from an earlier study (Walter and Buechler, 2008). All schemes show decreasing trends for
 569 lightning flash rates in Indonesia, although only the ICEFLUX scheme explicitly passed the significance test. In the North
 570 Atlantic, all schemes showed increasing lightning trends. Only the CTH scheme failed the significance test.

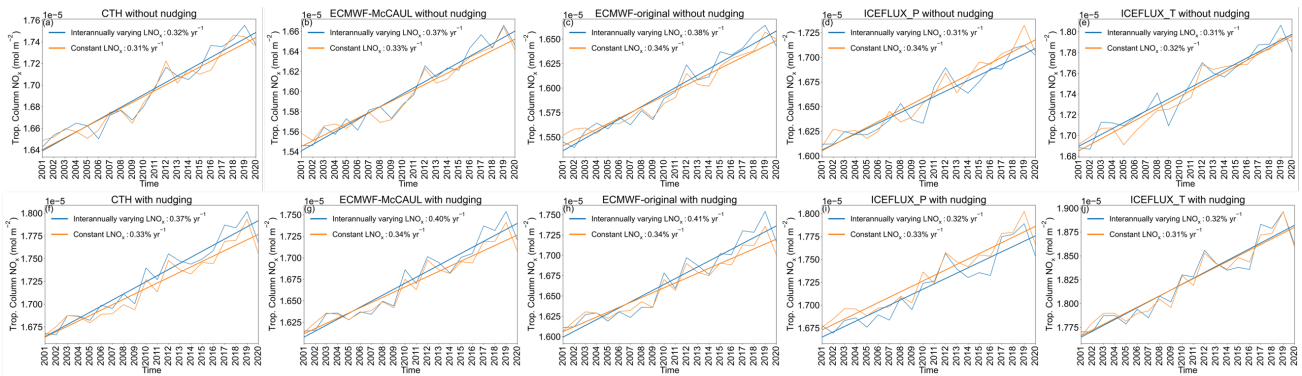
571 3.5 Effects of LNO_x emissions on trends of tropospheric O₃ and NO_x columns

572 The historical trends of lightning densities during 2001–2020 calculated using different lightning schemes have been
 573 discussed in Sect. 3.4. Increasing or decreasing trends of lightning can engender corresponding trends of LNO_x emissions,
 574 which can consequently influence trends of NO_x and O₃ concentrations. To ascertain the extent to which the LNO_x emissions
 575 influence NO_x and O₃ concentration trends, the effects of the LNO_x emissions on the trends of tropospheric NO_x and O₃
 576 columns have been estimated and discussed. We conducted two sets of experiments (Sect. 2.4), one of which interactively
 577 calculated LNO_x emission rates, whereas the other one maintained the 2001 LNO_x emission rates for simulations of the
 578 entire 20 years. The LNO_x emission effects on the trends of tropospheric NO_x and O₃ columns can be estimated
 579 quantitatively by comparing the results of these two sets of experiments. We also conducted the verification of the simulated
 580 trends of tropospheric NO_x and O₃ columns by the OMI satellite observations and the results are exhibited in Fig. S1 and Fig.
 581 S2. Generally, the model has well captured the trends of global averaged tropospheric NO₂ and O₃ columns even though the
 582 trends of both tropospheric NO₂ and O₃ columns are underestimated by the model. Overall, it is obvious that the modelled
 583 trends with interannually varying LNO_x emissions with nudging are most close to the OMI observations.



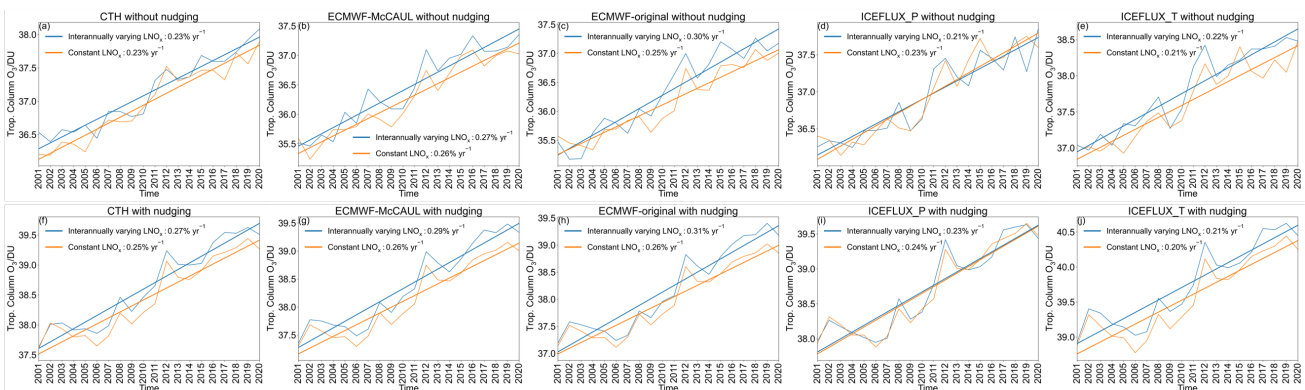
584
 585 **Figure 14: Trends of annual global LNO_x emissions calculated from simulation results (2001–2020) from different lightning schemes.**
 586 **Red lines are fitting curves. Figures 14(a–e) present results without nudging; Figs. 14(f–j) present results with nudging. The number**
 587 **in the title of each figure represents the trend corresponding to that figure in the unit of % yr⁻¹.**
 588

589 Figure 14 presents trends of annual global LNO_x emissions calculated from the simulation results (2001–2020) obtained
 590 using different lightning schemes. As Fig. 14 shows, the annual global LNO_x emission trends correspond to the trends of
 591 lightning presented in Fig. 12. Similar to the trends found for lightning, the trends of annual global LNO_x emissions are also
 592 increased by application of meteorological nudging. Only the ICEFLUX scheme simulated decreasing trends of annual
 593 global LNO_x emissions.



594
 595 **Figure 15: Trends of global mean tropospheric NO_x columns calculated from simulation results (2001–2020) using different lightning**
 596 **schemes. Straight lines in the figure are the fitting curves. The numbers in legends represent trends corresponding to that figure in**
 597 **the unit of % yr⁻¹. Figures 15(a–e) present results obtained without nudging; Figs. 15(f–j) present results obtained with nudging.**
 598

599 Figure 15 portrays trends of global mean tropospheric NO_x columns calculated from the first and second set of experiments
 600 (Table 2). As Fig. 14 and Fig. 15 depict, when the trends of annual global LNO_x emissions are not strong (e.g., Fig. 14a),
 601 their effects on the trends of global mean tropospheric NO_x columns are negligible. The marked increasing trends of annual
 602 global LNO_x emissions (Figs. 14f, g, h) led to great increases (12.12%–20.59%) of the increasing trends of tropospheric NO_x
 603 columns (Figs. 15f, g, h). In the case of the ICEFLUX_P scheme without nudging, because of the decreasing trends of LNO_x
 604 emissions, the increasing trends of the tropospheric NO_x columns decreased by around 10%.



605
 606 **Figure 16: Trends of global mean tropospheric O₃ columns calculated from simulation results (2001–2020) using different lightning**
 607 **schemes. Straight lines in the figure are the fitting curves. The number in the legend represents the trend corresponding to that**
 608 **figure in the unit of % yr⁻¹. Figures 16(a–e) present results obtained without nudging; Figs. 16(f–j) show results obtained with**
 609 **nudging.**

610

611 Figure 16 is similar to the results shown in Fig. 15, but for tropospheric O₃ columns. Because NO_x causes the formation of
612 O₃ by the fundamental chemical cycle of O_x–NO_x–HO_x, the trends of the global mean tropospheric O₃ columns are affected
613 strongly by trends of the global mean tropospheric NO_x columns. In some cases, the simulated trends of tropospheric O₃
614 columns are almost identical as portrayed in Figs. 16 a, b, e, i, j because the trends of tropospheric NO_x columns simulated
615 by the two sets of experiments are very similar (Figs. 15 a, b, e, i, j). As Fig. 14 and Fig. 16 show, the marked increasing
616 trends of annual global LNO_x emissions led to increases of the increasing trends of tropospheric O₃ columns by around 15%
617 (Figs. 16f, g, h). In the case of ICEFLUX_P without nudging, because of the decreasing trend of LNO_x emissions, the
618 increasing trend of the tropospheric O₃ columns decreased by around 10% (Fig. 16d). Note that the tropospheric NO_x or O₃
619 columns in 2001 simulated by the first set of experiments and the second set of experiments are not exactly the same. This is
620 because the blue lines show results with interactively calculated LNO_x emission rates (the time resolution is 10–30 minutes).
621 But the orange lines show results calculated by reading daily mean input data for LNO_x emission rates, which inhibits
622 interaction of LNO_x with meteorology in the model.

623

624 In conclusion, because the ICEFLUX scheme predicts the opposite trends of LNO_x emissions from the other lightning
625 schemes, they simulate opposite effects on the historical trends of global mean tropospheric NO_x and O₃ columns.
626 Furthermore, an evident trend of annual global LNO_x emissions has a strong effect on the trend of global mean tropospheric
627 NO_x and O₃ columns.

628 **4 Discussions and Conclusions**

629 Three new lightning schemes, the ICEFLUX, the original ECMWF, and the ECMWF-McCAUL schemes were implemented
630 into CHASER (MIROC), a global chemical climate model. Using LIS/OTD lightning observations as validation data, both
631 the ICEFLUX_P and ECMWF schemes simulated the spatial distribution of lightning more accurately on a global scale than
632 the CTH scheme did, and the lightning distribution in the ocean region was especially improved. The ECMWF-McCAUL
633 scheme showed the highest prediction accuracy for the spatial distribution of lightning on a global scale. It is noteworthy that
634 whilst the ice-based parametrisations showed superb prediction accuracy of lightning distribution under today's climate, they
635 have greater uncertainties associated with inputs, especially regarding the microphysics scheme used (Charn and Parishani,
636 2021).

637

638 To verify the LNO_x emissions of different lightning schemes, we used NO observations from ATom1 and ATom2. Overall,
639 both the ICEFLUX_P scheme and the ECMWF schemes partially reduced model biases typically over the dominant regions
640 of lightning activities compared to the CTH scheme. We also used TROPOMI tropospheric NO₂ columns to verify the LNO_x
641 emissions of different lightning schemes. Although the ICEFLUX_P and the ECMWF schemes have not shown
642 improvements of model biases of tropospheric NO₂ columns at an annual global scale, they generally led to an obvious
643 reduction of model biases in the prevailing seasons of lightning within the regions where LNO_x is a dominant source of NO_x.
644 Several studies have pointed out that the TROPOMI data used in this study biased negatively compared to the airborne or
645 ground-based observation data (Tack et al., 2021; Verhoelst et al., 2021; van Geffen et al., 2022). Since the TROPOMI data
646 used are generally negatively biased and the simulated tropospheric NO₂ columns are underestimated compared to the
647 TROPOMI observations. Therefore, the uncertainties that existed in the TROPOMI data have negligible impacts on the
648 conclusions of our study.

649

650 Effects of the newly implemented lightning schemes on the tropospheric chemical fields are evaluated compared to the CTH
651 scheme. Compared with the CTH scheme, the ECMWF schemes mainly led to a slight increase in NO_x, ozone, and OH
652 radical concentrations at low latitude regions and a decrease at middle-latitude to high-latitude regions. Effects of the
653 ICEFLUX_P scheme on the tropospheric chemical fields slightly differ from those of the ECMWF schemes. The
654 ICEFLUX_P scheme mainly causes a slight increase of NO_x, ozone, and OH radical concentrations from the tropics to the
655 Northern Hemisphere and a decrease in the concentrations of the three chemical species in the Southern Hemisphere except
656 the tropics. The commonality between the ECMWF schemes and the ICEFLUX_P scheme is that they both result in
657 decreasing concentrations of NO_x, ozone, and OH radical at the middle to high latitude regions of the Southern Hemisphere.
658 Although the newly implemented lightning schemes have little effect on the total oxidation capacity of the troposphere
659 compared to the CTH scheme, they led to marked changes of oxidation capacity in different regions of the atmosphere.

660

661 This study also analyzed the historical trends of lightning simulated by different lightning schemes under the short-term
662 surface warming during 2001–2020. The Mann–Kendall rank statistic was used to ascertain whether the lightning trends
663 were significant. Use of Mann–Kendall rank statistic tests revealed that all the simulated historical lightning trends are
664 significant, except the CTH and the ICEFLUX_T schemes without nudging and the ICEFLUX_P scheme with nudging, for
665 significance at 5%. All the lightning schemes predicted increasing lightning trends or no significant trends except the
666 ICEFLUX_P scheme without nudging, which predicted a decreasing lightning trend. The ICEFLUX_T scheme predicted a
667 decreasing trend without nudging even though the trend failed the significant test. If it's accepted that the non-inductive
668 charging mechanism is an appropriate basis for a lightning parametrisation, then the implication is that in the future if cloud
669 ice (and cloud ice fluxes) reduce then electrical charging will reduce too. This provides a line of scientific reasoning to
670 explain why lightning may reduce in the future. Moreover, findings showed that when nudging was not applied, the
671 ECMWF schemes predicted an increasing trend of lightning flash rate under the short-term surface warming by factors of 4
672 (ECMWF-McCAUL scheme) and 5 (original ECMWF scheme) compared to the CTH scheme. Although a considerable
673 degree of uncertainty remains in determining the sensitivity of lightning activity to changes in surface temperature on the
674 decadal timescale (Williams 2005), the majority of past estimates show the sensitivity tends average close to 10% K⁻¹ (Betz
675 et al., 2008, p. 521). This value is most consistent with the lightning increase rate predicted by the ECMWF-McCAUL
676 scheme without nudging in this study. Future research should be undertaken for specific examination of development of
677 lightning schemes that both accurately predict the global distribution of LNO_x, and which predict the changes in lightning
678 that are expected to occur concomitantly with global climate change. Finally, we quantitatively estimated the LNO_x emission
679 effects on tropospheric NO_x and O₃ column trends during 2001–2020. Results showed that a marked trend of annual global
680 LNO_x emissions significantly affects the trend of global mean tropospheric NO_x and O₃ columns.

681 **Code availability**

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

684 **Data availability**

685 The LIS/OTD data used for this study are available from <https://ghrc.nsstc.nasa.gov/hydro/?q=LRTS> (last access: 11 January
686 2022). The ATom data used for this study are available from https://daac.ornl.gov/ATOM/guides/ATom_merge.html (last
687 access: 11 January 2022). The TROPOMI data used for this study are available from
688 <https://s5phub.copernicus.eu/dhus/#/home> (last access: 11 January 2022). The OMI level-3 daily global gridded (0.25° ×
689 0.25°) Nitrogen Dioxide product (OMNO2d) used for this study is available from
690 https://disc.gsfc.nasa.gov/datasets/OMNO2d_003/summary (last access: 25 May 2022). The OMI/MSL tropospheric column
691 ozone data used for this study are available from https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html (last
692 access: 25 May 2022).

693 **Author contribution**

694 YFH introduced new lightning schemes into CHASER (MIROC) by adding new codes to CHASER (MIROC), conducted all
695 simulations, interpreted the results, and wrote the manuscript. KS developed the model code, conceived of the presented
696 idea, and supervised the findings of this work and the manuscript preparation. HMSH provided the TROPOMI data and the
697 relevant codes to pre-process the TROPOMI data.

698 **Competing interests**

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

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708 (https://ghrc.nsstc.nasa.gov/uso/ds_docs/lis_climatology/LISOTD_climatology_dataset.html, last access: 9 January 2022),
709 ATom data (<https://espo.nasa.gov/atom/content/ATom>, last access: 9 January 2022), and OMI satellite observation data
710 (https://disc.gsfc.nasa.gov/datasets/OMNO2d_003/summary, last access: 25 May 2022; [https://acd-
711 ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html](https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html), last access: 25 May 2022). We are grateful to ESA scientists
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