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Discussions

The impact of lightning on tropospheric ozone chemistry using a new global parametrisation

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Abstract. A lightning parametrisation based on upward cloud ice flux is implemented in a chemistry-climate model (CCM) for the first time. The UK Chemistry and Aerosols model is used to study the impact of these lightning nitric oxide (NO) emissions on ozone. Comparisons are then made between the new ice flux parametrisation and the commonly-used, cloud-top height parametrisation. The ice flux approach improves the simulation of lightning and the temporal correlations with ozone sonde measurements in the middle and upper troposphere. Peak values of ozone in these regions are attributed to high lightning NO emissions. The ice flux approach reduces the overestimation of tropical lightning apparent in this CCM when using the cloud-top approach. This results in less emission in the tropical upper troposphere and more in the extratropics when using the ice flux scheme. In the tropical upper troposphere the reduction in ozone concentration is around 5-10%. Surprisingly, there is only a small reduction in tropospheric ozone burden when using the ice flux approach. The greatest absolute change in ozone burden is found in the lower stratosphere suggesting that much of the ozone produced in the upper troposphere is transported to higher altitudes. Major differences in the frequency distribution of flash rates for the two approaches are found. The cloud-top height scheme has lower maximum flash rates and more mid-range flash rates than the ice flux scheme. The initial O_x (odd oxygen species) production associated with the frequency distribution of continental lightning is analysed to show that higher flash rates are less efficient at producing O_x - low flash rates produce around 10 times more O_x per flash than high-end flash rates. We find that the newly implemented lightning scheme performs favourably compared to the cloud-top scheme with respect to simulation of lightning and tropospheric ozone. This alternative lightning scheme shows spatial and temporal differences in ozone chemistry which may have implications for comparison on models and observations and for simulation of future changes in tropospheric ozone.

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1 Introduction

Lightning is a key source of nitric oxide (NO) in the troposphere. It is estimated to constitute around 10% of the global annual NO source (Schumann and Huntrieser, 2007). However, lightning has particular importance because it is the major source of NO directly in the free troposphere. In the middle and upper troposphere NO and NO_2 (together NO_x) have longer lifetimes and a disproportionately larger impact on tropospheric chemistry than emissions from the surface.

Through oxidation, NO is rapidly converted to NO_2 until an equilibrium is reached. NO_2 photolyses and forms atomic oxygen which reacts with an oxygen molecule to produce ozone, O_3. As a source of atomic oxygen, NO_2 is often considered together with O_3 as odd oxygen, O_x. Ozone acts as a greenhouse gas in the atmosphere and is most potent in the upper troposphere where temperature differences between the atmosphere and ground are greatest (Lacis et al., 1990; Dahlmann et al., 2011). Understanding lightning NO production and ozone formation in this region is important for determining the radiative forcing from ozone (Liaskos et al., 2015).

As reported by Lamarque et al. (2013), the parametrisation of lightning in chemistry transport and chemistry-climate models (CCMs) most often uses simulated cloud-top height to determine the flash rate as presented by Price and Rind (1992). However, this and other existing approaches have been shown to lead to large errors in the distribution of flashes compared to lightning observations (Tost et al., 2007). Several studies have shown that the global magnitude of lightning NO_x emissions is an important contributor to ozone and other trace gases especially in the upper tropical troposphere (Labrador et al., 2005; Wild, 2007; Liaskos et al., 2015). Each of these studies uses a single horizontal distribution of lightning so the impact of varying the lightning emission distribution is unknown. Murray et al. (2012, 2013) have shown that constraining simulated lightning to satellite observations results in a shift of activity from the tropics to extratropics, and that this constraint improves the representation of the ozone tropospheric column and its interannual variability. Finney et al. (2014) showed using reanalysis data that a similar shift in activity away from the tropics occurred when a more physically based parametrisation based on ice flux was applied.

The above studies and also that of Grewe et al. (2001) find that the largest impact of lightning emissions of trace gases occurs in the tropical upper troposphere. This is a particularly important region because it is the region of most efficient ozone production. Understanding how the magnitude of lightning flash rate or concentration of emissions affects ozone production is an ongoing area of research, and typically this has been done using simplified models of individual storms or small regions (Allen and Pickering, 2002; DeCaria et al., 2005; Apel et al., 2015). DeCaria et al. (2005) found that whilst there was little ozone enhancement at the time of the storm, there was much more ozone production downstream in the following days. They found a clear positive relationship between downstream ozone production and lightning NO_x concentration which was linear up to ~ 300 pptv but resulted in smaller ozone increases for NO_x increases above this concentration. Increasing ozone production downstream with more NO_x was also found by Apel et al. (2015). Allen and Pickering (2002) specifically explored the role of the flash frequency distribution on ozone production using a box model. They found that the cloud-top height scheme produces a high frequency of low flash rates and therefore $\mathrm{NO_x}$ concentrations which are unrealistic compared to observed flash rates. This results in a greater ozone production efficiency and therefore higher ozone production with the cloud-top height scheme. Differences in the frequency distribution between lightning parametrisations were also found across the broader region of the tropics and subtropics by Finney et al. (2014). The importance of differences in flash rate frequency distributions to ozone production over the global domain remains unknown.

In this study, the lightning parametrisation developed by Finney et al. (2014) which uses upward cloud ice flux at 440 hPa is implemented within the United Kingdom Chemistry and Aerosols model (UKCA). This parametrisation is closely linked to the Non-Inductive Charging Mechanism of thunderstorms (Reynolds et al., 1957) and was shown to perform well against existing parametrisations when applied to reanalysis data (Finney et al., 2014). Here the effect of the cloud-top height and ice flux parametrisations on tropospheric chemistry is quantified using a CCM, focussing especially on the location and frequency distributions. Section 2 describes the model and observational data used in the study. Section 3 compares the simulated lightning and ozone concentrations to observations. Section 4 analyses the ozone chemistry through use of $\mathrm{O_x}$ budgets. Section 5 then considers the differences in zonal and altitudinal distributions of chemical $\mathrm{O_x}$ production and ozone concentrations simulated for the different lightning schemes. Section 6 provides a novel approach to studying the effects of flash frequency distribution on ozone. Section 7 presents the conclusions.

2 Model and data description

2.1 Climate-chemistry model

The model used is the UK Chemistry and Aerosols model (UKCA) coupled to the atmosphere-only version of the UK Met Office Unified Model version 8.4. The atmosphere component is the Global Atmosphere 4.0 (GA4.0) as described by Walters et al. (2014). Tropospheric, stratospheric and aerosol chemistry are modelled, although the focus of this study is the troposphere. The UKCA tropospheric scheme is described and evaluated by O'Connor et al. (2014). The model is run at horizontal resolution N96 (1.875° longitude by 1.25° latitude). The vertical dimension has 85 terrain-following hybrid-height levels distributed from the surface to 85 km. The resolution is highest in the troposphere and lower stratosphere, with 65 levels up to $\sim$30 km. The model time step is 20 minutes with chemistry calculated on a 1 hour time step. The exception to this is for data used in section 6 where it was required that chemical reactions accurately coincide with time of emission and hence where the chemical time step was set to 20 minutes. The coupling is one-directional, applied only from the atmosphere to the chemistry scheme. This is so that the meteorology remains the same for all variations of the lightning scheme, and hence, differences in chemistry are solely due to differences in lightning $\mathrm{NO_x}$.

The cloud parametrisation (Walters et al., 2014) uses the Met Office Unified Model's prognostic cloud fraction and prognostic condensate (PC2) scheme (Wilson et al., 2008a, b) along with modifications to the cloud erosion parametrisation described by Morcrette (2012). PC2 uses prognostic variables for water vapour, liquid and ice mixing ratios as well as for liquid, ice and total cloud fraction. Cloud fields can be modified by shortwave and longwave radiation, boundary layer processes, convection, precipitation, small-scale mixing, advection and pressure changes due to large-scale vertical motion. The convection scheme calculates increments to the prognostic liquid and ice water contents by detraining condensate from the convective plume, whilst the cloud fractions are updated using the non-uniform forcing method of Bushell et al. (2003).

Simulations for this study were set up as a time-slice experiment using sea surface temperature and sea ice climatologies based on 1995-2005 analyses Reynolds et al. (2007), and emissions and background lower boundary GHG concentrations, including methane, are representative of the year 2000. A one year spin-up for each run was discarded and the following year used for analysis.

2.2 Lightning NO emission schemes

The flash rate in the lightning scheme in UKCA is based on cloud-top height by Price and Rind (1992, 1993), with energy per flash and NO emission per joule as parameters drawn from Schumann and Huntrieser (2007). The equations used to parametrise lightning are:

$$F_\mathrm{l} = 3.44 \times 10^{-5} H^{4.9} \tag{1}$$

$$F_\mathrm{o} = 6.2 \times 10^{-4} H^{1.73}, \tag{2}$$

where F is the total flash frequency ($\mathrm{fl.\,min^{-1}}$), H is the cloud-top height (km) and subscripts l and o are for land and ocean, respectively (Price and Rind, 1992). A resolution scaling factor, as suggested by Price and Rind (1994), is used although it is small and equal to 1.09. An area scaling factor is also applied to each grid cell which consists of the area of the cell divided by the area of a cell at 30° latitude.

This lightning $\mathrm{NO_x}$ scheme has been modified to have equal energy per cloud-to-ground and cloud-to-cloud flash based on recent literature (Ridley et al., 2005; Cooray et al., 2009; Ott et al., 2010). The energy of each flash is 1.2 GJ and NO production is 12.6×10^{16} NO molecules $\mathrm{J^{-1}}$ These correspond to 250 mol(NO) $\mathrm{fl.^{-1}}$ which is within the estimate of emission in the review by Schumann and Huntrieser (2007). It also ensures that changes in flash rate produce a proportional change in emission independent of location since different locations can have different proportions of cloud-to-ground and cloud-to-cloud flashes. The vertical emission distribution has been altered to use the recent prescribed distributions of Ott et al. (2010) and applied between the surface and cloud top. Whilst the Ott et al. (2010) approach is used for both lightning parametrisations, the resulting average global vertical distribution can vary because the two parametrisations distribute emissions in cells with different cloud top heights. This simulation with the cloud-top height approach will be referred to as CTH.

Two alternative simulations are also used within this study: 1) lightning emissions set to zero (ZERO), and 2) using the flash rate parametrisation of Finney et al. (2014) (ICEFLUX). The equations used by Finney et al. (2014) are:

$$f_{\mathrm{l}} = 6.58 \times 10^{-7} \phi_{\mathrm{ice}} \tag{3}$$

$$f_{\mathrm{o}} = 9.08 \times 10^{-8} \phi_{\mathrm{ice}}, \tag{4}$$

where f_{l} and f_{o} are the flash density (fl. $\mathrm{m^{-2}\,s^{-1}}$) of land and ocean, respectively. ϕ_{ice} is the upward ice flux at 440 hPa and is formed using the following equation:

$$\phi_{\mathrm{ice}} = \frac{q \times \Phi_{\mathrm{mass}}}{c}, \tag{5}$$

where q is specific cloud ice water content at 440 hPa ($\mathrm{kg\,kg^{-1}}$), Φ is the updraught mass flux at 440 hPa ($\mathrm{kg\,m^{-2}\,s^{-1}}$) and c is the fractional cloud cover at 440 hPa ($\mathrm{m^2\,m^{-2}}$). Upward ice flux was set to zero for instances where $c < 0.01\,\mathrm{m^2 m^{-2}}$. Where no convective cloud top is diagnosed, the flash rate is set to zero.

Both the CTH and ICEFLUX parametrisations when implemented in UKCA produce flash rates corresponding to global annual NO emissions within the range estimated by Schumann and Huntrieser (2007) of 2-8 $\mathrm{TgN\,yr^{-1}}$. However, for this study we choose to have the same flash rate and global annual $\mathrm{NO_x}$ emissions for both schemes. To achieve this the annual flash rate and NO per J were scaled to result in the satellite estimated flash rate by Cecil et al. (2014) of 46 fl. $\mathrm{s^{-1}}$ and then a total NO emission of 5 $\mathrm{TgN\,yr^{-1}}$. The flash rate scaling factors needed for implementation in UKCA were 1.57 for the Price and Rind (1992) scheme and 1.11 for the Finney et al. (2014) scheme. As stated earlier a parameter of 12.6×10^{16} NO molecules $\mathrm{J^{-1}}$ was used for both schemes. The factor applied to the ice flux parametrisation is similar to that used in Finney et al. (2014), who used a scaling of 1.09. This is some evidence for the parametrisation's robustness since the studies use different atmospheric models, however, the scaling may vary in other models.

2.3 Lightning observations

The global lightning flash rate observations used are a combined climatology product of satellite observations from the Optical Transient Detector (OTD) and the Lightning Imaging Sensor (LIS). The OTD observed between ±75° latitude from 1995-2000 while LIS observed between ±38° from 2001-2015 and a slightly narrower latitude range between 1998-2001. The satellites were low earth-orbit satellites so did not observe everywhere simultaneously. LIS, for example, took around 99 days to twice sample the full diurnal cycle at each location on the globe. The specific product used here is referred to as the High Resolution Monthly Climatology (HRMC) which provides 12 monthly values on a 1° horizontal resolution made up of all the measurements of OTD and LIS between 1995-2010. A detailed description of the product is provided by Cecil et al. (2014).

2.4 Ozone column and sonde observations

Two forms of ozone observations are used to compare and validate the model and lightning schemes. Firstly, a monthly climatology of tropospheric ozone column inferred by the difference between two satellite instrument datasets is used (Ziemke et al., 2011). These are the total column ozone estimated by the Ozone Monitoring Instrument (OMI) and the stratospheric column ozone estimated by the Microwave Limb Sounder (MLS). The climatology uses data covering October 2004 to December 2010. The production of the tropospheric column ozone climatology by Ziemke et al. (2011) uses the NCEP tropopause climatology so, for the purposes of evaluation, simulated ozone in this study is masked using the same tropopause. In Section 3.2, the simulated annual mean ozone column is regridded and compared directly to the satellite climatology without sampling along the satellite track.

Secondly, ozone sonde observations averaged into 4 latitude bands were used. The ozonesonde measurements are from datasets described by Logan (1999) (representative of 1980–1993) and Thompson (2003) (representative of 1997–2011), and consists of 48 stations, with 5, 15, 10 and 18 stations in the SH extratropics, SH tropics, NH tropics and NH extratropics respectively. In Section 3.2, the simulated annual ozone cycle is interpolated to the locations and pressure of the sonde measurements. The average of the interpolated points is then compared to the annual cycle of the sonde climatology without processing to sample the specific year or time of the sonde measurements. Both of these ozone datasets are the same as used in the ACCMIP multi-model comparison study by Young et al. (2013).

3 Comparison to observations

3.1 Global annual spatial and temporal lightning distributions

Using the combined OTD/LIS climatology allows extension of the evaluation over a smaller region made by Finney et al. (2014). Figure 1 shows the satellite annual flash rate climatology alongside the annual flash rate estimated by UKCA using CTH and ICEFLUX. The annual flash rate simulated by UKCA is broadly representative of the decade around the year 2000 as it uses SST and sea ice climatologies for that period. A spatial correlation of 0.78 between the flash rate climatology estimated by ICEFLUX and the satellite climatology is an improvement upon the correlation of flash rates estimated by CTH which is 0.65. Furthermore, the root mean square error (RMSE) of the ICEFLUX climatology to the satellite data of 3.7 fl. $\mathrm{km}^{-2}\,\mathrm{yr}^{-1}$ is favourably reduced compared to the 6.0 fl. $\mathrm{km}^{-2}\,\mathrm{yr}^{-1}$ RMSE of the CTH climatology.

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These results are similar to those found by Finney et al. (2014) who used offline ERA-Interim meteorology as the input to the parametrisation. Neither approach for simulating lightning achieves the observed ocean to land contrast despite using separate equations, and neither displays the large peak flash rate in central Africa. The ICEFLUX approach over the ocean provides a contrast to the CTH approach by being an overestimate instead of an underestimate compared to the satellite lightning observations. While not achieving the magnitude of the observed Central African peak the ICEFLUX scheme does yield closer agreement over the American and Asian tropical regions.

Figure 2 shows comparisons of the monthly mean flash rates for 4 latitude bands. The ICEFLUX approach simulates lightning well in the extratropics with good temporal correlations with LIS/OTD in both hemispheres. The correlation of CTH with LIS/OTD is higher in the southern extratropics but this improvement compared to ICEFLUX is contrasted by much larger absolute errors. Correlations for both approaches are lowest in the southern tropics. CTH also has very large absolute errors during December to April, with more detailed analysis (not shown) suggesting this is due to overestimation in the South American region. In the northern tropics the temporal correlation with LIS/OTD suggests CTH performs slightly better although Figure 2 shows that the CTH approach is not capturing the double peak characteristic of this latitude band. The ICEFLUX approach appears to simulate a double peak but it does not achieve the timing which leads to a poor correlation. In the northern tropics, both schemes failed to match the observed August peak of the American region and the duration of the lightning peak in the African region which lasts from June to September (not shown). The delay in the lightning peak that was apparent in annual cycles shown by Finney et al. (2014) over the tropics and subtropics is not so apparent here although there may be some delay in the southern tropics. The underestimation of ICEFLUX in the northern tropics and overestimation of CTH in the southern tropics found by Finney et al. (2014) is also found here. Overall, the ICEFLUX approach reduces the errors in the annual cycles of lightning and, on the whole, improves the correlation except in the northern tropics where both approaches for simulating lightning have difficulties.

3.2 Global annual spatial and temporal ozone distributions

Ozone has an average lifetime in the troposphere of a few weeks and can be transported long distances during that time. It can therefore be challenging to identify the sources of measured ozone but we use two types of measurements here to analyse how lightning emissions influence ozone distribution. Satellite column ozone measurements provide estimates of effect on the annual horizontal distribution of ozone whilst ozone sonde measurements demonstrate the altitudinal effect of lightning emissions on monthly varying ozone.

Comparisons with the MLS/OMI tropospheric column ozone climatology are made using Pearson correlations, root mean square error (RMSE) and mean bias assessments. The model ozone is masked to the troposphere by applying the NCEP tropopause climatology to each month and regridding to the 5° by 5° horizontal resolution of the MLS/OMI climatology. Table 1 gives the annual results for the three simulations using CTH, ICEFLUX and ZERO lightning.

The inclusion of lightning emissions from either scheme has a large effect on the amount of ozone in the column as shown by the reduced mean bias and RMSE compared to the ZERO simulation, however, there is little difference between the two lightning schemes. There is a slightly larger mean bias with the ICEFLUX approach. To analyse the error in distribution without the bias present an adjustment is made by subtracting the mean biases from the respective simulated ozone column distributions. Once this adjustment is made the ICEFLUX approach shows a slightly lower RMSE than the CTH approach (Table 1).

Figure 3 uses sonde measurements averaged over four latitudinal bands and taken at three pressure levels. The temporal correlations and mean biases of the model monthly means, interpolated to the same pressure and locations, against the sonde observations are shown.

Both lightning schemes show a reduction in mean bias compared to the ZERO run throughout all latitude bands and altitudes (Figure 3). The greatest impact of lightning is on the tropical, middle and upper troposphere. In these locations the ozone concentration simulated by the ICEFLUX scheme has a much better temporal correlation with sonde measurements than that simulated by the CTH scheme. The ICEFLUX approach has a larger bias than the CTH approach which is discussed further in the following paragraph.

Figure 4 shows the monthly ozone comparisons between sonde measurements and the model at 250 hPa and 500 hPa for the northern and southern tropics. It is clear that in the middle and upper troposphere the lightning scheme is important in achieving a reasonable magnitude of ozone, though both schemes still generally show an underestimate compared to observations (Figure 4). Other aspects of simulated ozone chemistry or uncertainty in total global lightning emissions, which is ±3 TgN on the 5 TgN used here, may also contribute to this bias. In Wild (2007) and Liaskos et al. (2015) the ozone burden and mean tropospheric column ozone respectively, scaled approximately linearly with increases in lightning emissions. Using the mean bias data in Table 1 we can calculate the mean increase in ozone column associated with each TgN emission from lightning. The average mean bias in ozone column of the ICEFLUX and CTH simulations is -3.0 DU, where as the mean bias of the ZERO simulation is -7.4 DU. Therefore, 5 TgN of lightning emissions has increased the mean ozone column by, on average, 4.4 DU. If we assume the effect of emissions is linear, these biases imply that the mean global effect of lightning on ozone column is 0.9 DU TgN^{-1}. Changing lightning emissions to 8 TgN could increase the ozone column by 2.7 DU and result in a bias of less than 1 DU. Such bias potentially introduced by the uncertainty in total emissions or other aspects of the model is much greater than the difference in mean bias between the two lightning schemes given in Table 1. Therefore, the small difference in mean bias between the two lightning schemes does not necessarily imply greater accuracy, instead the correlation values provide a more useful evaluation of parametrisation success.

In Figure 4 some features of the results from the simulations with lightning emissions stand out as being different from that in the ZERO run. These features occur as ozone peaks in April in the northern tropics (most notably at 500 hPa)(Figure 4D) and in October in the southern tropics (most notably at 250 hPa)(Figure 4A). The northern tropics peak in ozone improves the comparison to sondes at 500 hPa, if slightly underestimated. However, the 250 hPa April peak in Figure 4B does not appear in any of the model simulations. Potentially, the modelled advection is not transporting the lightning emissions or ozone produced to high enough altitudes. An anomalous southern tropical peak in March in Figures 4A and C, particularly shown by CTH, is not shown in the sonde measurements, but this corresponds to a month where the CTH scheme especially is overestimating lightning, as seen in Figure 2. The ICEFLUX scheme is a much closer match to the lightning activity in the southern tropics in March and correspondingly the modelled ozone is less anomalous compared to the ozone sonde measurements in that month. The well modelled lightning activity in the southern tropics in October (Figure 2C) results in a correctly matched peak in the ozone sonde measurements at both pressure levels which does not occur in the ZERO run. From these comparisons to ozone sondes we conclude that the lightning emissions have impacts in particular months which include the months of peak ozone. Figure 2 shows that these are not necessarily the month of highest lightning activity in the region, but instead as the lightning activity builds in the region. It may be of particular use for field campaigns studying the chemical impact of lightning to focus on these months.

4 The influence of lightning on the global annual O_x budget

The O_x budget considers the production and loss of odd oxygen in the troposphere. Several studies have used O_x budgets to study tropospheric ozone (Stevenson et al., 2006; Wu et al., 2007; Young et al., 2013; Banerjee et al., 2014). Here, the O_x approach has particular use because it responds more directly to the emission of NO than O_3 which may form in outflows of storms and take several days to fully convert between O_x species (Apel et al., 2015).

There are different definitions of O_x family species and here we use a broad definition that includes O_3, $O(1D)$, $O(3P)$, NO_2 and several NO_y species (Wu et al., 2007). The O_x species and the different terms of the budget are illustrated in Figure 5. Of particular relevance to this study is the chemical production of O_x, the majority of which occurs through oxidation of NO to NO_2.

The global annual O_x budgets for CTH, ICEFLUX and ZERO are given in Table 2. The terms are for the troposphere which is diagnosed each time step using the modelled meteorology to determine a tropopause defined as a combination of the pressures at 380 K and at 2 PVU. Clearly, the ZERO run demonstrates the large control that lightning has on these budget terms with changes of around 20% in the ozone burden and chemical production and losses (Table 2). Also because of reduced ozone concentrations, there is reduced deposition. The lifetime of ozone is less affected compared to other terms because the ozone burden has reduced as well as the loss terms. Using no lightning (ZERO) corresponds to a reduction of 5 TgN emissions over the year - less than the range of estimates for lightning emissions of 2-8 TgN emissions (Schumann and Huntrieser, 2007). Therefore large changes in O_x budget terms can be expected within the uncertainty range of the global lightning NO_x emission total.

It would seem that for constant emissions of 5 TgN and a reasonable change in the flash rate distribution using ICEFLUX, there are only small changes in the global O_x budget terms but this does not consider changes in composition of the lower stratosphere. Previous studies have found ozone produced from lightning is transported into the lower stratosphere (Grewe et al., 2002; Banerjee et al., 2014). In this study, we quantify the different transport between the two lightning schemes by considering changes in total atmospheric ozone burden against changes in tropospheric ozone burden. The difference in simulated total atmospheric ozone burden between ICEFLUX and CTH is -13 Tg. Given the -6 Tg difference in the troposphere, this means that the majority of the difference in ozone burden ($\sim$55%) occurs in the stratosphere. On the other hand, the difference in total atmospheric ozone burden in the ZERO run was -91 Tg. The tropospheric ozone burden difference was -62 Tg so accounts for around two thirds of the total difference in this case. The ICEFLUX approach has resulted in less lightning emissions in the upper tropical troposphere and therefore less ozone is available in the region to be transported into the stratosphere. We see that such a change in the lightning distribution, but maintaining the same level of total emissions, results in reduced net ozone production but that much, and even the majority, of this reduction in ozone can occur in lower stratospheric ozone.

5 Differences in the zonal-altitudinal distributions of O_x and O_3 between the two lightning schemes

The previous section showed that the global tropospheric O_x budget is affected principally by the magnitude of emissions and not the location of emissions as occurs in the switch from the CTH to the ICEFLUX scheme. This section now considers changes in the zonal and altitudinal location of O_x chemistry and ozone concentration as a result of changes in the lightning emission distribution. The zonal-altitudinal net chemical O_x production, as well as its components of gross production and loss, are shown in Figure 6A-C for the CTH scheme as well as changes as a result of using ICEFLUX instead of CTH in Figure 6D-F.

The difference in net O_x production when using the ICEFLUX scheme compared to the CTH scheme is dominated by the change in gross production (Figure 6D and E). Figure 6E shows a shift away from the tropical upper troposphere to the middle troposphere and the subtropics. There is over a 10% reduction in the upper troposphere net production and 100% changes in the subtropics (Figure 6D). However, the high subtropical percentage change is principally due to small net production in these regions. The changes in O_x production result as a shift in emissions which happens by: 1) reduced and more realistic lightning in the tropics (see Figure 7), and 2) decoupling of the vertical and horizontal emissions distributions by not using cloud-top in both aspects (as is the case in CTH). The latter means by basing the horizontal lightning distribution on cloud-top height and then distributing emissions to cloud-top, emissions are most effectively distributed to higher altitudes. Hence, a lightning parametrisation for which the horizontal distribution is different to that of cloud-top height will, to some extent, naturally distribute emissions at lower altitudes. This is demonstrated best in Figure 6E which shows gross production in the northern tropics. Whilst both lightning schemes have similar total lightning at these latitudes (shown in Figure 7), and therefore similar column O_x production, the gross O_x production occurs less in the upper troposphere and more in the middle troposphere when using the ICEFLUX scheme.

It is consistent with observations of lightning, that there is less lightning in the tropics than estimated by CTH here. It is also consistent with current understanding that the most intense lightning flash rates do not always occur in the highest clouds. We would therefore suggest that the change to the net O_x production of ICEFLUX is a more realistic representation of the distribution of production than with CTH. The improved sonde correlations presented in section 3.2 support this conclusion.

Whilst O_x gross production changes, mainly representing oxidation of NO to NO_2, show a close resemblance to the lightning NO emissions changes they are only part of the picture with regard to changes in the distribution of ozone. This is because the lifetime of ozone is much longer than the timescales for NO forming an equilibrium with NO_2. Furthermore, other O_x species are transported before then forming ozone. The difference in O_x production (Figure 6) between the two lightning schemes influences not only ozone locally but also downwind where ozone is transported to. Figure 8 presents the percentage changes in ozone distribution as a result of using the ICEFLUX scheme instead of the CTH scheme.

There is reduced tropical upper tropospheric ozone of up to 10% (Figure 8) due to reduced NO emission in that region. This results in less ozone transported into the lower stratosphere under the ICEFLUX scheme compared to the CTH scheme. The lower stratospheric ozone may also be lower due to less NO_x being available for transport, and therefore reduced chemical production in the stratosphere. Whilst ozone is lower in most of the lower stratosphere in the simulation with ICEFLUX the percentage changes are largest (up to 5%) nearer to the tropopause.

In the middle and lower tropical troposphere there is also a reduction in ozone concentration (Figure 8) despite increased net O_x production (Figure 6D). In the southern tropics this is because the increase in net O_x production is due to reduced O_x loss which is likely caused by the reduced ozone concentration itself. The reduced ozone concentrations in the northern and southern tropics is a result of less ozone available to be transported from the upper troposphere within the Hadley cell or other vertical subsidence. Note that both schemes experience the same meteorology because the chemistry is not coupled. The percentage changes in ozone in the northern tropics are less than in the southern tropics (Figure 8). This is likely to be in part due to offsetting through increased lightning emissions in the northern tropical middle troposphere. Finally, the increased lightning emissions in the subtropics with the ICEFLUX compared to the CTH scheme results in small changes in ozone throughout the extratropics.

It is worth noting that OH concentrations (not shown) respond in a similar manner to ozone concentration with the change from the CTH to the ICEFLUX scheme. These changes are more localised to emission changes but are still apparent in the lower stratosphere and extratropics. A change from the CTH to ICEFLUX scheme results in only small changes in the methane lifetime as a result of the changes in OH. Hence, in this setup we do not expect the ozone changes would be greatly modified with the use of interactive methane.

Liaskos et al. (2015) identified that even with the same total global emissions, the magnitude and distribution of radiative forcing resulting from lightning emissions is dependent on the method for distributing the emissions horizontally and vertically. The changes in zonal-altitudinal distribution discussed in this section show that these changes could be expected as a result of changes in ozone in the upper troposphere.

6 Frequency distributions of lightning and associated O_x production

Lightning is a highly dynamic process. This section presents analysis of the frequency distribution of flash rates as a means to study the finer scale effects.

The CTH scheme simulates extremely low flash rates over the ocean. For instance, the maximum September oceanic flash rate using CTH was $1.1 \times 10^{-4} fl.km^{-2} 20min^{-1}$ where as using ICEFLUX the maximum was over 100 times greater. This difference is not surprising given the difference in annual oceanic lightning activity shown in Figure 1. CTH tends to underestimate ocean lightning compared to satellite observations. The focus here will be on continental lightning. Other studies of frequency distribution in the literature have also focussed on continental locations so this work can be more directly compared to those.

Figure 9 shows the hourly continental flash rate frequency distribution for one model month (September). September was chosen as a month with a reasonable balance of lightning activity in between the hemispheres and where total lightning activity, and therefore emissions, was similar for the two lightning schemes.

When compared to the frequency distribution simulated by ICEFLUX, CTH has lower maximum flash rates, fewer occurrences of low flash rates and more occurrences of mid-range flash rates (Figure 9). Other studies have drawn similar conclusions regarding the frequency distributions of CTH when comparing to other parametrisations and lightning observations (Allen and Pickering, 2002; Wong et al., 2013; Finney et al., 2014). In Figure 9, the CTH frequency distribution displays some unusual periodic characteristics in the occurrence rate, most notably towards high flash frequencies.

These features are also apparent in the cloud-resolving simulations presented in Wong et al. (2013). We suggest here that these features may arise due to discretised nature of the cloud-top height input variable.

The importance of the global flash rate frequency distribution to atmospheric chemistry frequency distributions is currently unknown but simplified model studies have suggested some key features:

- Compared to a set of observations over the US, a simulation using the CTH approach led to a greater ozone production efficiency due to the non-linear nature of ozone production and $\mathrm{NO_x}$ (Allen and Pickering, 2002).
- Total ozone production increased approximately linearly up to 300 pptv of lightning $\mathrm{NO_x}$ and then increased at a slower rate beyond that. This may be due to the ozone production approaching the maximum possible for the given altitude, solar zenith angle and $\mathrm{HO_x}$ concentration (DeCaria et al., 2005).

In the following analysis we consider $\mathrm{O_x}$ production rather than ozone production because it exhibits a more immediate response to NO emission. This is important given the difficulty and errors associated with tracking ozone production associated with each emission source in a global model. However, there are some comparable results which we will compare to the previous findings above, as well as new insights into the consequences of different frequency distributions and lightning parametrisations.

Figure 10 presents two metrics of the gross column chemical $\mathrm{O_x}$ production resulting from continental lightning in each of the frequency bins of Figure 9. The metrics are: A) the mean column $\mathrm{O_x}$ production, and B) the mean $\mathrm{O_x}$ production per flash. Each flash corresponds to 250 mol(NO) emission so the $\mathrm{O_x}$ production per mole of emission can easily be inferred from the $\mathrm{O_x}$ production per flash. $\mathrm{O_x}$ production resulting from lightning is calculated as the difference between the model run with lightning and the model run with no lightning, using the grid cells from the no lightning run that correspond to the cells used in each bin for the relevant lightning parametrisation. This means that this work is focussing on the *initial* $\mathrm{O_x}$ production resulting from emission. This initial $\mathrm{O_x}$ production has been calculated to be approximately 15% of total $\mathrm{O_x}$ production associated with lightning for both parametrisations. The calculation was made as the difference between the total $\mathrm{O_x}$ production resulting from lightning in the sampled grid cells and the total $\mathrm{O_x}$ production resulting from lightning over the whole globe in all time steps. The remaining 85% of production must occur after the initial time step and be a result of advected emissions or changes to the large-scale distributions of constituents such as ozone or OH as discussed in section 5.

The mean column $\mathrm{O_x}$ production in Figure 10A shows, as expected, that increasing flash rate (i.e. more NO emissions in a cell) results in increased column $\mathrm{O_x}$ production. The higher extreme flash rates of ICEFLUX compared to CTH result in greater column $\mathrm{O_x}$ productions as a result of individual occurrences. The increase is linear up to approximately 0.02 fl. $\mathrm{km}^{-2} 20 min^{-1}$ at which point the two schemes produce 1 to 1.5 kg of O_x. Beyond this point, the O_x production simulated by the ICEFLUX approach increases still linearly but with a shallower gradient. The ICEFLUX scheme produces less O_x for a given flash rate than the CTH scheme at higher flash rates but more at lower flash rates (Figure 10A). This is due to emissions from high flash rates in ICEFLUX not necessarily being distributed to such high altitudes as with CTH. At the higher altitudes that emissions reach when using the CTH scheme, NO_x has a longer lifetime, as discussed in section 5. Conversely, in the ICEFLUX scheme, lower flash rates can occur in relatively deeper cloud so in these there can be greater O_x production efficiency compared to the CTH scheme because the CTH scheme will always place these low flash rates at lower altitudes. On larger scales, whilst high extreme flash rates produce more O_x, they occur relatively infrequently so do not greatly affect the global O_x budget.

Figure 10B shows the mean column O_x production per flash for each flash rate bin. It is derived by dividing the data in Figure 10A by the mid-point flash rate of each bin. Whilst Figure 10A shows that lower flash rates produce less O_x, they do produce O_x more efficiently than higher flash rates. Flash rates of 0.0005 fl. $\mathrm{km}^{-2} 20\,\mathrm{min}^{-1}$ produce $\sim$ 10 times more O_x per flash than flash rates of 0.05 fl. $\mathrm{km}^{-2} 20\mathrm{min}^{-1}$.ICEFLUX displays the greatest contrast in efficiency between high and low flash rates of the two parametrisations (Figure 10B). As with the column mean production, because the CTH scheme places the most emissions in the highest cloud tops it is more efficient at producing O_x at higher flash rates but the ICEFLUX scheme is more so at lower flash rates. The range of initial O_x production per mol of emission is 25 mol(O_x) mol^{-1}(NO) at low flash rates for ICEFLUX to less than 2 $\mathrm{mol}(O_x)\,\mathrm{mol}^{-1}(\mathrm{NO})$ for the highest flash rates in the ICEFLUX scheme (Figure 10B).

In summary, we find similarly to Allen and Pickering (2002) that O_x production becomes less efficient at higher flash rates. It is important to consider that in our case the higher flash rates are less efficient at the point of emission - the emissions may go on to produce O_x elsewhere following advection. Also, similarly to DeCaria et al. (2005), we find that the mean column O_x production increases linearly up to a point, in our case 0.05 fl. $\mathrm{km}^{-2} 20\mathrm{min}^{-1}$, then increases at a slower, but still linear rate beyond that. New insights provided through the use of a global model are:

- Both lightning schemes produce about 15% of the O_x associated with lightning at the time of emission
- For the CTH approach, oceanic flash rates are so low that associated O_x production at the time of emission is negligible for the global production
- Because CTH places the most emissions in the highest clouds (where NO_x lifetime is longer), more O_x is produced by the CTH scheme than ICEFLUX at high flash rates, but ICEFLUX produces more at low flash rates
- Initial O_x production per flash is approximately 10 times greater for low flash rates than high-end flash rates

These findings regarding the O_x production per flash provide a useful metric to evaluate lightning parametrisations with observations. Several differences between the CTH and ICEFLUX scheme suggest further study is needed to determine the true nature of O_x production. For instance, the almost negligible proportion of O_x production that will occur over the ocean when using the CTH scheme due to very low flash rates would benefit from oceanic measurements of ozone and NO_x in the vicinity of storms. An extension of the work here could be to run idealised experiments of pulse lightning emissions in a global model to see how the O_x and ozone production develop with time and hence, assess the lag between NO emission and ozone production.

7 Conclusions

A new lightning parametrisation based on upward cloud ice flux, developed by Finney et al. (2014), has been implemented in a chemistry-climate model (UKCA) for the first time. It is a physically based parametrisation closely linked to the Non-Inductive Charging Mechanism of thunderstorms. The horizontal distribution and annual cycle of flash rates as calculated through the new ice flux approach and the commonly-used, cloud-top height approach were compared to the LIS/OTD satellite climatology. The ice flux approach is shown to generally improve upon the performance of the cloud-top height approach. Of particular importance is the realistic representation of the zonal distribution of lightning using the ice flux approach, whereas the cloud-top height approach overestimates the amount of tropical lightning and underestimates extra-tropical lightning.

The ice flux approach greatly improves upon the cloud-top height approach in UKCA with regards to the temporal correlation to the observed annual cycle of ozone in the middle and upper tropical troposphere. Through considering a simulation without emissions and the simulated annual cycle of lightning, it is clear that the ice flux approach reduces the biases in ozone in months where the cloud-top height approach has the largest errors in simulating lightning.

The zonal flash rate distribution when using the ice flux approach instead of the cloud-top height approach results in a shift of O_x production away from the upper tropical troposphere. As a consequence there is a 5-10% reduction in upper tropical tropospheric ozone concentration along with smaller reductions in the lower stratosphere and small increases in the extratropical troposphere. These changes in ozone concentration are a result of the change in distribution of lightning emissions only, the total global emissions are the same for both schemes. We conclude that biases in zonal lightning distribution of the cloud-top height scheme increase ozone in the upper tropical troposphere and, as demonstrated by comparison to ozone sondes, this reduces the correlation to observations in ozone annual cycle in this region.

Analysis of the continental flash rate frequency distribution shows the cloud-top height approach has lower high-end extreme flash rates, more frequent mid-range flash rates and less frequent low-end flash rates, compared to the frequency distribution using the ice flux approach. Such features simulated by the cloud-top height approach have been found in comparisons to the observed frequency distribution over the US and this current evidence suggests such a frequency distribution is unrealistic. We apply a novel analysis to determine the impact of the differences in flash rate frequency distribution on the initial O_x production resulting from lightning emissions. As expected, the higher the flash rate, the more O_x is initially produced. However, the O_x production efficiency reduces for higher flash rates; lower flash rates initially produce approximately 10 times as much O_x as higher flash rates. Further study is warranted to determine how emissions produce ozone downstream of a storm in complex chemistry models, but the result here is relevant to aircraft campaigns measuring NO_x and ozone near to the thunderstorms. It would be useful to study such measurements to determine if less intense storms exhibit such a difference in O_x production efficiency.

The global lightning parametrisation of Finney et al. (2014) using upward cloud ice flux has proven to be robust at simulating present-day annual distributions of lightning and tropospheric ozone. The reduced ozone in the upper tropical troposphere could be important for the understanding of ozone radiative forcing. In addition, the differences in the frequency distribution when using different lightning schemes is shown to affect the chemical O_x production. The parametrisation is appropriate for testing in other chemistry transport and chemistry-climate models where it will be important to determine how the parametrisation behaves using different convective schemes. Furthermore, this new parametrisation offers an opportunity to diversify the estimates of the sensitivity of lightning to climate change which will be the focus of future work.

8 Author contribution

DLF, RMD, OW and NLA designed the experiments and interpreted the results. DLF performed the analysis. DLF and NLA developed the code and ran simulations. DLF prepared the manuscript with contributions from all co-authors.

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Table 1. Spatial comparisons of correlation, errors and bias of annual tropospheric ozone column between model runs and the MLS/OMI satellite climatology product. Adjusted root mean square error (RMSE) refers to the RMSE following the subtraction of the mean bias from the field.

Run	r	RMSE	Mean bias	adjusted RMSE
CTH	0.82	5.5	-2.8	4.1
ICEFLUX	0.84	5.7	-3.2	3.9
ZERO	0.83	10.7	-7.4	4.6

Table 2. Global annual tropospheric O_x budget terms for the year 2000 for three different simulations: CTH, ICEFLUX and Zero. All terms in $\mathrm{Tg\,yr^{-1}}$ except Burden which is in Tg and lifetime which is in days. In addition to the usual budget terms the whole atmospheric ozone burden is included.

	CTH	ICEFLUX	ZERO
Chem. prod.	4472	4443 (-1%)	3638 (-19%)
Chem. loss	3848	3821 (-1%)	3115 (-19%)
Net chem. prod.	624	622 (0%)	522 (-16%)
Deposition	1006	1006 (0%)	899 (-11%)
Strat. influx*	382	384 (0%)	376 (-2%)
Trop. O_3 burden	267	261 (-2%)	205 (-23%)
Whole atm. O_3 burden	3253	3240	3162
τ_{O_3}	19.8	19.5 (-2%)	18.4 (-7%)

* Stratospheric influx is inferred to complete the O_x budget through balancing the chemical loss and production and deposition.

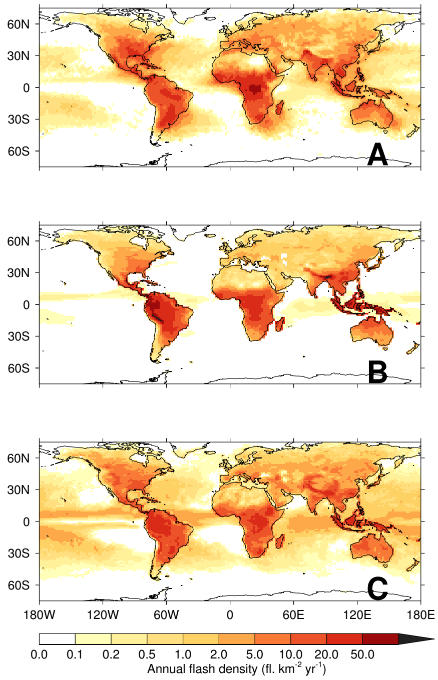


Figure 1. Annual flash rates from (A) a combined climatology from LIS/OTD satellite observations spanning 1995-2010, (B) the CTH scheme using the year 2000 of UKCA output and (C) the ICEFLUX scheme using the year 2000 of UKCA output. The horizontal resolution of the climatology product has been degraded to match that of the model which is 1.875° longitude by 1.25° latitude.

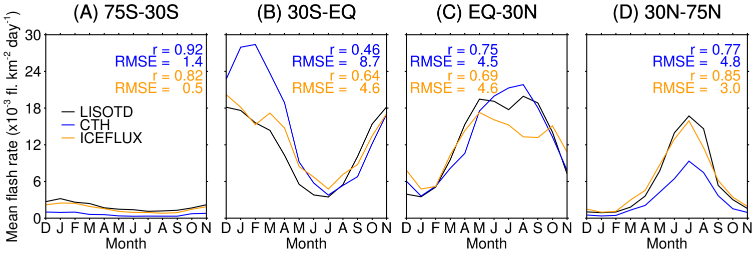
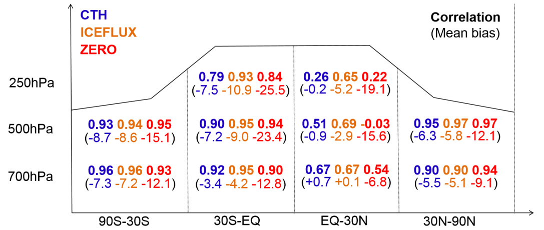


Figure 2. Mean monthly flash rate averaged over four latitudinal bands for the two different schemes for 2000 and the LIS/OTD climatology spanning 1995-2010. The points use one year of UKCA model output and a combined climatology from LIS/OTD satellite observations spanning 1995-2010. Also given are the temporal correlations (r) between the CTH model (blue) and LIS/OTD and between ICEFLUX (orange) and LIS/OTD. The corresponding root mean square errors (RMSE) are given in units of 10^{-3} fl. $\mathrm{km}^{-2}\mathrm{yr}^{-1}$.



	90S-30S	30S-EQ	EQ-30N	30N-90N
250hPa		0.79 0.93 0.84 (-7.5 -10.9 -25.5)	0.26 0.65 0.22 (-0.2 -5.2 -19.1)	
500hPa	0.93 0.94 0.95 (-8.7 -8.6 -15.1)	0.90 0.95 0.94 (-7.2 -9.0 -23.4)	0.51 0.69 -0.03 (-0.9 -2.9 -15.6)	0.95 0.97 0.97 (-6.3 -5.8 -12.1)
700hPa	0.96 0.96 0.93 (-7.3 -7.2 -12.1)	0.92 0.95 0.90 (-3.4 -4.2 -12.8)	0.67 0.67 0.54 (+0.7 +0.1 -6.8)	0.90 0.90 0.94 (-5.5 -5.1 -9.1)

Figure 3. Temporal correlations and mean biases of the annual cycle of modelled ozone in UKCA over the year 2000 compared to a climatology of ozone sonde measurements averaged over 1980-1993 and 1997-2011. The simulated ozone data was interpolated to the location and pressure level of the sonde measurements. The sonde and modelled ozone were then averaged into 4 latitude bands which correspond to the bands used in Figure 2.

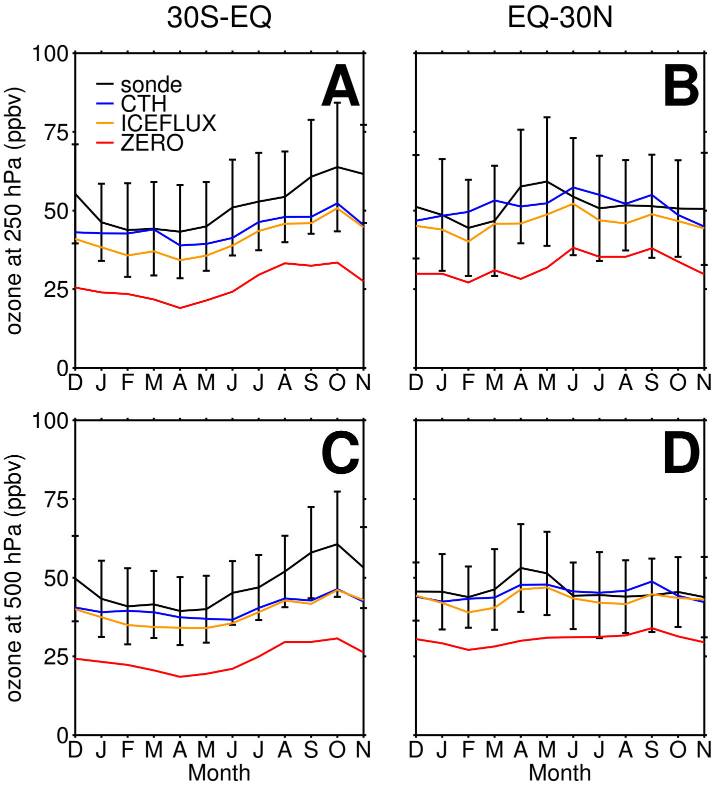


Figure 4. Middle and upper tropospheric UKCA simulated ozone concentration for the year 2000 compared to a climatology of sonde measurements averaged over 1980-1993 and 1997-2011. These cycles correspond to the 500 hPa and 250 hPa correlations for 30S-EQ and EQ-30N in Figure 3. The vertical black bars show the average interannual standard deviation for each group of stations.

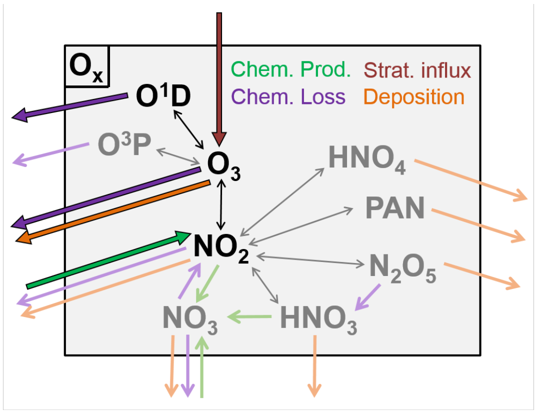


Figure 5. The UKCA definition of O_x species and the O_x budget. Major contributors are shown in bright colours and black outlines, minor contributors in pale colours. Black arrows are reactions between O_x species and therefore result in no production or loss. The burden of O_x is dominated by O_3.

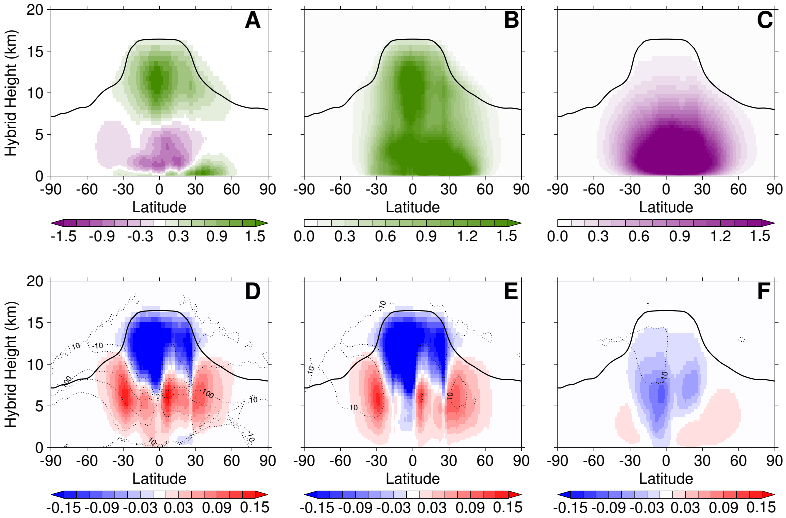


Figure 6. Annual total zonal-altitudinal distributions of O_x reaction fluxes for CTH for the year 2000. These fluxes are A) Net production, B) gross production, and C) gross loss of O_x. The respective differences between simulations using the ICEFLUX scheme and the CTH scheme are shown in D-F. All units are Tg(O_3). Values are annual and meridional totals. The solid line is the annual mean tropopause and dashed lines contour 10% and 100% changes. The O_x fluxes were masked with the model tropopause every time step.

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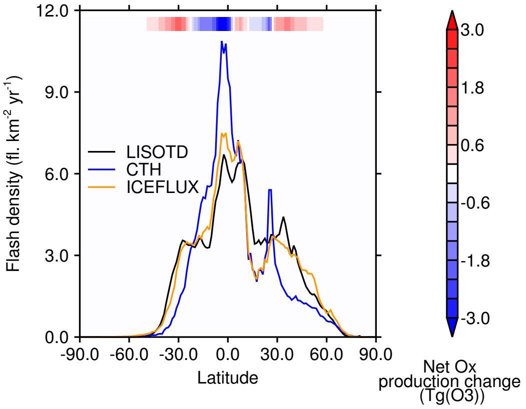

Figure 7. Zonal mean lightning flash rate from the LIS/OTD climatology and as modelled by CTH and ICEFLUX. The zonal changes in net tropospheric column O_x production (ICEFLUX-CTH) are shown by the colour bar. The units of O_x are expressed as a mass of ozone.

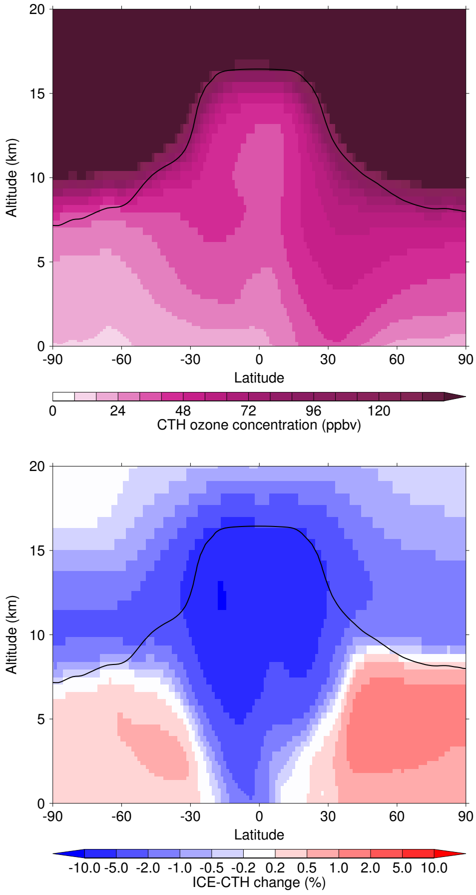

Figure 8. Annual mean distribution of ozone concentration modelled using the CTH approach, and the percentage difference between ICEFLUX and CTH simulated ozone concentration. The solid line shows the mean annual tropopause as diagnosed using the modelled meteorology.

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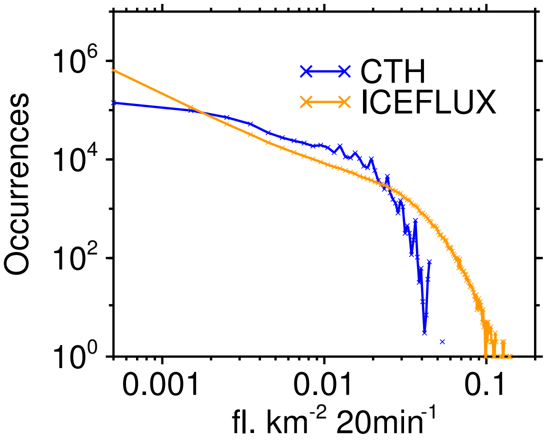

Figure 9. Frequency distribution of continental lightning flash rates using all time steps, for one month (September 2000) as modelled by the CTH and ICEFLUX schemes. The binsize used is 0.001 fl. $\mathrm{km}^{-2}20\mathrm{min}^{-1}$ with crosses placed at the centre value of each bin.

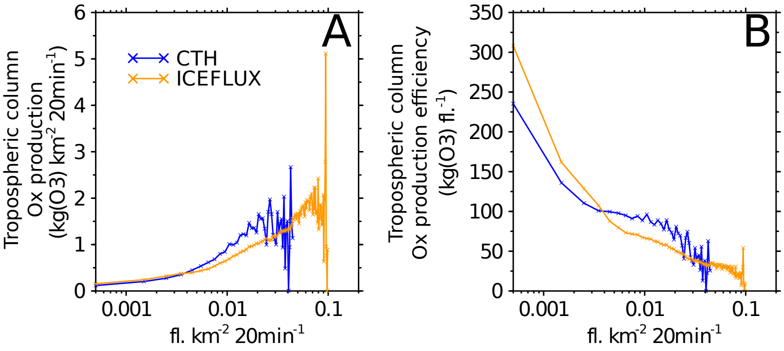

Figure 10. Two metrics of intial gross column O_x production as a result of continental lightning simulated by the CTH and ICEFLUX schemes. The cells used in each bin correspond to those used in Figure 9. The metrics are A) mean column O_x production in each bin, and B) mean column O_x production per flash in each bin. The O_x production resulting from lightning was determined by subtracting the column O_x production in the no lightning run from the each lightning parametrisation for the corresponding cells. To reduce noisiness, only data is only plotted up to the highest bin of each parametrisation where there are at least two occurrences in Figure 9. The units of O_x are expressed as a mass of ozone.