Dear Dr. Jöckel

We thank both reviewers for their dedication and insightful comments. Below are our responses to each reviewer. We have included line numbers with respect to the revised marked-up document to accommodate matching changes in the manuscript in response to these comments.

Response to reviewer 1

We thank the reviewer for their comments which have helped to improve and clarify several points in the manuscript. We are pleased that the reviewer found the manuscript to be well written and organised, and we address their comments below.

Significant Comments:

Lines 98-99: Are there separate variables for cloud ice and precipitation-size ice (snow and graupel)? If so, please be more specific here.

In the UKCA climate-chemistry model the different aspects of cloud ice, including snow, pristine ice and riming particles, are all considered together as part of one prognostic cloud ice variable. We have added the sentence "The cloud ice variable includes snow, pristine ice and riming particles." (Line 108). Microphysical processes such as melting snow and riming are represented but the prognostic variable provides the bulk response of all these components as cloud ice. The references included in the text in this paragraph provide further details of the microphysical scheme.

Line 171: Are there any biases in this tropospheric ozone product relative to sondes or other satellite observations?

The tropospheric ozone product used in this paper is that of Ziemke et al. (2011). Ziemke et al. (2011) evaluated the MLS/OMI product against ozonesondes and an alternative satellite product which combined SAGE and MLS with ozonesondes to estimate tropospheric ozone. There is no apparent systematic under- or over- estimation across the sonde sites. We have added the following sentences discussing this evaluation: "In an evaluation against ozone sondes with broad coverage across the globe, the MLS/OMI product generally simulated the annual cycle well (Ziemke et al., 2011). The annual mean tropospheric column ozone mixing ratio of the MLS/OMI product was found to have a root mean square error (RMSE) of 5.0 ppbv, and a correlation of 0.83, compared to all sonde measurements. The RMSE was lower and correlation higher (3.18ppbv and 0.94) for sonde locations within the latitude range 25°S to 50°N." (Lines 207-212)

Lines 219-220: The correlation is also not improved with ICEFLUX in the southern extratropics.

The reviewer is correct. This paragraph has been revised to: "Overall, the ICEFLUX approach reduces the errors in the annual cycles of lightning. This scheme improves the correlation between simulated and observed lightning compared to CTH scheme in the northern extratropics and southern tropics. It has a lower correlation in the northern tropics, where both approaches for simulating lightning have difficulties, and in the southern extratropics, where the magnitude of the bias is much reduced upon compared to the CTH approach." (Lines 263-268).

Lines 236-237: It would benefit the paper if these statistics were presented for the latitude bands.

We present an extended version of Table 1 with latitude bands included below. The top section of this table is the original version which remains in the manuscript. The other sections show statistics for other latitudes bands which we have decided not to add to the manuscript. The adjusted RMSE for each region was calculated using the mean bias over the full 60S-60N region. While we agree that it was worth examining these statistics, we feel they do not significantly add to the manuscript. An additional conclusion could be that the adjusted RMSE of the ZERO simulation is lower than that for the CTH and ICEFLUX simulations in northern midlatitudes. However, given the still comparatively large unadjusted RMSE in the region, this does not add sufficient value to merit inclusion of such a large amount of extra data.

Latitude band	Run	r	RMSE (DU)	Mean bias (DU)	adjusted RMSE (DU)
Full 60S-60N	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
60N-30N	CTH	0.65	4.0	–	3.6
	ICEFLUX	0.67	3.8	–	3.5
	ZERO	0.69	6.4	–	3.0
30N-Eq	CTH	0.95	3.3	–	3.4
	ICEFLUX	0.96	3.4	–	2.6
	ZERO	0.96	9.2	–	2.7
Eq-30S	CTH	0.90	4.0	–	2.4
	ICEFLUX	0.93	4.9	–	2.2
	ZERO	0.93	12.3	–	5.1
30S-60S	CTH	0.81	8.6	–	6.1
	ICEFLUX	0.81	8.9	–	5.8
	ZERO	0.81	13.5	–	6.3

We realise that we omitted an explicit description of the range of the MLS/OMI product in the manuscript (60S-60N), so we have added a comment to the data description section (Line 198) and the Table 2 caption.

Line 300: It is not clear how you are defining the tropopause. How are the 380K surface and the 2 PVU surface combined?

We have added a reference and modifed the text to read: "These budget terms are for the troposphere. Here, the tropopause is defined at each model time step using a combined isentropic-dynamical approach based on temperature lapse rate and potential vorticity (Hoerling1993)." (Line 391-395). The reference provides the details and motivation for the tropopause definition used in the UKCA model. It uses a thermal definition equatorward of 13 degrees, and a dynamical definition poleward of 28 degrees. In between there is a smooth transition using weightings of the two definitions. This definition of the tropopause overcomes issues with the individual definitions and produces a tropopause surface which is a continuous function of latitude.

Line 436: I am not sure what is the significance of this Ox production in the first 20 minutes. Isn't it primarily just the production of NO2 as it comes into equilibrium with NO in the atmosphere after flashes occur? Very little ozone production is going to be produced in 20 minutes.

Following NO emission, the NO is oxidised to bring NO into equilibrium with NO2. This happens principally though the reaction of NO with O3. The Ox production term diagnosed here does not include this reaction flux since the NO2 product is also an Ox species – no Ox is produced or lost from this reaction. Instead the Ox production term is dominated by the oxidation of NO by peroxy radicals. A small proportion of this Ox production in the first 20 minutes will be associated with equilibration, but the remainder reflects equilibrium NOx cycling and consequent O3 production. We have added the following text: "oxidation of NO to NO2 by peroxy radicals." to the Ox budget description in Section 4 (Line 388). The Ox budget diagram (Figure 6) demonstrates, terms through the use of a grey arrows, that reactions converting O3 to NO2 and vice versa are not counted in the Ox budget.

Table 2: What are the percentages? Are they the percentage changes from the CTH scheme? More meaningful might be to show the percentage changes of CTH and ICEFLUX from ZERO.

Good point. The percentage differences shown in Table 2 are with respect to the CTH scheme. We have now added a statement to this effect to the table caption. We choose to show changes with respect to CTH to demonstrate both the small effect of horizontal changes in distribution (the ICEFLUX column) and the large effect of emission changes (the ZERO column). We agree that percentages with respect to ZERO are useful but feel that percentages with respect to CTH make the above point most clearly.

Figure 5: There should be stratospheric influx of NO2 and other NOy species.

We thank the reviewer for highlighting this. The figure referred to is figure 6 in the new manuscript. The stratospheric influx is an inferred quantity which is derived to balance the other budget terms. It is not calculated for each species individually but is a value for the influx of total Ox, though this is dominated by the O3 contribution. We felt that the most accurate way to portray this was by including a stratospheric influx arrow pointing to the Ox label, instead of any individual species. We have added a statement describing the stratospheric influx term in the figure 6 caption.

Minor Comments:

Line 21: comparison of models

Changed to "between". Line 22

Line 183: define ACCMIP

Changed. Line 224

Line 267: .....instead the correlation values between the model and the sonde data (Figure 3) provide a more......

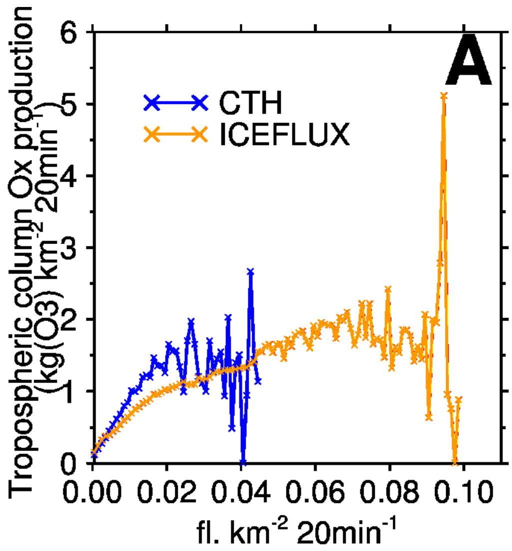
Changed. Line 356

Line 302: ....production and losses when lightning is added (Table 2).

Changed to “when lightning NOx emissions are removed”. Line 397

Line 446: The increase is linear up to approximately 0.006 fl km-2 min-1 and then becomes steeper up to o,02 fl km-2 min-1 at which....

We show below Figure 11A plotted with a linear x-axis which highlights the linear features which change at approximately 0.02 fl. km^{-2} 20min^{-1}. We have revised the text to read: “A linear increase in Ox production is apparent up to approximately 0.02 fl. km^{-2} 20min^{-1} at which point the two schemes produce 1 to 1.5 kg km^{-2} 20min^{-1} of Ox. Beyond this point, the Ox production simulated by the ICEFLUX approach increases still linearly but with a shallower gradient.” (Lines 568 - 570)



Response to reviewer 2

We thank reviewer 2 for their comments which clarified the manuscript and included useful suggestions on how to expand the analysis to be more relevant to future aircraft studies. We are very pleased that they suggest our new lightning parametrisation analysis would be an important addition to the literature. We address their comments below.

Specific Comments

Title. To convey what kind of parameterization, I would suggest modifying the title to say, “using a new lightning parameterization”.

The title has been changed to “…using a new global lightning parametrisation”

L. 126. How does the lightning-NOx scheme differentiate between cloud-to-ground (CG) and intracloud (IC) lightning? Does it need to make this difference if the production of NO per flash is the same for CG and IC flashes and the vertical distribution of NO sources is the same for CG and IC flashes?

Whilst the UKCA model does apply a method to determine the IC:CG ratio, it has no consequence in this study because, as the reviewer points out, we choose to have equal NO production for both types and because the vertical distribution is based on that of Ott et al. (2010) which is not dependent on the IC:CG ratio. A sentence has been added to clarify this:

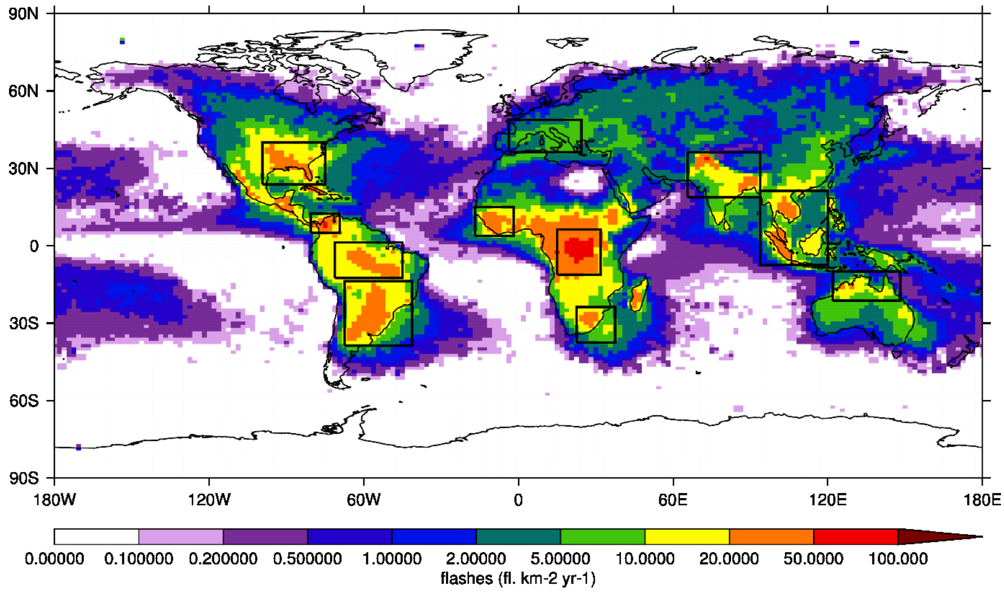
"As a consequence, the distinction between cloud-to-ground and cloud-to-cloud has no effect on the distribution or magnitude of lightning NOx emissions in this study." (Line 149).

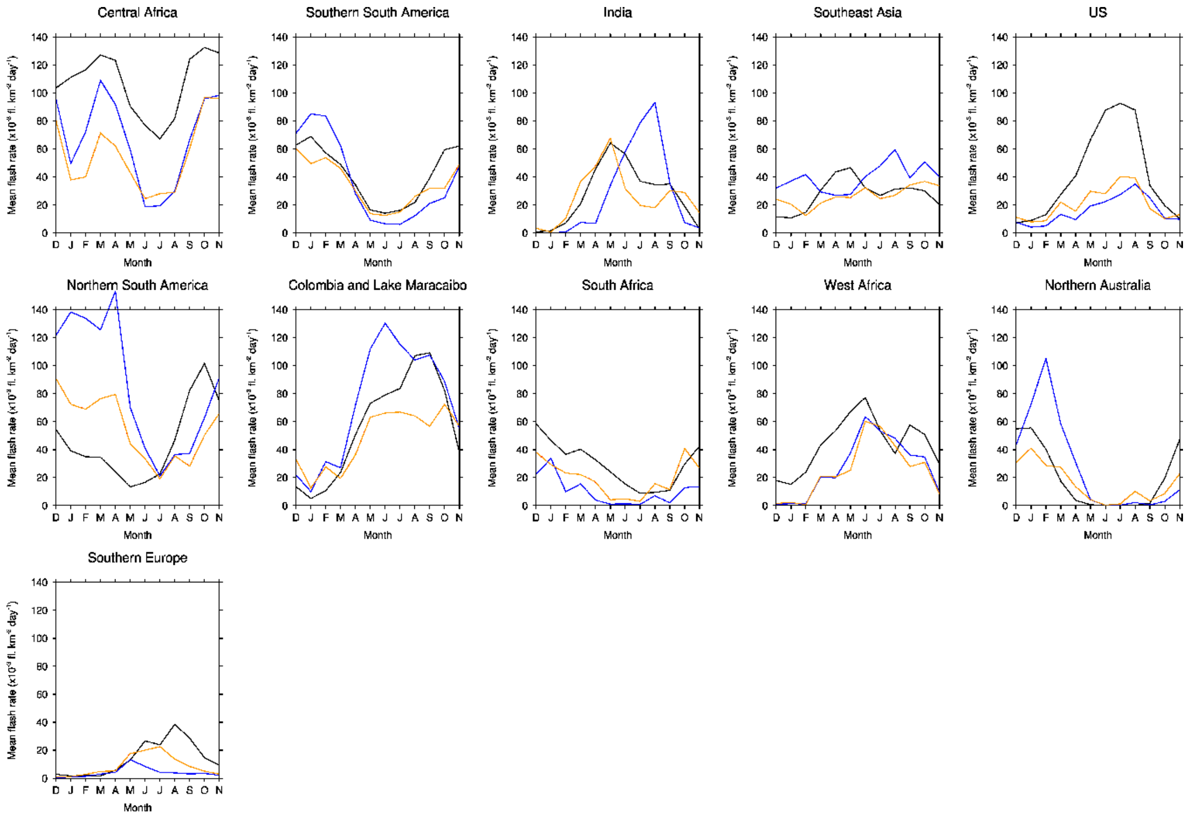
Section 3. Both lightning flash rate schemes depend on how well the model predicts cloud top height or ice mass flux. Has there been an attempt to evaluate these parameters (or a proxy) from the chemistry-climate model to a climatology of reanalysis data?

Several evaluations of the representation of clouds in the Unified Model have been performed in the literature and references for these have been added to Section 2.1, along with a paragraph describing the relevant results (Lines 114-127). No specific evaluation was carried out for this study since the upward cloud ice flux cannot be well constrained by observations on a global scale. We can infer that, given the similar distribution of lightning flash rates to the study of Finney et al. (2014), as well as similar total annual flashes, the upward cloud ice flux at 440hPa simulated in the Unified Model used here is similar to the ERA-Interim reanalysis data used by Finney et al. (2014).

Section 3.1 has a nice analysis of lightning flash rates in different latitude bands, and remarks upon differences in continental regions. I wonder if an additional figure showing how the model performs for different continents could be included and discussed. For example, showing the annual cycle of North American, South American, African, India and East Asia, and Australia (and maybe tropical oceanic region) lightning should give peaks at different times of the year. This type of figure would be a natural follow on to Figure 1 because the eye is drawn to each of these regions when viewing Figure 1.

We agree that focus on specific regions would be useful and we have therefore included six such regions in a new Figure 3 in the revised manuscript. We have also added text to discuss the new figure (Lines 269-307), as well as revised the previous paragraph over lines 249-362 to improve the flow of the text. Below, we show a map of land regions and annual cycles of regional-mean lightning flash rates for potential regions that were initially considered. We made a selection from these in order to offer an interesting but succinct discussion on regional performance.





L. 252. While both lightning-NOx schemes show a general underestimate of ozone in the middle and upper troposphere of the tropics, they are both within the variability of the observations (while no lightning-NOx is outside that variability). In fact, the northern tropics appears to have quite good agreement. If you want to point out the underestimation, restrict the comment to the southern tropics. Second, what is the variability in the model results?

We agree with the reviewer, although we also feel it worth noting in the text that the spring peak in the northern tropics is underestimated. The reviewer's point regarding the variability range is also correct. We have modifed the text to read "Both schemes still show an underestimate compared to observations all year round in the southern tropics and during spring in the northern tropics, but are within the variability of sonde measurements." (Line 339-341). There is no estimate of variability for the models as they are based on a single year run, however, by using a climatology of SSTs and emissions from year 2000, the simulation should broadly represent a tropospheric ozone climatology.

L. 284. I like the conclusion from the analysis that point to April and October as specific months to focus field campaigns. However, aircraft field campaigns can only cover a region (and not a latitude band). Can you recommend where field campaigns should focus? A similar analysis of continental regions would be helpful.

We feel that the additional analysis of the lightning annual cycle of particular regions (Figure 3) provides useful insights regarding this point. The biases in lightning in the southern tropics have been identified as originating in biases in northern South America, so this would be an appropriate region for studies. We have added a sentence to the ozone sonde discussion which refers back to this: "It may be of particular use for field campaigns studying the chemical impact of lightning to focus on these months and, as discussed in Section 3.1, South America could provide a useful region in which to develop understanding of lightning activity and therefore also its impacts on tropospheric ozone." (Line 375-378).

L. 297. I assume that the major Ox production is through oxidation of NO by peroxy radicals. This should be clarified to avoid confusion with NO + O3 producing NO2. It is curious that Table 2 discusses production and loss rates of Ox, but burdens of O3. I assume that is because O3 is the dominant Ox species (although it is of equal size to NO2 and O(1D) in Figure 5). It would be good to clarify in this paragraph why you discuss O3 burdens juxtaposed with the Ox production and loss rates discussion.

We thank the reviewer for this comment. We have added "by peroxy radicals" and a sentence to explain that only the ozone burden is considered because it makes up the majority of the Ox burden (Lines 388-390). The NO2 and O^1D species had been highlighted in the Ox budget diagram because they were involved in the production and loss key fluxes. However, to maintain a consistent approach to that used with the highlighting of reactions in Figure 6, the NO2 and O1D species have now been shaded grey along with the other Ox species apart from O3.

L. 303-304. Perhaps the characterization of the ZERO case could be revised. I think it should be described as the following. There is less production of Ox (or O3) without lightning-NOx emissions, resulting in a smaller O3 burden and therefore reduced Ox losses and shorter O3 lifetime. Can anything be said about linear or non-linear responses? For example, it seems that the lifetime decrease is less than the Ox loss rate decrease, and both are less than the decrease in Ox production.

This is a good point. The text has been modified to make the description of budget changes clearer (Line 398-403). We have commented that the lifetime changes by less and the reason for this, as we agree that this is an important point. However, to determine linearity, multiple experiments with different emissions would be needed.

Section 4. In addition to the comparison of the ZERO case with the two other cases, there should be a statement pointing out similarities between ICEFLUX and CTH, including the point at the beginning of Section 5.

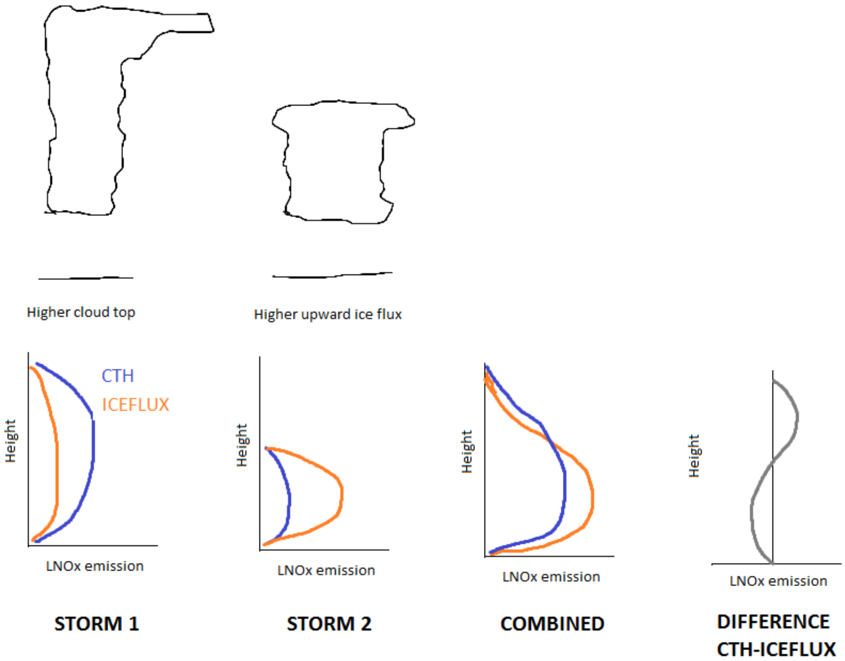
An additional sentence has been added: "The largest differences between the Ox budgets of the ICEFLUX and CTH approaches are in the ozone burden and lifetime but these are only 2%." (Line 413). At the beginning of section 5, an additional sentence has been added to point out that the two schemes produce similar values for the global Ox budget (Lines 438-440).

L. 341-344. I think it would be helpful to the reader to repeat how the NO lightning emissions are placed vertically for each scheme. It is also not clear to me how the horizontal distribution affects the vertical distribution. My interpretation is that ICEFLUX predicts lower lightning-NOx emissions in the tropics based on the storm parameters and more in the extratropics. While the magnitude of NO emissions is less in the ICEFLUX scheme for the tropics, those emissions are still distributed according to the Ott et al. (2010) curves to cloud top height (lines 126-128; and cloud top height should be the same in the two simulations). However, I think the authors are trying to say that the ICEFLUX scheme produces a lot of lightning-NO emissions in storms with lower cloud tops. There is also the point that because the CTH scheme has greater NO emissions in taller clouds, there is a substantial difference in where the NO emissions are found vertically. I think this could easily be supported by a plot of lightning-NO emissions versus cloud top height for different latitude bands.

The method to distribute LNOx vertically has been restated (Lines 453-455) and we have included a reference to the model parametrisation description in section 2.2. Regarding how the horizontal distribution of lightning affects the vertical distribution of LNOx emissions, we agree with the reviewer's interpretation and discuss the point below. The new section in the manuscript reads: "As described in section 2.2, the column LNOx is distributed up to the cloud-top, and this is how a coupling exists between the horizontal LNOx distribution simulated by the CTH approach and the height that LNOx emissions reach. This means that, by basing the horizontal lightning distribution on cloud-top height and then distributing emissions to cloud top, LNOx is most effectively distributed to higher altitudes." (Lines 453-457).

The vertical LNOx distribution is determined by the cloud top height at those (horizontal) locations where lightning has been diagnosed to occur. If cloud-top height is used to determine the horizontal distribution of lightning then the highest emissions will occur where the cloud tops are highest and therefore those emissions will be distributed to as high a level as possible for the given set of modelled cloud top heights.

An illustration of this is shown below for two storms (ultimately grid cells in the model): one with the highest cloud top, and one with the highest upward ice flux. Assuming the largest emission for the CTH approach is comparable to the largest emission for the ICEFLUX approach, the emissions are skewed to lower altitudes where using the ICEFLUX parametrisation. Figure 7E shows the gross Ox production zonal-altitudinal distribution and demonstrates that in the northern tropics there is a shift of Ox production to lower altitudes reflecting emissions at lower altitudes.



Regarding the reviewer's final suggestion, a plot of lightning NO emissions against cloud-top height would show the relationship used by the CTH approach scaled by the NO per flash parameter. For the ICEFLUX approach, it would show a different relationship because cloud top height is not used to determine the size of emission. This second plot would approximately show the relationship between upward ice flux and cloud-top height in the model. We feel that the inclusion of such plots would be somewhat of a digression and would not make the point clearer to the reader.

L. 369-375. Could this be clarified? It was already established that a reduction in Ox production decreased O3 mixing ratios and therefore Ox loss rates (Section 4). However, in these lines it says there is an increase in Ox production in the middle and lower troposphere but a reduction of O3 concentrations, when comparing ICEFLUX and CTH results. Is Ox partitioned differently, meaning there is more HNO3 that can be removed? What loss process dominates (O3 chemical loss or Ox wet deposition)?

The reasoning in the paragraph (Lines 485-492) is that there is an increase in net chemical production but that this is because the chemical loss is reduced in the region due to reduced ozone concentrations. Ozone concentrations have reduced because less ozone is produced in the upper troposphere to be transported within the tropics in the Hadley cell. The paragraph has been modified to focus on the upper tropospheric change impacting the whole tropical troposphere (Lines 485-492). The NOy wet deposition is not discussed but given there is no change in the global total deposition and NOy wet deposition makes up a small proportion of total deposition terms, it is reasonable to assume that it isn't a key driver in the ozone concentration distribution.

L. 405-410. I was surprised that the ICEFLUX lightning flash rate frequency distribution was not discussed. Also, although it is not the point of section 5, I wonder if it would be useful to include LIS/OTD frequency distribution in Figure 9.

The LIS/OTD product used here is a monthly climatology, hence cannot be compared to the 20 minute flash rates simulated by the model shown in in Figure 9. Whilst the individual LIS/OTD observations could be used to produce a frequency distribution, because the satellites do not measure all locations simultaneously, much care would be required to sample the model at the same locations and times. We therefore choose to describe the CTH distribution relative to the ICEFLUX distribution. An additional sentence has been added to give some context to the text on the ICEFLUX distribution: "The ICEFLUX approach produces a similar distribution to that produced by the same scheme applied in the study by Finney et al. (2014.) In that study the ICEFLUX frequency distribution had a fairly average distribution compared to four other lightning parametrisations with slightly more occurrences of low flash rates.“ (Lines 527-530).

L. 460. It is an interesting finding that Ox production efficiency is less for higher flash rates (at least initial Ox production). Could the authors speculate why this would happen? Or suggest analysis that could be done in order to explain why. I would imagine the HO2 and RO2 abundance might play a role. Are there connections between flash rate and location to VOC sources? For example, Barth et al. (2012) showed more O3 produced from storms occurring over VOC-rich regions (e.g. southeast U.S.).

This is definitely an interesting point. When extra NO is added there is a corresponding increase in Ox production. However, each additional NO molecule produces less Ox, as NOx cycling becomes less efficient for higher NOx levels. This is to be expected because other species involved in the NOx cycle such as HO2/VOCs do not increase in step with Ox to maintain the same production efficiency. Whilst location and background levels of ozone precursors play a role, the figures are based on global data and shows that this relationship holds for LNOx in general, though obviously high LNOx will have a tendency to occur in particular hotspots. The Barth et al. (2012) paper is a useful example of how VOCs interact with LNOx to affect ozone production. A reference to Barth et al. (2012) and three extra sentences have been added: "This suggests that as the NO increases, NOx cycling and therefore ozone production decreases in efficiency. This is likely a result of peroxy radical availability and VOC abundance limiting the rate of NOx cycling. Evidence for such control of VOC precursors on ozone production in US thunderstorms has been presented by Barth et al. (2012)." (Lines 585-588).

L. 465. How did the authors translate the Ox production efficiencies to Ox produced per mole of NO?

We have added the following at the beginning of the sentence "Using the NO production per flash of 250 mol(NO) fl.$^{-1}$ stated in Section 2.2,…" (Line 591)

L. 477. Here, the authors argue that more Ox is produced by the CTH scheme because NOx has a longer lifetime at higher altitudes. However, the analysis is for the initial Ox production ("at the time of emission")? How does the NOx lifetime affect the Ox production shown in Figure 10, which is "at the time of emission"?

This is a good point. There are several factors that can lead to increased ozone production efficiency from NOx at higher altitudes: the longer lifetime of NOx, the rate of NOx cycling, and efficiency of NOx cycling. The focus of the original text on the lifetime of NOx in this case was too specific as the lifetime will not play a substantive role in determining the ozone production efficiency over the initial 20 minute time step. We have amended the text to say "where ozone production efficiency is greater", since the reason for this is a combination of these factors (Line 606). The text on "NOx lifetime" mentioned earlier in the section and has also been broadened (Line 575).

<u>Technical Comments</u>

L. 9 Insert "NO" before emission.

Changed. Line 8

L. 17 Replace "-" with ";"

Changed. Line 17

L. 16-18 I suggest adding a caveat that more ozone production can subsequently occur from the high flash rate regions.

The term "initially" Ox production is added to the abstract (Lines 18). In the Section 6, we have added additional text "This study has analysed the Ox production occurring in the first 20 minutes, but further Ox production can occur over longer time periods." (Lines 616)

L. 21-22 Change to "for comparison between models and observations : : :".

Changed. Line 22

L. 27 NO2 lifetime may be shorter in the upper troposphere because its photolysis rate is greater. I think it would be better to rewrite the sentence to say NOx lifetime is longer in the upper troposphere (rather than the individual species).

Changed. Lines 28

L. 51 Could a reference be cited supporting that the upper troposphere is the region with most efficient ozone production?

We add the reference of Dahlmann et al. (2011) (Line 53) which addresses the ozone production efficiency of different sources including lightning and aircraft NOx and finds that these two sources have a greater ozone production efficiency because of their location.

L. 53 Please delete "simplified". I find cloud chemistry models to be rather complex.

This has now been removed. (Line 55)

L. 63-64 It would be better written as, ": : : of low flash rates, which are unrealistic compared to observed flash rates. This results in low NOx concentrations and greater ozone production efficiency : : :."

Changed. Lines 64-66

L. 86 Please add more information about the chemistry represented in the model. Is it the "standard troposphere" chemistry or does it have the added isoprene chemistry, both described in O'Conner et al. (2014)? I suggest including number of species, stating it describes methane, ethane, and propane (and maybe isoprene) hydrocarbon chemistry.

More information has been added which addresses the comment. Isoprene chemistry is included and appropriate references are given. Lines 90-94

L. 147-151 Could this be rewritten? It appears that only lightning flash rates are scaled to obtain a global values of 46 fl/s, because the NO production per energy is the same for both cases. Is the energy per flash changed? I suggest rewriting to first address the scaling for the flash rates, including the comment that the scaling factor is very similar to Finney et al (2014). Then discuss the scaling applied to get 5 Tg N per year globally.

A scaling factor is calculated for each parametrisation to achieve the same global annual flash rate. Each flash has equal energy. Then the NO production per Joule is chosen in order to produce 5 TgN per year given the total number of flashes (which is the same for each parametrisation). The ordering and wording of the sentences has been altered to make this clearer: "However, for this study we choose to have the same flash rate and global annual NOx emissions for both schemes. A scaling factor was used for each parametrisation that results in the satellite estimated flash rate of 46 fl./s, as given by Cecil et al. (2012). … Given that each parametrisation produces the same number of flashes each year and each flash has the same energy, a single value for NO production can be used. As above, a value of 12.6 X 10^{16} NO molecules J^{-1} was used for both schemes which results in a total annual emission of 5 TgN yr^{-1}." (Lines 171-182)

L. 164 I think it would be good to include in the text what is said in the caption of Figure 1 regarding the satellite data are regridded to the model grid.

Changed. Lines 194-195

L. 174 The model ozone column is regridded. I assume that it is placed on the same grid as the satellite climatology (which is what in degrees latitude and longitude?). Could the sentence be clarified? ": : : is regridded to the satellite grid of x by y degrees and then compared on this grid. The model ozone column was not sampled the satellite track. (perhaps this last sentence is placed before the previous sentence).

The model is regridded to the MLS/OMI grid of 5x5 degrees. The sentence has been rephrased as "In Section 3.2, the simulated annual mean ozone column is regridded to the MLS/OMI grid of 5° by 5° and compared directly to the satellite climatology without sampling along the satellite track." (Lines 204-206)

L. 178 Hard to believe Thompson (2003) included data until 2011! It looks like 2011 should be 2000.

The Thompson [et al. *now corrected*] (2003) paper describes the sites but this data set has since been extended. The sentence has been revised to say this. Lines 214-216

L. 179 Perhaps add values of latitudes for the 4 regions.

Added. Lines 218

L. 187 What does ": : : extension of the evaluation over a smaller region : : :" mean? I assume that this paper evaluates lightning over a larger region than what was used by Finney et al. (2014).

Yes, the region used in Finney (2014) was smaller. This sentence has been revised for clarity. Line 228

L. 275 Insert "NOx" before emissions.

Changed. Line 364

L. 303 Add "in the ZERO simulation" in stating which case has reduced deposition.

Referred to ZERO at the beginning of the sentence. Line 398

L. 305 is not clear. Is not the ZERO simulation corresponding to a reduction of N emissions by definition? That is, it is how the simulation is configured. What is the point of "less than the range of estimates for lightning emissions"?

The difference in LNOx (5 TgN/yr) is similar in magnitude to the uncertainty in the total lightning NOx source (~6 TgN/yr based on 2-8 TgN/yr). The sentences have been modified to try and make the point clearer: "There is uncertainty in the global lightning NOx source of 2-8 TgN emissions (Schumann and Huntrieser 2007), and there will be an associated uncertainty in the Ox budgets. Using no lightning (ZERO) corresponds to a reduction of 5 TgN emissions over the year - less than the range of uncertainty in LNOx. " Lines 404-407

L. 315 Use "whole" instead of "total" to be consistent with table.

Changed for all instances in the paragraph. Lines 415,420,422 and 426

L. 315-319 Why not just say "less than by 13 Tg" instead of "difference of -13 Tg"? I think your meaning may become clearer. Likewise, for the other differences stated in this paragraph.

Changed. Lines 423-428

L. 309-324. Consider revising the construction of this paragraph, which is making the point that location of the emissions (tropics versus extratropics) matters because production of O3 in the tropical upper troposphere will result in more O3 transported into the stratosphere. Previous studies found this result, and your results do as well. Implement basic paragraph construction: Topic of paragraph (or point being made), support of this topic, concluding sentence.

Agreed. The beginning of the paragraph has been altered. Line 415

L. 326-333 Remind the reader that although the ICEFLUX and CTH simulations were designed to have the same magnitude of lightning flashes and lightning-NOx production, the location of the lightning and lightning-NOx differs between simulations, citing Figure 1 or other supporting information.

The following sentence has been added, "In the previous section, it was demonstrated that the global tropospheric Ox budget is affected principally by the magnitude of emissions and not the location of emissions. This was achieved by using the same total emissions but different distributions of lightning in the CTH and ICEFLUX approaches (Figure 1), which simulate little difference in the global Ox budget terms." Lines 436-440

L. 355 add "by peroxy radicals".

Changed. Line 469

L. 358-359. Change to "Ox precursors are transported downwind of convection before they form ozone".

Amended the sentence to: "Furthermore, ozone precursors are transported downwind of convection before they form ozone.". Line 473

L. 361-363. The last sentence of the paragraph should be the first sentence of the next paragraph.

Changed. Line 477

L. 473. When the authors say, "at the time of emission", do they mean within the model time step? In other words, 15% of the Ox production associated with lightning occurs within 20 minutes of the lightning flash (or NO emission)?

Yes, in the model time step including the emission. This has been clarified in this instance and at the first use of *initial*. Lines 557 and 601

Table 1. Add units for RMSE and mean bias.

Added.

Table 2. Add information about values in parentheses.

Added to Table caption.

Manuscript prepared for Atmos. Chem. Phys.
with version 2015/04/24 7.83 Copernicus papers of the LaTeX class copernicus.cls.
Date: 23 May 2016

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

D. L. Finney[1], R. M. Doherty[1], O. Wild[2], and N. L. Abraham[3,4]

[1]School of GeoSciences, The University of Edinburgh, Edinburgh, UK
[2]Lancaster Environment Centre, Lancaster University, Lancaster, UK
[3]Department of Chemistry, University of Cambridge, Cambridge, UK
[4]National Centre for Atmospheric Science, University of Cambridge, Cambridge, UK

Correspondence to: D. L. Finney (d.finney@ed.ac.uk)

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 NO 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 initially 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~~ between models and observations, and for simulation of future changes in tropospheric ozone.

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. The oxidation of NO forms NO_2 and the sum of these is referred to as NO_x. In the middle and upper troposphere ~~and (together~~ NO_x ~~) have longer lifetimes~~ has a longer lifetime 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~~ changes in radiative flux resulting from changes in 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 (Dahlmann et al., 2011). 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~~ so far has focussed on 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 concentrations~~ which are unrealistic compared to ~~observed flash rates~~the observed flash rate distribution. This results in ~~a~~ lower NO_x concentrations and 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 O_x budgets. Section 5 then considers the differences in zonal and altitudinal distributions of chemical 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~~ and stratospheric 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) ~~.~~ and the stratospheric scheme by Morgenstern et al. (2009). This combined *CheST* chemistry scheme has been used by Banerjee et al. (2014) in an earlier configuration of the Unified Model. There are 75 species with 285 reactions considering the oxidation of methane, ethane, propane, and isoprene. Isoprene oxidation is included using the Mainz Isoprene Mechanism of Pöschl et al. (2000). Squire et al. (2015) gives a more detailed discussion of the isoprene scheme used here.

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 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. The cloud ice variable includes snow, pristine ice and riming particles. 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).

Evaluation of the distribution of cloud depths and heights simulated by the Unified Model has been performed in the literature. For example, Klein et al. (2013) conclude that across a range of models, the most recent models improve the representation of clouds. They find that HadGEM2-A, a predecessor of the model used in this study, simulates cloud fractions of high and deep clouds in good agreement with the International Satellite Cloud Climatology Project (ISCCP) climatology. In addition, Hardiman et al. (2015) studied a version of the Unified Model which used the same cloud and convective parametrisations as used here. They found that over the tropical Pacific warm pool that high cloud of 10-16 km occurred too often compared to measurements by the CALIPSO satellite. This will bias a lightning parametrisation based on cloud-top height, over this region. Cloud ice content and updraught mass flux, which are used in the ice flux based lightning parametrisation presented in this study, are are not well constrained by observations and represent an uncertainty in the simulated lightning. However, these variables are fundamental components of the Non-Inductive Charging Mechanism and therefore it is appropriate to consider a parametrisation which includes such aspects.

Simulations for this study were set up as a time-slice experiment using sea surface temperature and sea ice climatologies based on ~~1995-2005~~ 1995-2004 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 (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 J^{-1} These correspond to 250 mol(NO) 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. As a consequence, the distinction between cloud-to-ground and cloud-to-cloud has no effect on the distribution or magnitude of lightning $\mathrm{NO_x}$ emissions in this study. 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. $\mathrm{m}^{-2}\,\mathrm{s}^{-1}$) 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 per J were scaled to result~~ A scaling factor was used for each parametrisation that results in the satellite estimated flash rate ~~by Cecil et al. (2014)~~ of 46~~fl. and then a total emission of 5 TgN~~ $\mathrm{fl.\,s^{-1}}$, as given by Cecil et al. (2014). 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} molecules 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. Given that each parametrisation produces the same number of flashes each year and each flash has the same energy, a single value for NO production can be used. As above, a value of 12.6×10^{16} $\mathrm{NO molecules\,J^{-1}}$ was used for both schemes which results in a total annual emission of 5 $\mathrm{TgN\,yr^{-1}}$.

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 $\pm 75°$ latitude from 1995-2000 while LIS observed between $\pm 38°$ 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°~~ 0.5° horizontal resolution made up of all the measurements of OTD and LIS between ~~1995-2010. A~~ May 1995 - December 2011. Cecil et al. (2014) provides a detailed description of the product ~~is provided by Cecil et al. (2014)~~using data for 1995-2010, which had been extended to 2011 when data was obtained for this study. The LIS/OTD climatology product was regridded to the resolution of the model (1.875° longitude by 1.25° latitude) for comparison.

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 between $\pm 60°$ latitude, 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 to the MLS/OMI grid of 5° by 5° and compared directly to the satellite climatology without sampling along the satellite track.

In an evaluation against ozone sondes with broad coverage across the globe, the MLS/OMI product generally simulated the annual cycle well (Ziemke et al., 2011). The annual mean tropospheric column ozone mixing ratio of the MLS/OMI product was found to have a root mean square error (RMSE) of 5.0 ppbv, and a correlation of 0.83, compared to all sonde measurements. The RMSE was lower and correlation higher (3.18 ppbv and 0.94) for sonde locations within the latitude range 25°S to 50°N.

Secondly, ozone sonde observations averaged into 4 latitude bands were used. The ~~ozonesonde~~ ozone sonde measurements are from ~~datasets~~ the dataset described by Logan (1999) (representative of 1980–1993) and ~~Thompson et al. (2003) (~~from sites described by Thompson et al. (2003) for which the data has since been extended to be representative of 1997–2011~~), and~~. The data consists of 48 stations, with 5, 15, 10 and 18 stations in the ~~SH extratropics , SH tropics , NH tropics and NH extratropics~~ southern extratropics (90S-30S), southern tropics (30S-Equator), northern tropics (Equator-30N) and northern extratropics (30N-90N) 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 observational ozone datasets are the same as used in the ~~ACCMIP multi-model comparison~~ Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) 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 made by Finney et al. (2014) which was 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.

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~~

Figure 2B shows that CTH has very large ~~absolute~~ root mean square errors during December to April ~~, with~~ in the southern tropics. A more detailed analysis (not shown) ~~suggesting this is~~ suggests that these errors are due to overestimation ~~in the South American region~~over South America. In the northern tropics the temporal correlation with LIS/OTD suggests CTH performs slightly better than the ICEFLUX approach, although Figure 2C 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, the more detailed analysis found that both schemes failed to match the observed magnitude of the August peak of ~~the American region and the~~ Central America and the Southern US, nor the duration of the lightning peak ~~in the African region~~ over Northern Africa 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,~~. This scheme improves the correlation ~~except~~ between simulated and observed lightning compared to CTH scheme in the northern ~~tropics~~ extratropics and southern tropics. It has a lower correlation in the northern tropics, where both approaches for simulating lightning have difficulties, and in the southern extratropics, where the magnitude of the bias is much reduced upon compared to the CTH approach.

To further understand how the schemes perform on a regional scale, the annual cycles of the simulated and observed lightning, for a selection of key regions, are shown in Figure 3A. A box showing each region is plotted on Figure 1. The regions of Figure 3 include many of the peak areas of lightning shown in Figure 1A or, in the case of Europe, are an area in which a higher density of measurement studies are undertaken including using ground-based lightning detectors.

Figure 3A shows the Central African peak lightning region where both parametrisations successfully simulate the observed peak months of lightning in the LIS/OTD data. For the most part, both parametrisations produce similar flash rates. However the simulated flash rates generally underestimate lightning compared to the observations. Interestingly, the ICEFLUX approach has a greater underestimation of the observed Spring lightning peak compared to the CTH approach. This suggests that the input meteorology for the ICEFLUX scheme over the Central African region is less well simulated during this season, or that the ICEFLUX scheme does not capture some necessary aspect of thunderstorm activity during the season. Over the Indian region (Figure 3B), the two schemes substantially differ in their flash estimates. The ICEFLUX scheme achieves a much more realistic annual cycle than the CTH scheme. This suggests that aspects of charging during the Indian monsoon seasons may not be captured by the cloud-top height approach. Two regions in South America are shown in Figure 3 C and D. Both schemes capture the southern South American annual cycle of lightning flash rates well but both perform poorly in the northern region (the ICEFLUX approach results in a much lower bias). Biomass burning aerosols could be a key control on lightning activity in the region, as was shown by (Altaratz et al., 2010). The flash rate peak in the southern USA region is greatly underestimated by both schemes 3. The lack of difference between the two schemes suggests that it may not be the best study region for distinguishing which is a more successful parametrisation. Finally, over the southern European region, both schemes show an underestimation of flash rates compared to LIS/OTD, although the bias is less in the case of the ICEFLUX approach. The August peak in this region is not captured by either approach, which may relate to lightning activity over the Mediterranean Sea, given that both schemes also underestimate the annual flash rate over the Mediterranean Sea as shown in Figure 1.

The analysis of the annual cycle of flash rates in some key regions has shown that the ICEFLUX scheme is similar to or improves upon the simulated annual cycle by the CTH scheme when compared to the LIS/OTD satellite climatology. The exception is for the Central African peak in Spring. Any future studies of the Central African region could explore this difference further. Neither parametrisation captures the magnitude of flash rates over the southern USA or southern European regions. Given the high density of measurements in these regions it should be possible to study why this underestimation occurs in future studies. Finally, we suggest that one of the greatest sources of bias in the flash rate estimates by the CTH scheme are over northern South America. The ICEFLUX scheme reduces this bias but still does not capture the annual cycle. In southern South America both parametrisations reproduce the observed annual cycle of lightning. Therefore, we suggest that field campaigns comparing the southern and northern regions of South America would be particularly useful in improving the understanding of lightning processes and finding reasons for large-scale biases in models.

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)~~ 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 4 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 4). 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 5 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~~ . Both schemes still show an underestimate compared to observations ~~(Figure 5)~~all year round in the southern tropics and during spring in the northern tropics, but are within the variability of sonde measurements. 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 between the model and sonde data (Figure 4) provide a more useful evaluation of parametrisation success.

In Figure 5 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 5D) and in October in the southern tropics (most notably at 250 hPa)(Figure 5A). 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 5B does not appear in any of the model simulations. Potentially, the modelled advection is not transporting the lightning $\mathrm{NO_x}$ emissions or ozone produced to high enough altitudes. An anomalous southern tropical peak in March in Figures 5A 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 and, as discussed in Section 3.1, South America could provide a useful region in which to develop understanding of lightning activity and therefore also its impacts on tropospheric chemistry.

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 6. 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 ~~.~~ by peroxy radicals. The ozone burden is considered along with the budget terms as it is the key species of interest and it makes up the majority of the O_x burden.

The global annual O_x budgets for CTH, ICEFLUX and ZERO are given in Table 2. ~~The~~ These budget terms are for the troposphere~~which is diagnosed each~~. Here, the tropopause is defined at each model time step using ~~the modelled meteorology to determine a tropopause defined as a combination of the pressures at 380 K and at 2 PVU~~a combined isentropic-dynamical approach based on temperature lapse rate and potential vorticity (Hoerling et al., 1993). Clearly, the ZERO ~~run~~ simulation 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 when lightning NO_x emissions are removed (Table 2). ~~Also because of reduced ozone concentrations~~The O_x budget for the ZERO simulation shows that through reduced ozone production, there is reduced ~~deposition~~ozone burden and therefore chemical losses and deposition fluxes are reduced. The lifetime of ozone is ~~less affected compared to other terms because the ozone burden has reduced as well as the loss terms.~~ given by the burden divided by the losses. Since the burden decreases more than the losses, the ozone lifetime reduces overall, although to a lesser extent than the burden and loss terms individually.

There is uncertainty in the global lightning NO_x source of 2-8 TgN emissions (Schumann and Huntrieser, 2007), and there will be an associated uncertainty in the O_x budgets. 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)~~uncertainty in LNO_x. Therefore large changes in O_x budget terms can be expected within the uncertainty range of the global lightning NO_x emission total.

~~It~~ In contrast, it would seem that for constant emissions of 5 TgN and a reasonable change in the flash rate distribution ~~using ICEFLUX~~by using the ICEFLUX approach instead the CTH approach, there are only small ~~changes~~ differences in the global O_x budget terms~~but this does not consider changes in composition of the lower stratosphere~~. The largest differences between the O_x budgets of the ICEFLUX and CTH approaches are in the ozone burden and lifetime but these are only 2 %.

The O_x budget discussed so far represents the troposphere, but if the whole atmospheric ozone burden is considered (Table 2) then it is apparent that there is an also a reduction in ozone in stratosphere which must be due to changes in the troposphere-stratosphere exchange of ozone. Previous studies have also 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~~ differences in whole atmospheric ozone burden against ~~changes~~ differences in tropospheric ozone burden. The ~~difference in simulated total~~ whole atmospheric ozone burden ~~between ICEFLUX and CTH is -13 Tg~~ simulated with ICEFLUX approach is 13 Tg less than that simulated by the CTH approach. Given the ~~-6 Tg difference in the troposphere~~tropospheric ozone burden simulated by the ICEFLUX approach is only 6 Tg less that that of the CTH approach, 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~~ whole atmospheric ozone burden simulated in the ZERO run was ~~-91 Tg~~ 91 Tg less than that of the CTH approach. The tropospheric ozone burden ~~difference was -62 Tg~~ was 62 Tg less 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 sectionshowed~~ In the previous section, it was demonstrated 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 was achieved by using the same total emissions but different distributions of lightning in the CTH and ICEFLUX approaches (Figure 1), which simulate little difference in the global O_x budget terms. 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 7A-C for the CTH scheme as well as changes as a result of using ICEFLUX instead of CTH in Figure 7D-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 7D and E). Figure 7E 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 7D). 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 8), 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~~ As described in section 2.2, the column LNO_x is distributed up to the cloud-top, and this is how a coupling exists between the horizontal LNO_x distribution simulated by the CTH approach and the height that LNO_x emissions reach. This means that, by basing the horizontal lightning distribution on cloud-top height and then distributing emissions to ~~cloud-top, emissions are~~ cloud top, LNO_x is 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 7E which shows gross production in the northern tropics. Whilst both lightning schemes have similar total lightning at these latitudes (shown in Figure 8), 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 by peroxy radicals, 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 species are transported before then forming~~ ozone precursors are transported downwind of convection before they form ozone. The difference in O_x production (Figure 7) between the two lightning schemes influences not only ozone locally but also downwind where ozone is transported to.

Figure 9 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 9) 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 9) despite increased net O_x production (Figure 7D). This is because there is less ozone produced in the upper troposphere, and therefore there are lower ozone concentrations in the air transported within the vertical circulation in the tropics. In the southern tropics~~this is because the increase in~~, the net O_x production increase 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~~ as a result of ~~less ozone available to be transported from the upper troposphere within the Hadley cell or other vertical subsidence~~lower ozone concentrations in the region. 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 9). 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 10 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 10). 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). The ICEFLUX approach produces a similar distribution to that produced by the same scheme applied in the study by Finney et al. (2014). In that study the ICEFLUX frequency distribution had a fairly average distribution compared to four other lightning parametrisations with slightly more occurrences of low flash rates.

In Figure 10, 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 NO_x (Allen and Pickering, 2002).

- Total ozone production increased approximately linearly up to 300 pptv of lightning 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 HO_x concentration (DeCaria et al., 2005).

In the following analysis we consider 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 11 presents two metrics of the gross column chemical O_x production resulting from continental lightning in each of the frequency bins of Figure 10. The metrics are: A) the mean column O_x production, and B) the mean O_x production per flash. Each flash corresponds to 250~~mol()~~ mol(NO) emission so the O_x production per mole of emission can easily be inferred from the O_x production per flash. 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* O_x production ~~resulting from emission~~occurring in the 20 minute time step in which emissions are produced. This initial O_x production has been calculated to be approximately 15% of total O_x production associated with lightning for both parametrisations. The calculation was made as the difference between the total O_x production resulting from lightning in the sampled grid cells and the total 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 O_x production in Figure 11A shows, as expected, that increasing flash rate (i.e. more NO emissions in a cell) results in increased column O_x production. The higher extreme flash rates of ICEFLUX compared to CTH result in greater column O_x productions as a result of individual occurrences. ~~The increase is linear~~ A linear increase in O_x production is apparent up to approximately 0.02~~fl. km^{-2}20min^{-1}~~ $\mathrm{fl.\,km^{-2}\,20min^{-1}}$ at which point the two schemes produce 1 to 1.5~~kg~~ $\mathrm{kg\,km^{-2}\,20min^{-1}}$ 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 11A). 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~~greater ozone production efficiency, 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 11B shows the mean column O_x production per flash for each flash rate bin. It is derived by dividing the data in Figure 11A by the mid-point flash rate of each bin. Whilst Figure 11A 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.~~ $\mathrm{fl.\,km^{-2}\,20\,min^{-1}}$ produce ~ 10 times more O_x per flash than flash rates of 0.05~~fl. .~~ $\mathrm{fl.\,km^{-2}\,20min^{-1}}$. This suggests that as the NO increases, NO_x cycling and therefore ozone production decreases in efficiency. This is likely a result of peroxy radical availability and VOC abundance limiting the rate of NO_x cycling. Evidence for such control of VOC precursors on ozone production in US thunderstorms has been presented by Barth et al. (2012).

ICEFLUX displays the greatest contrast in efficiency between high and low flash rates of the two parametrisations (Figure 11B). 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~~ Using the NO production per flash of 250 $\mathrm{mol(NO)\,fl.^{-1}}$ stated in Section 2.2, the range of initial O_x production per mol of emission is 25~~mol() mol^{-1}()~~ $\mathrm{mol(O_x)\,mol^{-1}(NO)}$ at low flash rates for ICEFLUX to less than 2~~mol()~~ $\mathrm{mol(O_x)\,mol^{-1}(NO)}$ for the highest flash rates in the ICEFLUX scheme (Figure 11B).

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.~~ 0.02 $\mathrm{fl.\,km^{-2}\,20min^{-1}}$, 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~~ in the first 20 minutes after 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 ~~lifetime is longer~~ozone production efficiency is greater), 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. This study has analysed the O_x production occurring in the first 20 minutes, but further O_x production can occur over longer time periods. 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.

Acknowledgements. This work has been supported by a Natural Environment Research Council grant NE/K500835/1. We thank the TRMM satellite team for access to the Lightning Imaging Sensor products. Thanks to Paul Young for providing and assisting with use of the ozone column and sonde observations~~.~~, and Jonathan Wilkinson for guidance regarding implementation of the lightning parametrisation based on ice flux in the Met Office Unified Model. Finally, we thank the two anonymous reviewers who greatly helped to improve the manuscript.

References

Allen, D. J. and Pickering, K. E.: Evaluation of lightning flash rate parameterizations for use in a global chemical transport model, ~~Journal of Geophysical Research~~J. Geophys. Res., 107, 4711, doi:10.1029/2002JD002066, 2002.

Altaratz, O., Koren, I., Yair, Y., and Price, C.: Lightning response to smoke from Amazonian fires, Geophys. Res. Lett., 37, 1–6, doi:10.1029/2010GL042679, 2010.

Apel, E. C., Hornbrook, R. S., Hills, A. J., Blake, N. J., Barth, M. C., Weinheimer, A., Cantrell, C., Rutledge, S. A., Basarab, B., Crawford, J., Diskin, G., Homeyer, C. R., Campos, T., Flocke, F., Fried, A., Blake, D. R., Brune, W., Pollack, I., Peischl, J., Ryerson, T., Wennberg, P. O., Crounse, J. D., Wisthaler, A., Mikoviny, T., Huey, G., Heikes, B., Sullivan, D. O., and Riemer, D. D.: Upper tropospheric ozone production from lightning NOx-impacted convection: Smoke ingestion case study from the DC3 campaign, J. Geophys. Res. Atmos., 120, 1–19, doi:10.1002/2014JD022121, 2015.

Banerjee, A., Archibald, A. T. A. M., Telford, P., Abraham, N. L., Yang, X., Braesicke, P., and Pyle, J.: Lightning NOx, a key chemistry-climate interaction: impacts of future climate change and consequences for tropospheric oxidising capacity, Atmos. Chem. Phys., 14, 9871–9881, doi:10.5194/acp-14-9871-2014, 2014.

Barth, M. C., Lee, J., Hodzic, A., Pfister, G., Skamarock, W. C., Worden, J., Wong, J., and Noone, D.: Thunderstorms and upper troposphere chemistry during the early stages of the 2006 North American Monsoon, Atmos. Chem. Phys, 12, 11 003–11 026, doi:10.5194/acp-12-11003-2012, 2012.

Bushell, A. C., Wilson, D. R., and Gregory, D.: A description of cloud production by non-uniformly distributed processes, Q. J. Roy. Meteor. Soc., 129, 1435–1455, doi:10.1256/qj.01.110, 2003.

Cecil, D. J., Buechler, D. E., and Blakeslee, R. J.: Gridded lightning climatology from TRMM-LIS and OTD: Dataset description, Atmos. Res., 135-136, 404–414, doi:10.1016/j.atmosres.2012.06.028, 2014.

Cooray, V., Rahman, M., and Rakov, V.: On the NOx production by laboratory electrical discharges and lightning, J. Atmos. Sol.-Terr. Phys., 71, 1877–1889, doi:10.1016/j.jastp.2009.07.009, 2009.

Dahlmann, K., Grewe, V., Ponater, M., and Matthes, S.: Quantifying the contributions of individual NOx sources to the trend in ozone radiative forcing, Atmos. Environ., 45, 2860–2868, doi:10.1016/j.atmosenv.2011.02.071, 2011.

DeCaria, A. J., Pickering, K. E., Stenchikov, G. L., and Ott, L. E.: Lightning-generated NOx and its impact on tropospheric ozone production: A three-dimensional modeling study of a Stratosphere-Troposphere Experiment: Radiation, Aerosols and Ozone (STERAO-A) thunderstorm, J. Geophys. Res., 110, D14 303, doi:10.1029/2004JD005556, 2005.

Finney, D. L., Doherty, R. M., Wild, O., Huntrieser, H., Pumphrey, H. C., and Blyth, A. M.: Using cloud ice flux to parametrise large-scale lightning, Atmos. Chem. Phys., 14, 12 665–12 682, doi:10.5194/acp-14-12665-2014, 2014.

Grewe, V., Brunner, D., Dameris, M., Grenfell, J., Hein, R., Shindell, D., and Staehelin, J.: Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes, Atmos. Environ., 35, 3421–3433, doi:10.1016/S1352-2310(01)00134-0, 2001.

Grewe, V., Reithmeier, C., and Shindell, D. T.: Dynamic-chemical coupling of the upper troposphere and lower stratosphere region., Chemosphere, 47, 851–61, 2002.

Hardiman, S. C., Boutle, I. A., Bushell, A. C., Butchart, N., Cullen, M. J. P., Field, P. R., Furtado, K., Manners, J. C., Milton, S. F., Morcrette, C., O'Connor, F. M., Shipway, B. J., Smith, C., Walters, D. N., Willett, M. R., Williams, K. D., Wood, N., Lukeabraham, N., Keeble, J., Maycock, A. C., Thuburn, J., and Woodhouse, M. T.: Processes controlling tropical tropopause temperature and stratospheric water vapor in climate models, J. Climate, 28, 6516–6535, doi:10.1175/JCLI-D-15-0075.1, 2015.

Hoerling, M. P., Schaack, T. K., and Lenzen, A. J.: A global analysis of stratospheric?tropospheric exchange during northern winter, doi:10.1175/1520-0493(1993)121<0162:AGAOSE>2.0.CO;2, 1993.

Klein, S. A., Zhang, Y., Zelinka, M. D., Pincus, R., Boyle, J., and Gleckler, P. J.: Are climate model simulations of clouds improving? An evaluation using the ISCCP simulator, J. Geophys. Res. Atmos., 118, 1329–1342, doi:10.1002/jgrd.50141, 2013.

Labrador, L. J., von Kuhlmann, R., and Lawrence, M. G.: The effects of lightning-produced NOx and its vertical distribution on atmospheric chemistry : sensitivity simulations with MATCH-MPIC, Atmos. Chem. Phys., 5, 1815–1834, doi:10.5194/acp-5-1815-2005, 2005.

Lacis, A. A., Wuebbles, D. J., and Logan, J. A.: Radiative forcing of climate by changes in the vertical distribution of ozone, ~~Jo~~J. Geophys. Res., 95, 9971–9981, doi:10.1029/JD095iD07p09971, 1990.

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): Overview and description of models, simulations and climate diagnostics, ~~Geoscientific Model Development~~Geosci. Model Dev., 6, 179–206, doi:10.5194/gmd-6-179-2013, 2013.

Liaskos, C. E., Allen, D. J., and Pickering, K. E.: Sensitivity of Tropical Tropospheric Composition to Lightning NO x Production as Determined by the NASA GEOS-Replay Model, J. Geophys. Res. Atmos., 120, 8512–8534, doi:10.1002/2014JD022987, 2015.

Logan, J. a.: An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models and development of a gridded climatology for tropospheric ozone, J. Geophys. Res., 104, 16 115–16 149, doi:10.1029/1998JD100096,1999.

Morcrette, C. J.: Improvements to a prognostic cloud scheme through changes to its cloud erosion parametrization, ~~Atmospheric~~ Atmos. Sci. Lett., 13, 95–102, doi:10.1002/asl.374, 2012.

Morgenstern, O., Braesicke, P., O'Connor, F. M., Bushell, A. C., Johnson, C. E., Osprey, S. M., and Pyle, J. A.: Evaluation of the new UKCA climate-composition model ? Part 1: The stratosphere, Geosci. Model Dev., 2, 43–57, doi:10.5194/gmd-2-43-2009, 2009.

Murray, L. T., Jacob, D. J., Logan, J. A., Hudman, R. C., and Koshak, W. J.: Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data, J. Geophys. Res., 117, D20 307, doi:10.1029/2012JD017934, 2012.

Murray, L. T., Logan, J. A., and Jacob, D. J.: Interannual variability in tropical tropospheric ozone and OH : the role of lightning, J. Geophys. Res., 118, 11 468–11 480, doi:10.1002/jgrd.50857, 2013.

O'Connor, F. M., Johnson, C. E., Morgenstern, O., Abraham, N. L., Braesicke, P., Dalvi, M., Folberth, G. a., Sanderson, M. G., Telford, P. J., Voulgarakis, A., Young, P. J., Zeng, G., Collins, W. J., and Pyle, J. a.:

Evaluation of the new UKCA climate-composition model - Part 2: The Troposphere, Geosci. Model Dev., 7, 41–91, doi:10.5194/gmd-7-41-2014, 2014.

Ott, L. E., Pickering, K. E., Stenchikov, G. L., Allen, D. J., DeCaria, A. J., Ridley, B., Lin, R.-F., Lang, S., and Tao, W.-K.: Production of lightning NOx and its vertical distribution calculated from three-dimensional cloud-scale chemical transport model simulations, J. Geophys. Res., 115, D04 301, doi:10.1029/2009JD011880, 2010.

Pöschl, U., von Kuhlmann, R., Poisson, N., and Crutzen, P. J.: Development and Intercomparison of Condensed Isoprene Oxidation Mechanisms for Global Atmospheric Modeling, J. Atmos. Chem, 37, 29–52, doi:10.1023/A:1006391009798, 2000.

Price, C. and Rind, D.: A simple lightning parameterization for calculating global lightning distributions, J. Geophys. Res., 97, 9919–9933, doi:10.1029/92JD00719, 1992.

Price, C. and Rind, D.: What determines the cloud-to-ground lightning fraction in thunderstorms?, Geophys. Res. Lett., 20, 463–466, doi:10.1029/93GL00226, 1993.

Price, C. and Rind, D.: Modeling global lightning distributions in a general circulation model, Mon. Weather Rev., 122, 1930–1939, 1994.

Reynolds, R. W., Smith, T. M., Liu, C., Chelton, D. B., Casey, K. S., and Schlax, M. G.: Daily ~~High-Resolution-Blended Analyses for Sea Surface Temperature~~high-resolution-blended analyses for sea surface temperature, J. Climate, 20, 5473–5496, doi:10.1175/2007JCLI1824.1, 2007.

Reynolds, S. E., Brook, M., and Gourley, M. F.: Thunderstorm charge separation, ~~Journal of Meteorology~~J. Meteorol., 14, 426–436, 1957.

Ridley, B., Pickering, K., and Dye, J.: Comments on the parameterization of lightning-produced NO in global chemistry-transport models, Atmos. Environ., 39, 6184–6187, doi:10.1016/j.atmosenv.2005.06.054, 2005.

Schumann, U. and Huntrieser, H.: The global lightning-induced nitrogen oxides source, Atmos. Chem. Phys., 7, 3823–3907, doi:10.5194/acpd-7-2623-2007, 2007.

Squire, O. J., Archibald, A. T., Griffiths, P. T., Jenkin, M. E., Smith, D., and Pyle, J. A.: Influence of isoprene chemical mechanism on modelled changes in tropospheric ozone due to climate and land use over the 21st century, Atmos. Chem. Phys, 15, 5123–5143, doi:10.5194/acp-15-5123-2015, 2015.

Stevenson, D. S., Dentener, F. J., Schultz, M. G., Ellingsen, K., van Noije, T. P. C., Wild, O., Zeng, G., Amann, M., Atherton, C. S., Bell, N., Bergmann, D. J., Bey, I., Butler, T., Cofala, J., Collins, W. J., Derwent, R. G., Doherty, R. M., Drevet, J., Eskes, H. J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J.-F., Lawrence, M. G., Montanaro, V., Müller, J.-F., Pitari, G., Prather, M. J., Pyle, J. A., Rast, S., Rodriguez, J. M., Sanderson, M. G., Savage, N. H., Shindell, D. T., Strahan, S. E., Sudo, K., and Szopa, S.: Multimodel ensemble simulations of present-day and near-future tropospheric ozone, J. Geophys. Res., 111, D08 301, doi:10.1029/2005JD006338, 2006.

Thompson, A. M., Witte, J. C., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Fortuin, J. P. F., and Kelder, H. M.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology 2. Tropospheric variability and the zonal wave-one, J. Geophys. Res., 108, 8241, doi:10.1029/2002JD002241, 2003.

Tost, H., Jöckel, P., and Lelieveld, J.: Lightning and convection parameterisations - uncertainties in global modelling, Atmos. Chem. Phys., 7, 4553–4568, doi:10.5194/acpd-7-6767-2007,2007.

Walters, D. N., Williams, K. D., Boutle, I. A., Bushell, A. C., Edwards, J. M., Field, P. R., Lock, A. P., Morcrette, C. J., Stratton, R. A., Wilkinson, J. M., Willett, M. R., Bellouin, N., Bodas-Salcedo, A., Brooks, M. E., Copsey, D., Earnshaw, P. D., Hardiman, S. C., Harris, C. M., Levine, R. C., MacLachlan, C., Manners, J. C., Martin, G. M., Milton, S. F., Palmer, M. D., Roberts, M. J., Rodríguez, J. M., Tennant, W. J., and Vidale, P. L.: Geoscientific Model Development The Met Office Unified Model Global Atmosphere 4.0 and JULES Global Land 4.0 configurations, Geosci. Model Dev., 7, 361–386, doi:10.5194/gmd-7-361-2014, 2014.

Wild, O.: Modelling the global tropospheric ozone budget: Exploring the variability in current models, Atmos. Chem. Phys., 7, 2643–2660, doi:10.5194/acp-7-2643-2007,2007.

Wilson, D. R., Bushell, A. C., Kerr-Munslow, A. M., Price, J. D., and Morcrette, C. J.: PC2: A prognostic cloud fraction and condensation scheme. I: Scheme description, Q. J. Roy. Meteor. Soc., 134, 2093–2107, doi:10.1002/qj.333, 2008a.

Wilson, D. R., Bushell, A. C., Kerr-Munslow, A. M., Price, J. D., Morcrette, C. J., and Bodas-Salcedob, A.: PC2: A prognostic cloud fraction and condensation scheme. II: Climate model simulations, Q. J. Roy. Meteor. Soc., 134, 2109–2125, doi:10.1002/qj.332, 2008b.

Wong, J., Barth, M. C., and Noone, D.: Evaluating a lightning parameterization based on cloud-top height for mesoscale numerical model simulations, Geosci. Model Dev., 6, 429–443, doi:10.5194/gmd-6-429-2013,2013.

Wu, S., Mickley, L. J., Jacob, D. J., Logan, J. A., Yantosca, R. M., and Rind, D.: Why are there large differences between models in global budgets of tropospheric ozone?, J. Geophys. Res., 112, D05 302, doi:10.1029/2006JD007801, 2007.

Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J.-F., Naik, V., Stevenson, D. S., Tilmes, S., Voulgarakis, A., Wild, O., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsø ren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Horowitz, L. W., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R. B., Shindell, D. T., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 2063–2090, doi:10.5194/acp-13-2063-2013, 2013.

Ziemke, J. R., Chandra, S., Labow, G. J., Bhartia, P. K., Froidevaux, L., and Witte, J. C.: A global climatology of tropospheric and stratospheric ozone derived from Aura OMI and MLS measurements, Atmos. Chem. Phys., 11, 9237–9251, doi:10.5194/acp-11-9237-2011, 2011.

Table 1. Spatial comparisons of correlation, errors and bias of annual tropospheric ozone column between model runs and the MLS/OMI satellite climatology product over the range ±60°. Adjusted root mean square error (RMSE) refers to the RMSE following the subtraction of the mean bias from the field.

Run	r	RMSE (DU)	Mean bias (DU)	adjusted RMSE (DU)
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~~ZERO. All terms in Tg yr^{-1} except Burden which is in Tg and lifetime which is in days. The percentage difference with respect to the CTH budget is shown in brackets. In addition to the ~~usual~~ tropospheric budget terms, the whole atmospheric ozone burden is also 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.

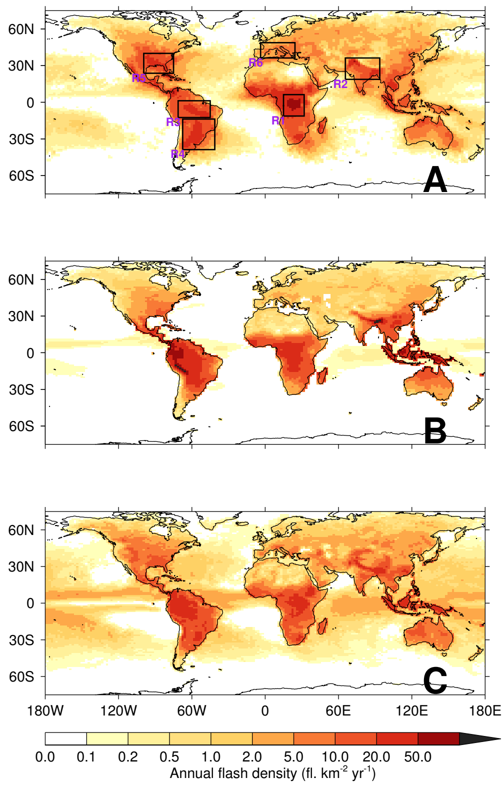


Figure 1. Annual flash rates from (A) a combined climatology from LIS/OTD satellite observations spanning ~~1995-2010~~1995-2011, (B) the CTH scheme using the year 2000 of UKCA output and (C) the ICEFLUX scheme using the year 2000 of UKCA output. The horizontal resolution of the climatology product has been degraded to match that of the model which is 1.875° longitude by 1.25° latitude. Boxes for the regions R1–R6 correspond to regions of interest for which the annual cycles are shown in Figure 3.

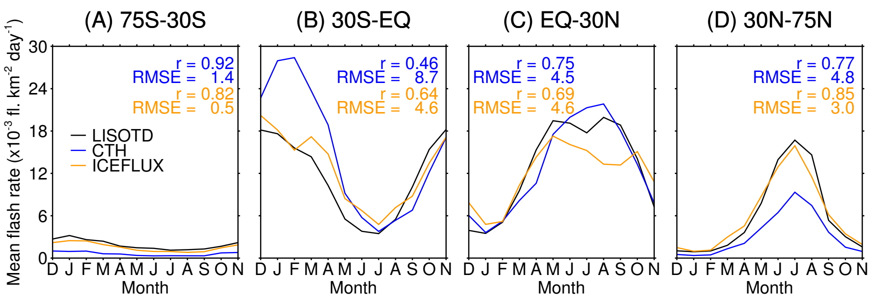


Figure 2. Mean monthly flash rate averaged over four latitudinal bands for the two different schemes for 2000 and the LIS/OTD climatology spanning ~~1995-2010~~1995-2011. The points use one year of UKCA model output and a combined climatology from LIS/OTD satellite observations spanning ~~1995-2010~~1995-2011. Also given are the temporal correlations (r) between the CTH ~~model~~ scheme (blue) and LIS/OTD and between ICEFLUX (orange) and LIS/OTD. The corresponding root mean square errors (RMSE) are given in units of $10^{-3}\,\mathrm{fl.\,km^{-2}yr^{-1}}$.

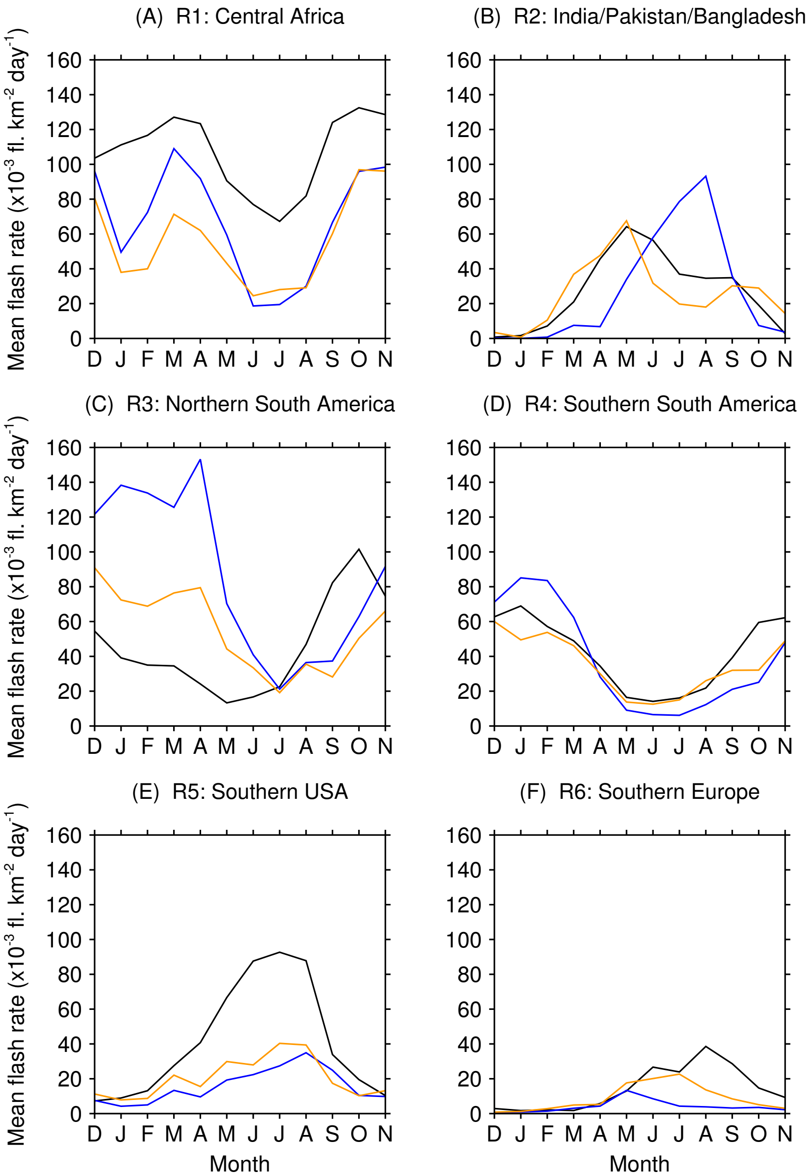


Figure 3. Mean monthly flash rate averaged over six regions (R1–R6) for the two different schemes for year 2000 and the LIS/OTD climatology spanning 1995-2011. Lines represent the lightning simulated using the CTH approach (blue) and the ICEFLUX approach (orange), and the LIS/OTD observed climatology (black). Regions R1-R6 are shown as boxes on Figure 1.

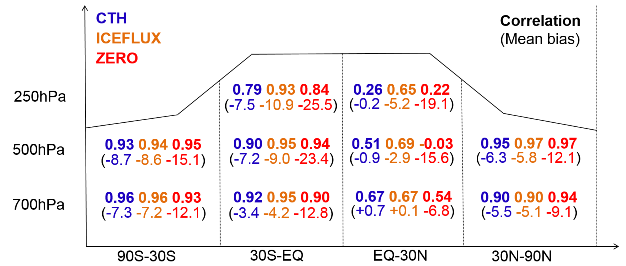


Figure 4. Temporal correlations and mean biases of the annual cycle of modelled ozone in UKCA over the year 2000 compared to a climatology of ozone sonde measurements averaged over 1980-1993 and 1997-2011. The simulated ozone data was interpolated to the location and pressure level of the sonde measurements.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

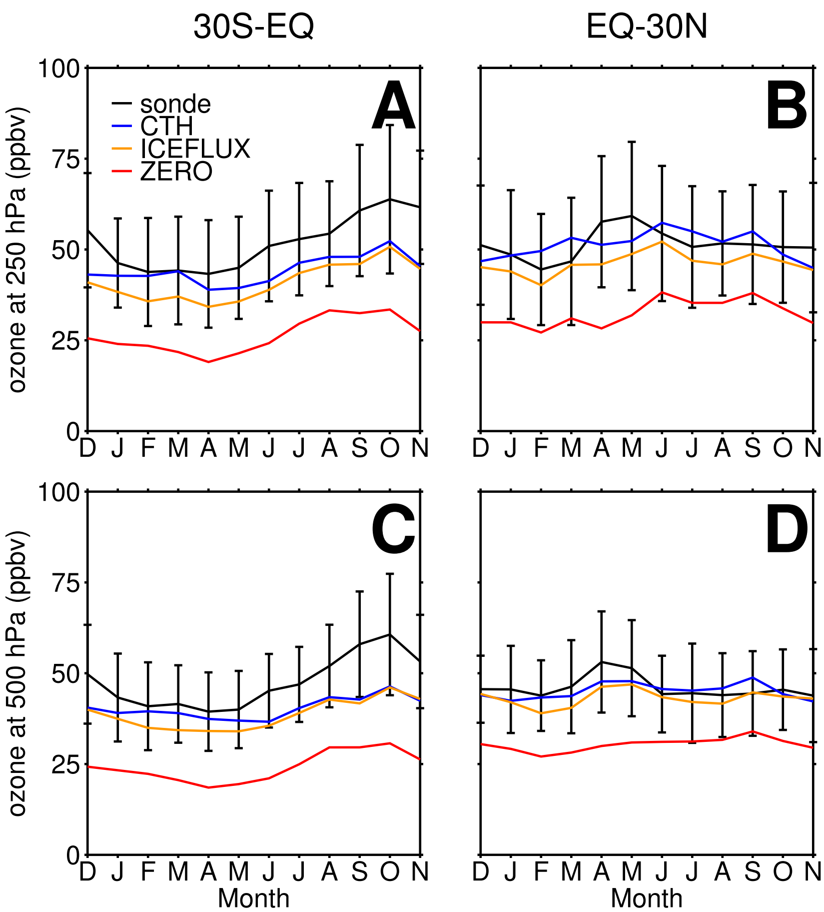


Figure 5. Middle and upper tropospheric UKCA simulated ozone concentration for the year 2000 compared to a climatology of sonde measurements averaged over 1980-1993 and 1997-2011. These cycles correspond to the 500 hPa and 250 hPa correlations for 30S-EQ and EQ-30N in Figure 4. The vertical black bars show the average interannual standard deviation for each group of stations.

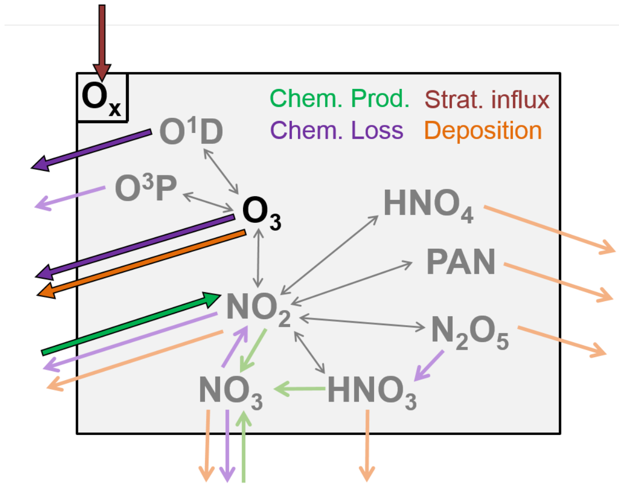


Figure 6. The UKCA definition of O_x species and the O_x budget. Major contributors are shown in bright colours and black outlines, minor contributors in pale colours. ~~Black~~ Grey arrows are reactions between O_x species and therefore result in no production or loss. The stratospheric influx is not determined for individual species. Instead the total O_x influx is inferred to balance the production and loss terms. The burden and stratospheric influx of O_x ~~is~~ are dominated by the burden and stratospheric influx of O_3.

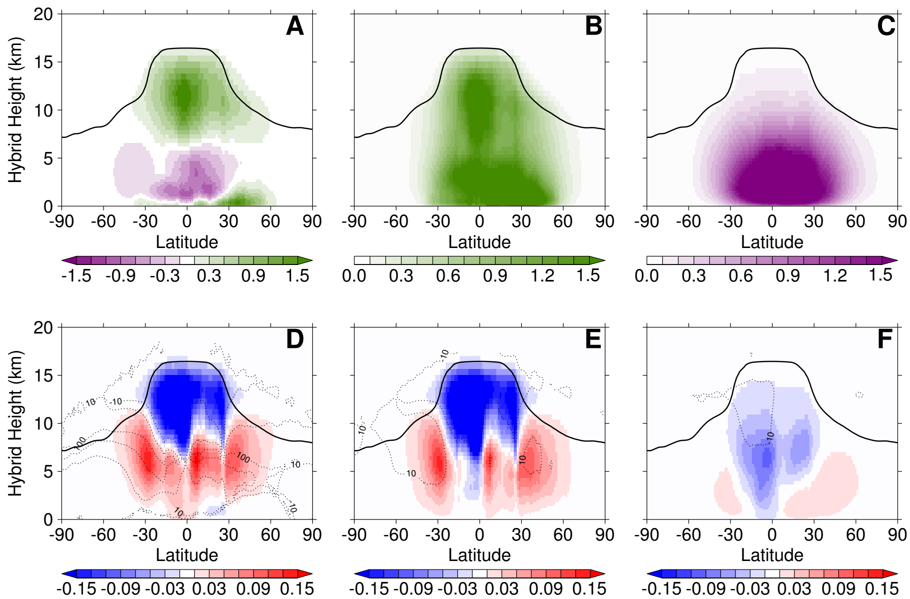


Figure 7. Annual total zonal-altitudinal distributions of O_x reaction fluxes for CTH for the year 2000. These fluxes are A) Net production, B) gross production, and C) gross loss of O_x. The respective differences between simulations using the ICEFLUX scheme and the CTH scheme are shown in D-F. All 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.

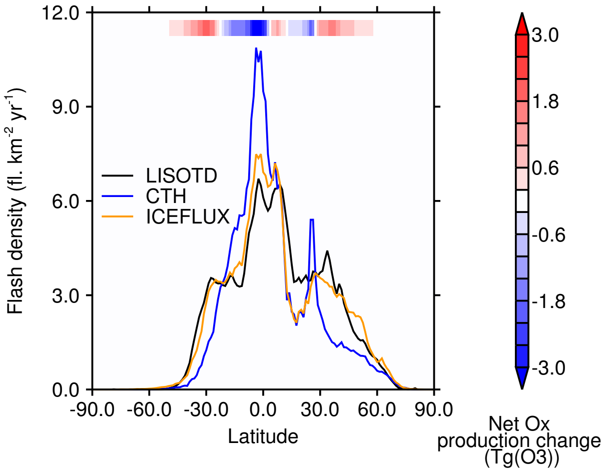


Figure 8. Zonal mean lightning flash rate from the LIS/OTD climatology and as modelled by CTH and ICEFLUX. The zonal changes in net tropospheric column O_x production (ICEFLUX-CTH) are shown by the colour bar. The units of O_x are expressed as a mass of ozone.

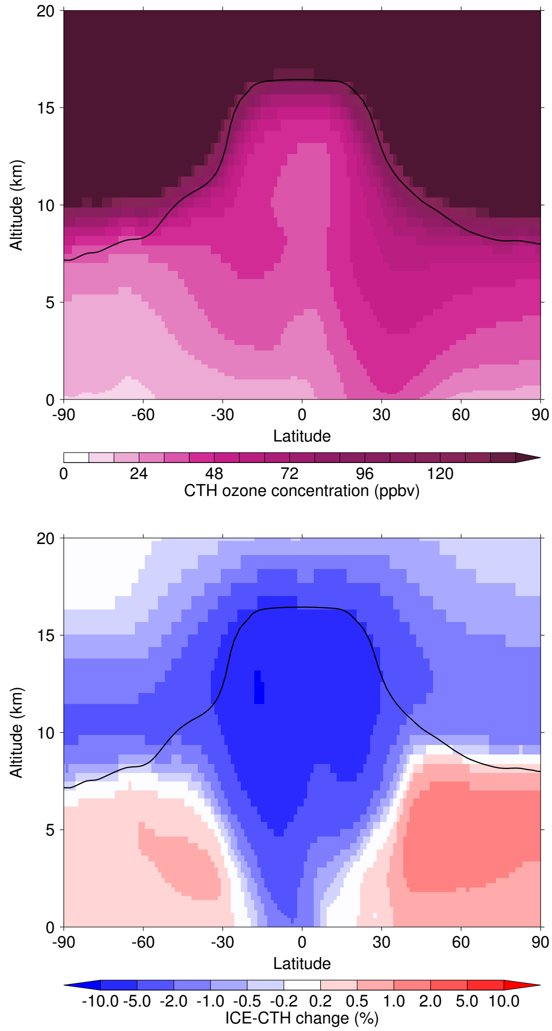


Figure 9. Annual mean distribution of ozone concentration modelled using the CTH approach, and the percentage difference between ICEFLUX and CTH simulated ozone concentration. The solid line shows the mean annual tropopause as diagnosed using the modelled meteorology.

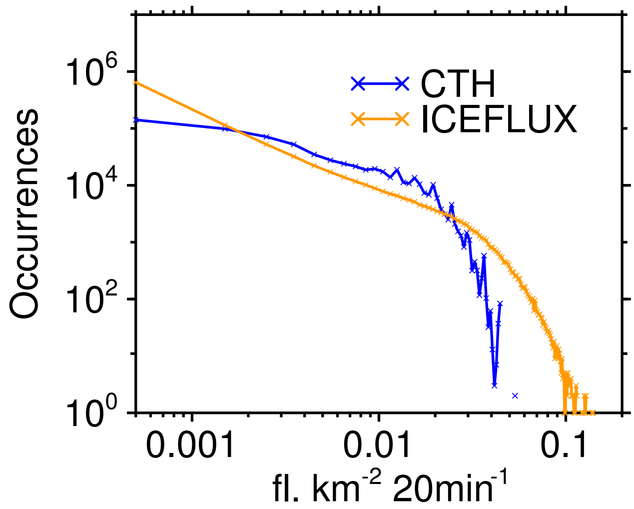


Figure 10. Frequency distribution of continental lightning flash rates using all time steps, for one month (September 2000) as modelled by the CTH and ICEFLUX schemes. The binsize used is 0.001 $\mathrm{fl.\,km^{-2}20min^{-1}}$ with crosses placed at the centre value of each bin.

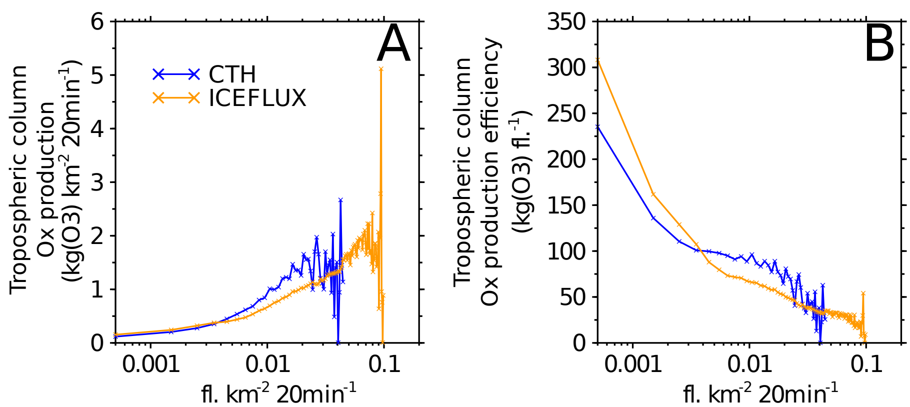


Figure 11. Two metrics of intial gross column O_x production as a result of continental lightning simulated by the CTH and ICEFLUX schemes. The cells used in each bin correspond to those used in Figure 10. The metrics are A) mean column O_x production in each bin, and B) mean column O_x production per 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 10. The units of O_x are expressed as a mass of ozone.