Global impact of lightning-produced oxidants

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Abstract

Lightning plays a major role in tropospheric oxidation, and its role on modulating tropospheric chemistry was thought to be emissions of nitrogen oxides (NO_x). Recent field and laboratory measurements demonstrate that lightning generates extremely large amounts of oxidants, including hydrogen oxides (HO_x) and O_3. We here implement the lightning-produced oxidants in a global chemical transport model to examine its global impact on tropospheric composition. We find that lightning-produced oxidants can increase global mass weighted OH by 0.3-10%, and affect CO, O_3, and reactive nitrogen substantially, depending on the emission strength of oxidants from lightning. Our work highlights the importance and uncertainties of lightning-produced oxidants, as well as the need for rethinking the role of lightning in tropospheric oxidation chemistry.

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Abstract

Lightning plays a major role in tropospheric oxidation, and its role on modulating tropospheric chemistry was thought to be emissions of nitrogen oxides (NO_x). Recent field and laboratory measurements demonstrate that lightning generates extremely large amounts of oxidants, including hydrogen oxides (HO_x) and O_3. We here implement the lightning-produced oxidants in a global chemical transport model to examine its global impact on tropospheric composition. We find that lightning-produced oxidants can increase global mass weighted OH by 0.3-10%, and affect CO, O_3, and reactive nitrogen substantially, depending on the emission strength of oxidants from lightning. Our work highlights the importance and uncertainties of lightning-produced oxidants, as well as the need for rethinking the role of lightning in tropospheric oxidation chemistry.

1. Introduction

Lightning plays a major role in tropospheric oxidation chemistry [*Murray et al.*, 2013]. It can produce nitrogen oxides, hydrogen oxides, and ozone through visible flashes, subvisible charges, and corona. The production of NO by lightning is through the Zel'dovich mechanism involving the dissociation of O_2 and N_2 and very high temperature (>4000 K) with visible flashes [*Schumann & Huntrieser*, 2007]. As the dissociation energy of O_2 (0.50 MJ mol^{-1}) and H_2O (0.50 MJ mol^{-1}) are lower than that of N_2 (0.94 MJ mol^{-1}) [*Howard & Rees*, 1996], producing O_3 and HO_x by lightning requires less energy.

The role of lightning on modulating global oxidation was mainly considered through emissions of nitrogen oxides (NO_x) [*Chameides et al.*, 1977; *Murray et al.*, 2012], which leads to the production of the major tropospheric oxidants, OH and ozone. The global lightning NO_x (LNO_x) emission is estimated to be about 2-8 Tg N/yr [*Schumann & Huntrieser*, 2007]. The lightning production of hydrogen oxides (LHO_x) was considered unimportant due to their short lifetime [*Hill & Rinker*, 1981; *Bhetanabhotla et al.*, 1985]. The lightning production of O_3 (LO_3) is shown in the laboratory to occur by corona discharges in higher amounts than LNO_x by a factor of 5-30 [*Hill et al.*, 1988; *Peyrous & Lapeyre*, 1982; *Simek & Clupek*, 2002], but lower production of O_3 was found in visible flashes [*Wang et al.*, 1998]. LO_3 by corona discharge is further supported by field measurements [*Bharali et al.*, 2015; *Bozem et al.*, 2014; *Kotsakis et al.*, 2017; *Minschwaner et al.*, 2008]. Recent studies show that extremely high amounts of HO_x and O_3 can be produced by visible flashes and subvisible charges in electrified storms [*Brune et al.*, 2021; *Jenkins et al.*, 2021].

Here we implement a simplistic parameterization for lightning HO_x and O_3 into a global chemical transport model (GEOS-Chem) to investigate the global impact of this overlooked

oxidant source (LHO_x and LO_3). Given the large uncertainties associated with current estimates of LHO_x and LO_3, we only focus on their potential global impact in this work.

2. Methods

GEOS-Chem is a global chemical transport model with transport driven by assimilated meteorological fields from the NASA Global Modeling and Assimilation Office's Modern-Era Retrospective analysis for Research and Applications, version 2 (MERRA-2) system [*Bey et al.*, 2001]. We use GEOS-Chem v12.5.0 (10.5281/zenodo.3403111) at a horizontal resolution of 4° latitude ×5°longitude with fully coupled O_3-NO_x-HO_x-VOC-aerosol chemistry ("tropchem" mechanism)[*Mao et al.*, 2010, 2013; *Park et al.*, 2004]. Methane is prescribed with monthly maps of spatially-interpolated NOAA flask data, but is allowed to advect and react [*Murray*, 2016].

The lighting NO_x in GEOS-Chem largely follows Murray et al. [2012], with lightning flash densities and convective cloud depths calculated at the native GEOS-FP meteorology resolution. The simulated climatology is further constrained by the satellite observations from the Optical Transient Detector (OTD) and Lightning Imaging Sensor (LIS), with an annual mean global flash rate of 46 flashes per second [*Christian*, 2003]. GEOS-Chem applies 500 mol N per flash for all lightning in the northern extratropics (north of 35°N), and 260 mol N per flash for the rest of the world. This approach results in a total lightning emission of 6 Tg N per year. The vertical distribution of lightning follows Ott et al. [2010], which redistributes lightning emissions vertically based on different surface types (tropical continental, tropical marine, subtropical and mid-latitude).

We scale lightning HO_x and O_3 with lightning NO_x by a factor of 10 and 100, to examine its global impact. Our estimated lightning HO_x is based on the following. The OH generated by LHO_x in each electrically active convective cell is estimated to be $3.1\times10^{25} - 2.7\times10^{26}$ molecules per second [*Brune et al.*, 2021]. Assuming globally there are 1800 electrically active convective cells every second, the global production is 3-30 T mol OH/yr. As global lightning NO_x is about 6 Tg N/yr (0.4 T mol N/yr) [*Murray*, 2016], we scale lightning HO_x by a factor of 10 and 100 on a molar basis. The scaling of O_3 is based on two facts. First, the O_3 production rate was estimated to be $0.4 - 98 \times10^{27}$ molecules per flash [*Bozem et al.*, 2014; *Kotsakis et al.*, 2017; *Minschwaner et al.*, 2008], while the NO_x production rate was estimated to be one or two orders of magnitude lower than that of O_3, with $2- 40 \times10^{25}$ molecules per flash [*Schumann & Huntrieser*, 2007]. Second, O_2 and H_2O have similar dissociation energy as mentioned above. We note that the resulting LO_3 is in the range of 140-1400 Tg O_3/yr, comparable to stratosphere-troposphere exchange (STE) ozone flux [*McLinden et al.*, 2000].

We add lightning HO_x and O_3 in a similar fashion as lightning NO_x in the model. As GEOS-Chem is run with operator splitting, we allow radicals (OH + HO_2) and O_3 to accumulate over the course of the emission step (20 mins in our model setup), which leads to a pulse of HO_x radicals at the beginning of the chemistry timestep (also 20 mins). We find in our current model setup that at the end emission timestep, OH and HO_2 are built up to the order of $\sim10^8$ molecules/cm^3, an order of magnitude smaller than observed [*Brune et al., 2021*]. Once the chemistry time step starts, the spikes of OH and HO_2 rapidly decrease due to the dominant loss

of radicals through the $OH+HO_2$ reaction, similar to the box model simulations of HO_x produced from lightning [*Brune et al.*, 2021]. Within seconds, the radical levels return to background levels, while their impact on OH reactants (CO, CH_4) can be significant due to high levels of OH exposure. We consider this treatment a better representation of the impact of lightning, rather than assuming a constant radical source throughout the whole chemistry time step, in which case $OH+HO_2$ reaction would be much less of a HO_x sink. In contrast, treating lightning O_3 production as a pulse or time averaged production should not make much difference on ozone, as the lifetime of ozone is on the order of months in the middle and upper troposphere.

We conducted five model simulations for the year of 2016, as illustrated in Table 1.

Table 1. Model set up for base run and sensitivity tests

Model run	Lightning emissions	Magnitude (molar basis)
Base	LNO_x	
H10	$LNO_x + LHO_x$	$LOH=10\times LNO_x$, $LHO_2=10\times LNO_x$
H100	$LNO_x + LHO_x$	$LOH=100\times LNO_x$, $LHO_2=100\times LNO_x$
H10_O10	$LNO_x + LHO_x + LO_3$	$LOH=10\times LNO_x$, $LHO_2=10\times LNO_x$, $LO_3=10\times LNO_x$
H100_O100	$LNO_x + LHO_x + LO_3$	$LOH=100\times LNO_x$, $LHO_2=100\times LNO_x$, $LO_3=100\times LNO_x$

3. Results

Figure 1 shows the global impact of lighting produced oxidants on annual ozone in the upper troposphere. We find that adding LHO_x alone (H10 and H100) will reduce ozone concentrations in the middle and upper troposphere, due to enhanced ozone loss through $OH/HO_2 + O_3$ as well as reduced ozone production efficiency through $OH + NO_2$ [*Hu et al.*, 2017]. For the case of H10, we find that LHO_x decreases O_3 in the upper troposphere by 1-2 ppbv on an annual mean basis, mainly over regions where lightning flashes are intense. For the run of H100, O_3 can be reduced by 3-7 ppbv in the upper troposphere.

The O_3 decrease due to LHO_x can be compensated by the addition of LO_3. We show in Figure 1 that with the case of H10_O10, annual mean O_3 is in fact enhanced by 1-3 ppbv in the upper troposphere. For the case of H100_O100, annual mean O_3 is enhanced by 10-30 ppbv mainly over lightning-intense regions. As the O_3 lifetime is on the order of ~1 month, LO_3 can effectively increase ozone in the middle and upper troposphere.

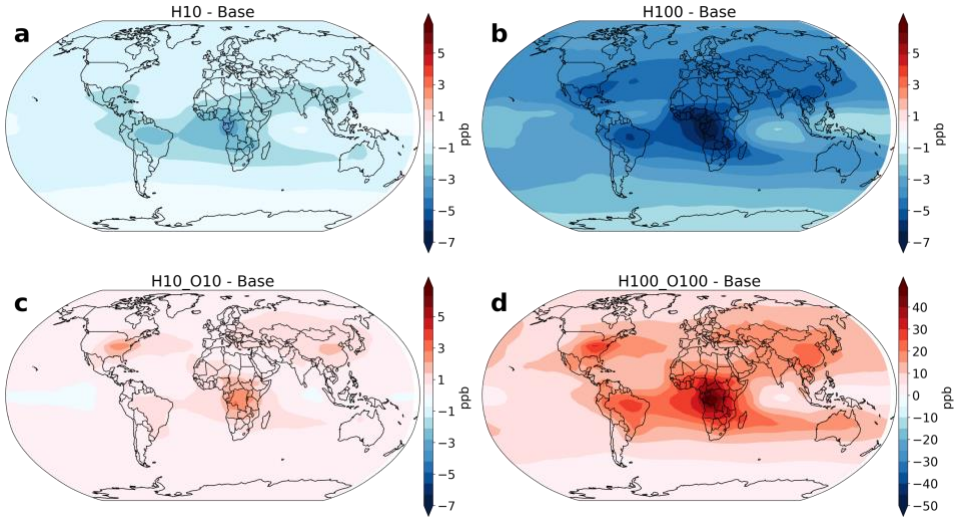

Figure 1 Global impact of lightning oxidants on annual mean O_3 in the upper troposphere (8 km). Each panel represents the difference between a sensitivity run and base model run: (a) H10 – Base (b) H100 – Base (c) H10_O10 – Base (d) H100_O100 – Base. H10 and H100 are referred to the runs with LHO_x, and H10_O10 and H100_100 are referred to the runs with LHO_x and LO_3 (see Table 1 for details).

Figure 2 shows the impact of lightning oxidants on OH, HO_2, and CO. We note that both H100 and H100_O100 increases OH in the upper troposphere by up to 10%. The increase of OH is mainly due to the decrease of CO, which allows OH to reach another steady state with higher concentrations, as CO accounts for more than 50% loss of ambient OH [*Mao et al.*, 2009]. The increase of HO_2 is in part due to direct emission and in part due to OH+CO. We see a mild decease of CO with H10 and H10_O10, but a much bigger decrease with H100 and H100_O100.

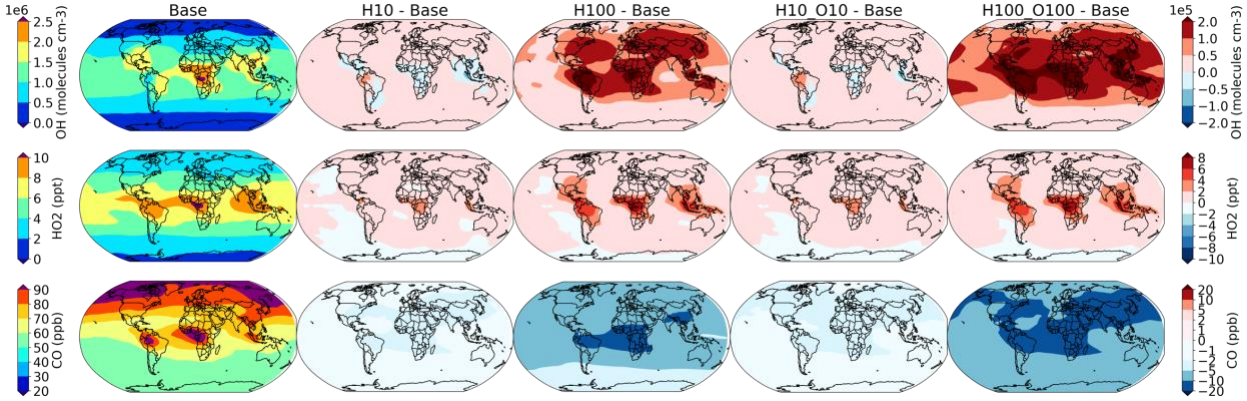

Figure 2 Effect of lightning-produced oxidants on OH (top), HO_2 (middle) and CO (bottom) in the upper troposphere (8 km). Each panel represents the difference between a sensitivity run and base model run: (a) H10 – Base (b) H100 – Base (c) H10_O10 – Base (d) H100_O100 – Base. H10 and H100 are referred to the runs with LHO_x, and H10_O10 and H100_100 are referred to the runs with LHO_x and LO_3 (see Table 1 for details).

Table 2 summarizes the global impact of different sensitivity runs. In the base run, global OH production and loss are 220 T mol/yr, in agreement with other model studies [*Lelieveld et al.*, 2016]. The imbalance between Prod(OH) and Loss(OH) in sensitivity runs (H_10, H_100, H10_O10, and H100_O100) reflects the added oxidants (OH, HO_2 and O_3) from lightning before the chemistry timestep. We show that mass-weight global mean OH increases by 0.3%, 3%, 0.8% and 9% with H_10, H_100, H10_O10, and H100_O100 respectively, with little difference on the OH Northern hemisphere to Southern hemisphere ratio. We note that the impact on global mean OH is smaller than previously estimated by Brune et al. [2021], likely due to two reasons. First, the estimate by Brune et al. [2021] is the direct impact on instantaneous global OH, i.e., a snapshot of the global OH field with pulses from LHO_x included, while our calculation is based on the OH concentrations after chemistry timestep (20 min), during which HO_x pulses decay to background levels within the first few seconds. The impact on global OH from our estimate is mainly resulting from changes on the burden of OH sources and sinks, such as O_3 and CO. Second, the global mass-weighted OH is weighted towards the lower troposphere [*Lawrence et al.*, 2001], while LHO_x in our model is mainly distributed into the middle and upper troposphere. As a result, the global mass-weighted OH is relatively insensitive to the changes of OH field in the middle and upper troposphere

Lightning-produced oxidants also impact the global CH_4 budget. We find in Table 2 that the global loss of CH_4 increases by 15-110 Tg CH_4/yr from our sensitivity runs. As CH_4 oxidation is rather slow in the upper troposphere, we find that the impact on CH_4 is mainly in the lower troposphere where the potential for LHO_x generation is currently unknown.

Lightning-produced O_3 offers an alternative explanation to ozone layering in the free troposphere. Atmospheric observations often show layers of high O_3 with high moisture [*Newell et al.*, 1999; *Oltmans et al.*, 1996], and these ozone layers are unlikely from stratosphere intrusion because of the high moisture. On the other hand, if these O_3 layer are produced during lightning, they can be transported thousands of kilometers away from the source region because the O_3 lifetime in the upper troposphere is about a month. Lightning-produced O_3 is also consistent with the seasonality of ozone layering, which shows a summer maximum in northern mid-latitude[*Colette & Ancellet*, 2005].

Table 2 Global impact of lighting produced oxidants on tropospheric composition

	Base	H10	H100	H10_O10	H100_O100
Global mass-weighted OH (10^6 molecules/cm^3)	1.212	1.216	1.252	1.222	1.324
OH NH/SH ratio	1.21	1.21	1.22	1.21	1.23
Prod Ox (Tg/yr)	5027	5058	5088	4983	4450
Loss Ox (Tg/yr)	4763	4804	4871	4918	6115
Prod OH (Tmol/yr)	222.1	223.8	230.4	225.5	249.8
Loss OH (Tmol/yr)	222.1	228.1	274.1	229.9	293.5
Prod CO (Tmol/yr)	57.2	58.0	61.3	58.2	63.5
Loss CO (Tmol/yr)	87.6	88.7	93.0	88.9	95.4
Loss CH_4 (Tg CH_4/yr)	564.8	579.2	633.6	582.4	676.8

Prod HNO_3 (Tmol/yr)	3.81	3.84	3.93	3.84	3.95
Prod HNO_2 (Tmol/yr)	1.41	1.35	1.36	1.31	1.11
Loss HNO_2 (Tmol/yr)	1.41	1.35	1.36	1.31	1.11

Figure 3 shows the global impact of lighting produced oxidants on the partitioning of reactive nitrogen in the upper troposphere. With newly added OH and HO_2 produced by lightning, $OH+NO_2$ is thus enhanced in the middle and upper troposphere, leading to a higher production of HNO_3 and lower ozone production efficiency (Table 2). We find that both NO and NO_2 in the upper troposphere decreased by 10-20 pptv on an annual mean basis over the tropics and subtropics where lightning activity is high. In the meantime, we see an increase in most nitrogen reservoirs including HNO_3, peroxyacetyl nitrate (PAN) and peroxynitric acid (HNO_4). The only exception is PAN in H100 and H100_O100, likely due to enhanced loss of PAN through its reaction with OH. This shift of NOx towards their reservoirs may have important implication on nitrogen chemistry in the upper troposphere.

Our results in this work are mainly based on the annual mean, and we expect the impact on shorter time scales to be different. For example, we expect a significant increase of HONO on a short time scale (on the order of hours to days) due to the production of OH and NO as shown in box model simulations [*Brune et al.*, 2021]. Once HONO is photolyzed and returns OH and NO, enhanced OH will lead to higher peroxy radicals that then convert NO_2 to peroxy nitrates and other nitrogen reservoirs, resulting in lower concentrations of NO and NO_2. Consequently, we see a decrease of annual mean HONO production and loss in sensitivity runs in Table 2.

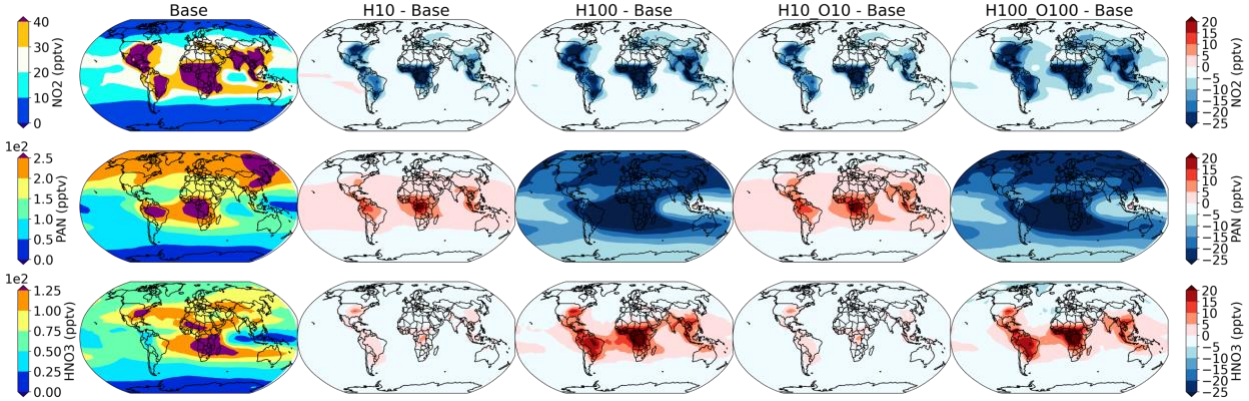

Figure 3 Effect of lightning-produced oxidants on NO_2 (top), PAN (middle) and HNO_3 (bottom) in the upper troposphere (8 km). Each panel represents the difference between a sensitivity run and base model run: (a) H10 – Base (b) H100 – Base (c) H10_O10 – Base (d) H100_O100 – Base. H10 and H100 are referred to the runs with LHO_x, and H10_O10 and H100_100 are referred to the runs with LHO_x and LO_3 (see Table 1 for details).

4. Discussion

Here we implement a new source of oxidants ($OH + HO_2 + O_3$) from lighting into a global chemical transport model, to examine its potential impact on tropospheric chemistry. Due to

large uncertainties associated with lightning and its emissions, we conduct only a few sensitivity tests to provide a qualitative assessment. However, we find that this new source of oxidants can increase global mass weighted OH by 0.3-10%, and affect CO, O_3 CH_4 and reactive nitrogen substantially, depending on the emission strength of oxidants from lightning (Table 2).

Large uncertainties remain in many aspects. First, we assume that lightning NO_x and oxidants are instantly mixed in each model grid box when there is lightning. In fact, field observations suggest that NO_x, HO_x, and ozone are likely produced in different parts of storm clouds (NO_x dominates in visible flashes, HO_x and O_3 dominates in subvisible discharges and coronas) [*Jenkins et al.*, 2021; *Brune et al.*, 2021]. It remains unclear how this instant mixing would affect the non-linear behavior of HO_x-NO_x-O_3 chemistry and its possible consequence[*Gressent et al.*, 2016]. Second, the volume of a model grid box in the upper troposphere (approximately 400 km × 500 km × 1 km) is about 1000 times bigger than the typical lightning mapping array (LMA) volume for one convective cell ($1x10^{17}$ cm^3) [*Brune et al.*, 2021], leading to a dilution effect on radical loss through OH + HO_2. However, there are typically many electrically active convective cells occupying one model grid box, so the grid cell box might be only 10 to 100 times larger than the volume of all the convection within that cell. Also, the fact that we allow the model to build up radicals over the emission timestep (20 mins or 1200 seconds) can somewhat compensate this dilution effect (Figure 4). These effects also imply that our model results may vary with model resolution and the choice of emission timesteps.

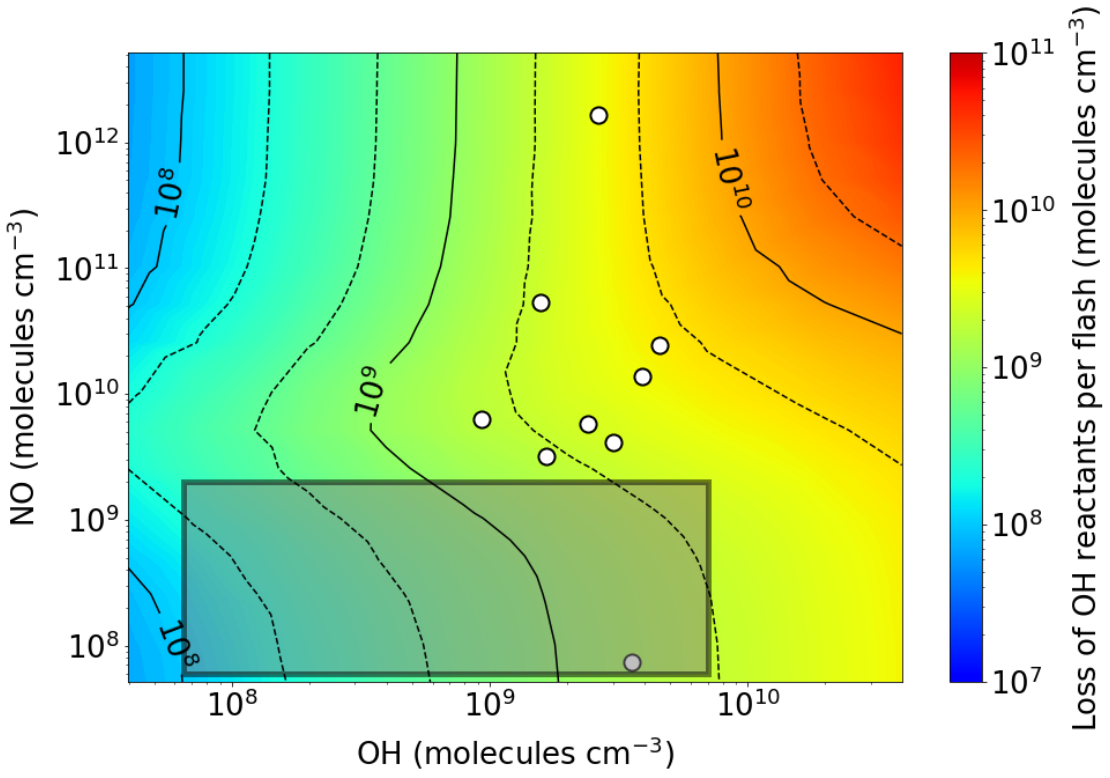

Figure 4 OH loss per flash through reactions with all OH reactants (excluding HO_2 and NO_2), calculated by a box model [*Brune et al.*, 2021]. We exclude HO_2 and NO_2 because their reactions with OH are considered permanent HO_x sinks. The white dots represent the observations from

DC3 aircraft campaign, and the gray box represents the range of model values in the upper troposphere after the emission timestep but before the chemistry timestep.

Our results are further complicated by the non-linear HO_x-NO_x-O_3 chemistry. We show in Figure 4 that the extent of OH loss through reactants other than HO_2 and NO_2, is largely dependent on the relative concentrations of OH and NO. In fact, NO could effectively extend OH lifetimes by producing HONO and reducing OH loss through OH+HO_2 in the first few seconds. As HONO photolyzes and returns OH, OH+HO_2 becomes a minor loss for OH. As shown in Figure 4, the high concentrations of observed NO_x from lightning are not reproduced in the global model, in part due to instant mixing, leading to a lower fraction of OH loss through CO and other OH reactants. This non-linear chemistry is therefore sensitive to co-location of LHO_x, LNO_x and LO_3, as well as model configurations.

Our work suggests the strong need of revisiting current estimates of global lightning NO_x emissions, with newly added HO_x and O_3. On one hand, OH and HO_2 may further shorten NO_x lifetimes in upper troposphere (Figure 2 and 3), pointing to a higher level of global LNO_x [*Nault et al.*, 2017]. On the other hand, LO_3 offers an additional source for ozone in the free troposphere, indicating a need for reducing lightning NO_x emission [*Sauvage et al.*, 2007]. In addition, we show that the nitrogen partitioning is indeed sensitive to lightning-produced oxidants (Figure 3). The role of lightning in tropospheric chemistry may be redefined when LNO_x, LHO_x, and LO_3 are all taken into account.

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Data Availability Statement

The model output for both base run and sensitivity runs can be accessed online (at Dryad, Dataset, https://doi.org/10.5061/dryad.t4b8gtj24).

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