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Lightning NO_x emissions over the USA investigated using TES, NLDN, LRLDN, IONS data and the GEOS-Chem model

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Abstract

Knowledge of the lightning source of NO_x is required to better understand NO_x and ozone distributions and changes in the troposphere, the oxidising capacity of the troposphere as well as some of the feedbacks between chemistry and climate change. In this paper, we use the National Lightning Detection (NLDN) and the Long Range Lightning Detection Network (LRLDN) data as well as the HYPSLIT transport and dispersion model to show the presence of ozone enhanced layers downwind of convective events in the ozone vertical profiles from the Tropospheric Emission Spectrometer (TES) instrument over the USA in July 2006. We use the TES dataset in conjunction with ozonesonde measurements from the Intercontinental Chemical Transport Experiment (INTEX) Ozonesonde Network Study (IONS) 2006 to test the parameterization of the lightning source of NO_x in a global chemistry transport model. We find that the global 3-D chemistry transport model GEOS-Chem with a lightning activity calculated with a parameterization based on cloud top height, scaled regionally and monthly to OTD/LIS (Optical Transient Detector/Lightning Imaging Sensor) climatology, and with a production of 260 mol NO/Flash captures the ozone enhancements seen by TES, but underestimates their intensities. We show that the model's ability to reproduce the location of the enhancements is due to the fact that this model reproduces the pattern of the convective events occurrence on a daily basis during the summer of 2006, even though it does not well represent the relative distribution of lightning intensities. By imposing an updated lightning NO production value of 520 mol NO/Flash, we decrease the bias between TES and GEOS-Chem over the US in July 2006 by 40%.

1 Introduction

Research on the strength and distribution of the source of nitrogen oxides (NO_x=NO+NO_2) from lightning has been the subject of increasing literature in the past decade (for a review, see Schumann and Huntrieser, 2007). Indeed, a better

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knowledge of this source is required to understand NO_x and ozone distributions in the troposphere, in particular in the upper troposphere where most of the lightning NO_x emissions are deposited by thunderstorms (Pickering et al., 1998). Because ozone production is not a linear function of the NO_x level, the knowledge of this source is also needed to assess the present and future impact of anthropogenic sources on upper tropospheric ozone. NO_x has an indirect climate impact via its effect on ozone and hydroxyl radical (OH) concentrations. Hence, quantifying this source is also critical to understanding feedbacks between climate change and atmospheric chemistry (IPCC, 2007).

Over the United States, lightning contributes substantially to the ozone maximum observed in the upper troposphere over the southeastern United States in summer (Li et al. 2005; Cooper et al., 2006, 2007). Analyses of observation campaigns have also demonstrated lightning contributions to the ozone and NO_x concentrations over the middle US (e.g.: Ridley et al., 1994; Jaegle et al., 1998; Stith et al., 1999; DeCaria et al., 2000, 2005), the East Coast of the US (Hudman et al., 2007) and over the downwind Atlantic region (e.g.: Brunner et al., 1998, 2001; Liu et al., 1999; Thompson et al., 1999; Jeker et al., 2000; Crawford et al., 2000; Allen et al., 2000) in summer and fall. The relative contribution of this source to the ozone observed over these regions vs. the contributions of the transport from the boundary layer by convection and from the stratosphere is a source of debate. Li et al. (2005) show that the elevated ozone concentrations in the upper troposphere over the Southeastern region in summer are mainly due to convective transport of pollution created in the boundary layer which are lifted and trapped in an anticyclone, while ozone production in-situ, from ozone precursors and NO_x from lightning, also contributes to this enhancement but plays a secondary role. Thompson et al. (2007a,b), using ozone and P-T-U soundings, found persistent stratospheric influences over northeastern North America free tropospheric ozone in summer 2004 (IONS-04; Intercontinental Transport Experiment (INTEX) Ozonesonde Network Study) and south central North America in summer 2006 (Thompson et al., 2008). In both cases, stratospheric origins were implicated

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in 20–25% of the tropospheric ozone column, with convection and lightning sources somewhat less, 10–15%. Detailed analysis of the IONS site at Beltsville, Maryland (39 N, 77 W) over four summers, 2004–2007, found that stratospheric influences on tropospheric ozone above the boundary layer, were typically >30% of the column (Yorks et al., 2008). Cooper et al. (2006, 2007) used a FLEXPART approach to IONS sonde analysis. When stratospheric ozone influences are filtered from the IONS sondes, upper tropospheric ozone origins are dominated by lightning NO_x, more than 80% between 9–11 km.

Here, we report new satellite ozone observations that can be used to test and constrain the parameterization of the lightning source of NO_x in global models. In the present study, the vertical ozone profiles from TES are used in conjunction with NLDN and LRLDN lightning data, ozonesonde measurements from the IONS project, and the GEOS-Chem model to investigate our understanding of lightning emissions over the United States during summer 2006.

2 Data, model and simulations

2.1 TES observations

The TES instrument is an infrared Fourier transform spectrometer with a spectral resolution of 0.1 cm^{-1} and a spectral range from 650–2250 cm^{-1} (Beer et al., 2001) which was launched aboard the NASA's Aura satellite in July 2004. Standard products include vertical profiles of ozone, CO and water vapor obtained from nadir observations. The vertical resolution of TES nadir ozone retrievals is about 6 km for cloud-free scenes, with sensitivity to both the lower and upper troposphere (Worden et al., 2004; Bowman et al., 2006; Jourdain et al., 2007). The quality of TES ozone retrievals in the troposphere has been evaluated using ozonesonde measurements (Worden et al., 2007; Nassar et al., 2008) as well as lidar measurements (Richards et al., 2008). TES total and stratospheric ozone columns have also been computed and compared with

the observations from the Ozone Monitoring Instrument and Microwave Limb Sounder (Osterman et al., 2008). The algorithm used for retrieving the vertical trace gas profiles from TES radiances is based on an optimal estimation method (Rodgers, 2000) and is described in Worden et al. (2004) and Bowman et al. (2006). The relationship between elements of the true trace gas profile and of the retrieved profile can be expressed as:

$$\hat{\boldsymbol{x}} = \boldsymbol{x}_a + \mathbf{A}(\boldsymbol{x} - \boldsymbol{x}_a) + \mathbf{G}\boldsymbol{\varepsilon} \quad (1)$$

where $\hat{\boldsymbol{x}}$ is the logarithm of the retrieved profile, $\boldsymbol{x}_a$ is the logarithm of the a priori constraint obtained from monthly mean profiles simulated with the MOZART-3 model (Horowitz et al., 2003) and binned in 10° latitude ×60° longitude grids, $\boldsymbol{x}$ is the logarithm of the true profile, $\boldsymbol{\varepsilon}$ is the radiance measurement noise, and $\mathbf{G}$ is the gain matrix converting the noise to spectral measurement error. The averaging kernel ($\mathbf{A}$) describes the sensitivity of the retrieved profile to the perturbations of the true state.

TES has several modes of observations (Beer et al., 2006). In this study, we use the Step and Stare mode that has nadir target spaced about 0.4° apart and typically covers a 50° latitude range. In order to study the ozone budget over the United States during the summertime and the pollution export from North America, 144 Step and Stare were performed over the North American and North Atlantic regions between 4 July and 21 August 2006. Figure 1 shows an example of the Step and Stare track performed during that period for 16 days coverage. For this study, we use the 39 Step and Stare performed over North America during July 2006.

2.2 Lightning data

Lightning data are used for two purposes in this study. First, we used them to relate ozone enhanced layers in the TES data to lightning events. Secondly, we also impose the distribution of lightning in the model according to these observations for our baseline and sensitivity simulations (see details in Sect. 2.3).

The National Lightning Detection Network (NLDN) detects cloud-to-ground (CG) flashes over the continental USA with a detection efficiency of at least 90% (Grogan,

2004). For regions outside the continental USA, CG flashes were detected with the Long Range Lightning Detection Network (LRLDN). The detection efficiency is 60–80% at 60 N and 40–60% at 21 N as reported in Cooper et al. (2006). We took a detection efficiency of 50% for all the LRLDN data and used these data only above 25 N. Note also that LRLDN does not include data over Canada in July 2006.

We also used the LIS/OTD 2.5 Degree Low Resolution Annual Climatology Time Series (Christian et al., 2003) to regionally adjust lightning flash rates in the model on a monthly basis (see details in Sect. 2.3).

2.3 GEOS-Chem simulations

We use the GEOS-Chem v7.04.09 global 3-D tropospheric chemistry and transport model with a horizontal resolution of 2°×2.5° and 55 layers in the vertical (http://www-as.harvard.edu/chemistry/trop/geos). This model has been described by Park et al. (2004) and a simulation driven by the assimilated meteorological GEOS-4 data from the NASA Global Modeling and Assimilation Office (GMAO) has been recently globally evaluated by Wu et al. (2006). Below we describe the different simulations performed for this study:

$-S_{\text{base}}$

For this simulation, the lightning emission parameterization is based on Price and Rind (1992) and described in Wang et al. (1998). The lightning source is scaled globally to 6 Tg N/yr. Previous work showed that NO_x concentrations predicted by the GEOS-Chem model were underestimated in the upper troposphere over the midlatitudes in summer (Martin et al., 2006; Hudman et al., 2007). Martin et al. (2006) attribute this problem to the incorrect spatial distribution of lightning activity in the model. Indeed, because lightning NO_x emissions are only scaled globally, any shortcoming in the lightning distribution in the model is reflected in the NO_x emissions distribution. Sauvage et al. (2007) shows the importance of lightning repartition on the ozone field predicted by the GEOS-Chem model. In the present study, we have regionally scaled the lightning in

the model to match the OTD/LIS climatology (Christian et al., 2003) on a monthly basis. To do this, we have run the lightning scheme of the GEOS-Chem model "off line" for the period August 2005–August 2006. The lightning flashes are then scaled regionally to match OTD/LIS on a monthly basis. The resulting flashes are also scaled to obtain an annual mean of 44 F/s globally, again consistent with OTDL/LIS climatology (Christian et al., 2003). We then simulate again the same period and NO_x emissions are scaled to 6 Tg N/yr. Over North America midlatitudes (25–50 N), the resulting NO_x emissions total 0.1 Tg N for July 2006. It is two times higher than without this scaling. This is due to the fact that the original GEOS-Chem model tends to overestimate lightning in the tropical regions and hence place too much of the 6 Tg N/yr in these regions, as noted in Martin et al. (2006). This is also shown in Fig. 2, where the initial and modified distributions of lightning for July 2006 in GEOS-Chem are presented. Note that the mean NO production per flash over the United States during July 2006 is 260 mol of NO/Flash. The value used in our baseline simulation is very close to the most recent best estimate by Schumann and Huntrieser (2007) of 250 mol NO/Flash. Finally, we have performed a full simulation with the GEOS-Chem model for the period January 2006–August 2006 driven by the GEOS-4 data updated every 3–6 h using the scaling factors for the lightning parameterization described before. Note that this is now the method adopted to distribute lightning in the most recent version of the GEOS-Chem model (Lee Murray, personal communication), but this was not available in the standard GEOS-Chem at the time of our study. Initial concentrations are from the Near-Real-time Simulation of the GEOS-Chem model.

$-S_{\text{noanth}}$

In this simulation, anthropogenic sources over North America, for latitude ranging between 25 N–50 N, have been turned off.

$-S_{\text{noligh}}$

To study the influence of the lightning NO_x emissions from midlatitudes North America on the ozone field predicted by GEOS-Chem, we have also performed an additional

simulation with the GEOS-Chem model, in which lightning NO_x emissions over North America, for latitude ranging between 25 N–50 N, have been turned off.

$-S_{\text{noligh+noanth}}$

In this simulation, lightning and anthropogenic sources over North America, for latitude ranging between 25 N–50 N, have been turned off.

$-S_{\text{NLDN}}$

We performed another simulation in which the lightning in the GEOS-Chem model for July 2006 are scaled on a daily basis to NLDN and LRLDN observations. Note that, to account for the intracloud fashes (IC), we have applied to the NLDN and LRLDN data a IC:CG ratio of 3, which corresponds to the mean value of this ratio over the USA calculated by Boccippio et al. (2001). We have then gridded the lightning data on the GEOS-Chem grid and taken the daily average. This resulting lightning distribution is used to scale the lightning activity in the model over the United States on a daily basis. The new GEOS-Chem distribution is shown in Fig. 2. The resulting NO_x source totals 0.14 Tg N in July 2006 over United States.

$-S_{\text{lighx2}}$

In this simulation, the NO_x production by flash is increased by 2 in midlatitudes compared to S_{NLDN}. The production of NO_x by flash is around 520 mol/Flash in mean. This value is close to the updated NO_x production per flash of De Caria et al. (2005) of 460 mol/Flash based on the analysis of the STERAO (Stratosphere-Troposphere Experiment-Radiation, Aerosols and Ozone) campaign and the mean value of 500 mol/Flash reported by Ott et al. (2008) from cloud-resolved modeling analyses of midlatitude and subtropical storms constrained by observed flash rates and anvil aircraft observations. Cooper et al. (2006) found that using a production rate of 460 mol/flash in the FLEXPART model provided acceptable comparison with aircraft data from the International Consortium on Atmospheric Transport and Transformation

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(ICARTT). Hudman et al. (2007) showed that with a value of 500 mol/Flash, the model NO_x field agrees better with observations from the ICARTT aircraft campaign over the Eastern part of the United States, but the model was still too low. Assuming larger NO_x production per flash in midlatitudes is also consistent with the analysis of Huntrieser et al. (2008). Indeed, they suggest based on field experiments measurements that the higher vertical wind shear in subtropical and midlatitudes thunderstorms lead to longer stroke lengths and thus higher Nox production per flash than in tropical thunderstorms. The resulting source totals 0.28 Tg N over North America midlatitudes (25–50 N) for July 2006. Note that this value is consistent with Hudman et al. (2007), who simulated a lightning source of 0.45 Tg N for 1 July to 15 August 2004 period when using 500 mol NO/Flash.

$-S_{\text{strat}}$

We performed another baseline simulation in which the treatment of the stratosphere differs from the method used in the original model. In the original version, the ozone is treated by SyNOZ (McLinden et al., 2000). But the ozone mixing ratios are largely underestimated in upper troposphere in midlatitudes in S_{base}. In this version, we use the Li and Shine (1995) monthly mean ozone climatology to prescribe the ozone in the overworld (potential temperature >380 K). Below, in the lowermost stratosphere, the ozone in the model is free to keep consistency between ozone distribution and dynamic field. The comparison between this version and the original version will be shown by comparison with ozonesonde measurements in Sect. 2.5. For lightning, the setting is the same as in S_{NLDN}.

Note that the simulations S_{noligh}, $S_{\text{noligh+noanth}}$, S_{noanth}, and S_{lighx2} start the 1 March 2006. The simulation S_{NLDN} starts the 1 July 2006. The simulation S_{strat} starts the 1 January 2006. All the sensitivity simulations begin with initial concentrations taken from the simulation S_{base}.

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2.4 Evaluation of TES and GEOS-Chem ozone with IONS ozonesondes

TES ozone profiles and GEOS-Chem profiles used in this study have been compared to the ozonesonde measurements from the Intercontinental chemical transport experiment Ozoneseonde Network Study (IONS) 2006 campaign (Thompson et al., 2007a, b, 2008) to provide validation specific to the North American summer conditions. The coincidence criteria for the comparison with sondes are ±3 h and ±200 km, the number of comparisons is 108. Note that for this comparison, the GEOS-Chem profiles and ozonesonde profiles are transformed using the TES observation operator, which is composed of the averaging kernel and the a priori profile using Eq. (1) without the measurement error term $\mathbf{G}\boldsymbol{\varepsilon}$ (Bowman et al., 2006; Worden et al., 2007).

Figure 3 shows the mean ozonesonde profiles, the mean TES profiles, along with the mean GEOS-chem profiles for the different simulations (S_{base}, S_{strat}, S_{lighx2}) and the mean TES apriori profiles. TES data compare better to the ozonesondes than the TES apriori but the ozone in TES is overestimated by 5 to 15 ppbv in the upper troposphere. The overestimation of TES in the upper troposphere is consistent with the previous work of validation of TES using ozonesondes measurements in Worden et al. (2007) and Nassar et al. (2008) as well as with lidar measurements by Richards et al. (2008). We will focus our study at 300 hPa where the bias of TES compared to ozonesondes is about +5 ppbv.

The GEOS-Chem baseline simulation (S_{base}) underestimates ozone mixing ratio by up to 30 ppbv in the upper troposphere. At 300 hPa, the bias is −17 ppbv. A similar underestimation of the GEOS-Chem model in the upper troposphere compared to IONS 2006 was also found by Parrington et al. (2008). In the simulation S_{lighx2}, where NO_x production per flash is increased by a factor of 2 and flash rates are based on NLDN and LRLDN, ozone in GEOS-Chem increases by about 10 ppbv throughout the upper troposphere. This shows that ozone mixing ratios simulated by GEOS-Chem are very sensitive to the lightning NO_x emissions parameterization. This suggests that an underestimation of this source could contribute to the discrepancy between GEOS-Chem

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and measurements (TES and IONS). However, the variation of the bias with altitude suggests also an underestimation of the downward ozone flux from the stratosphere by the model. Indeed, the difference between S_{base} and S_{lighx2} at 120 hPa is zero while the bias is 20 ppbv. In the S_{strat} simulation, with a different treatment of the stratosphere, the bias decreases and ranges now between −15 and −10 ppbv in the upper troposphere. The difference between S_{base} and S_{strat} is around 5–10 ppbv at 300 hPa. To conclude, the differences between GEOS-Chem and measurements (IONS and TES) and the sensitivity simulations suggest that contributions from the stratosphere and/or lightning could be larger than predicted by the GEOS-CHEM model. The present study focuses on the treatment of the lightning source in the GEOS-Chem model using profiles from TES, which have better spatial coverage than sondes, as well as NLDN and LRLDN data and the Hysplit model.

3 Results

3.1 Evidence of lightning NO_x emissions influence in the TES data

3.1.1 Example of the 12 July 2007

Figure 4 shows a case where ozone enhanced layers seen by TES on 12 July 2006 can be related to lightning events. In this figure, the forward trajectories are calculated with the HYSPLIT transport and dispersion model (Draxler and Rolph, 2003) and initialized from locations where the 1×1 gridded and hourly averaged NLDN and LRLDN indicates flashes. The trajectories shown are those that intersect the TES track within ±1 degree latitude, ±1 degree longitude and ±1 h. These trajectories show that the air parcels are trapped in an anticyclone, a typical feature of the upper atmospheric circulation during summer over the US (Li et al., 2005). The ozone retrievals along the TES track are shown in Fig. 4. The intersections between the TES track and the trajectories, indicated by black symbols in Fig. 4, are collocated with an ozone enhancement

layer in the middle and upper troposphere. In this layer, TES observes ozone values up to 100 ppbv. We have calculated the degrees of freedom for signal (DOF), as defined in Rodgers (2000), to quantify TES vertical sensitivity to the atmospheric variability. The DOF for the whole tropospheric profile varies between 1.5 and 1.8 along the TES track as shown in Fig. 5. For the profiles showing the ozone enhanced layers between 25–30 degrees, the middle/upper tropospheric (500 hPa to tropopause) part of the retrievals have a DOF larger than 1. This shows that TES is sensitive to the ozone variability in this region. This is further confirmed by the analysis of the a priori (not shown), which does not present this ozone enhanced layer. For comparisons with TES, the model profiles from the simulation S_{base} are sampled along the Aura orbit track at the observation times, and interpolated onto the TES pressure grid. In order to take into account the vertical sensitivity of each TES retrieval, the model profiles are transformed using the TES observation operator, which is composed of the averaging kernel and the a priori profile and is equivalent to Eq. (1) without the gain matrix (Worden et al., 2007). We find that the GEOS-Chem model also shows the ozone enhanced layer seen by TES. The difference between the GEOS-Chem simulations with and without the lightning source is also shown along the TES track, confirming that lightning NO_x emissions contribute to the ozone enhancement seen by TES. In particular, the model predicts an increase of the ozone up to 16 ppbv due to the lightning emissions. However, the ozone values in the ozone enhanced layer in the model simulation are weaker than in TES by about 25 ppbv. As described in Bowman et al. (2006), differences between TES and GEOS-Chem should be compared to TES reported observational errors (sum of the measurement and cross-state errors estimates). This error is typically of about 5–10 ppbv for the retrievals between 25–30 in latitude in the middle/upper troposphere. This suggests that the difference between TES and GEOS-Chem is significant. The reasons for discrepancies are investigated in Sect. 3.2.

3.1.2 Generalization to North America

We now show results for TES and GEOS-Chem over all of North America. Figure 6 shows a map of the ozone mixing ratios retrieved by TES over the USA averaged over the 250 and 350 hPa vertical domain. Only the measurements taken between the 4 and 18 July 2006 are shown here on the map to avoid the overlapping of symbols. In this map, we have distinguished measurements recently influenced by lightning (large filled dots) and non-recently influenced (small filled dots) according to our analysis using HYSPLIT and flashes from NLDN and LRLDN. For this analysis, "recently influenced by lightning" means that the air masses sampled by TES should have passed through a region of lightning activity less than 5 days before the TES measurements. In Fig. 6, we show as well the corresponding GEOS-Chem (S_{base}) ozone mixing ratios sampled along the TES track and modified as explained in the paragraph above for comparison with TES. We also show the difference between TES and GEOS-Chem and the ozone changes due to the lightning source in the model.

As for the particular case presented previously, these figures show ozone enhancements in TES at intersections with forward trajectories emanating from NLDN and LRLDN flashes, i.e.: downwind of convective events. These ozone enhancements are clearly seen over the Pacific Ocean between 25–30 N, West of Mexico and North West of the Gulf of Mexico, where color gradients between recently influenced by lightning measurements (large filled dots) and non recently influenced by lightning measurements (small filled dots) are observed. In the southeastern United States, the influence of recent convection (as shown by the presence of large filled dots) is observed everywhere. The Fig. 6 also shows that ozone mixing ratios in the upper troposphere in the GEOS-Chem simulation are generally lower and less variable than in TES. The differences TES – GEOS-Chem vary between −10 to +25 ppbv. But again, as for the particular case presented previously, the difference between the GEOS-Chem simulations with and without the lightning source shows that the model generally predicts an ozone enhancement due to lightning at the TES locations where recent influence from

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lightning (large filled dots) is expected according to the analysis of forward trajectories emanating from NLDN and LRLDN observations. This suggests that the model captures the spatial pattern of ozone enhancement due to lightning. Cooper et al. (2007) based on IONS campaign and model simulations find a very similar ozone enhancement pattern due to lightning for summer 2006 (see their Fig. 2c and 3c). This also further confirms the signature of convection and lightning events in the TES data indicating that TES data can be used to investigate lightning emissions over the US in conjunction with the GEOS-Chem model.

However, the last figure in Fig. 6 shows clearly that differences between GEOS-Chem and TES are not only observed in the air masses recently influenced by lightning but also everywhere, in particular at high latitudes, where low influence from lighting is expected. The spatial pattern of the difference between GEOS-Chem and TES suggests that the underestimation of several sources and among them the stratospheric source could potentially contribute to the underestimation of the ozone concentrations in the upper troposphere, as suggested by the analysis of the vertical variation of the difference between GEOS-Chem and sondes (Sect. 2.5). In the following, we focus on the treatment of the lightning source in global models (here GEOS-Chem) using information from TES ozone, NLDN and LRLDN data. More particularly, we test whether improving the lightning distribution and increasing the NO_x production by lightning in the model would reduce the discrepancy between TES and GEOS-Chem.

3.2 Sensitivity studies to the lightning distributions and NO_x production per flash

In this section, we perform two sensitivity studies, one to the lightning distribution and the other one to the value of the NO_x production per flash in the GEOS-Chem model. We compare TES and GEOS-Chem in terms of correlation, biases and centered pattern RMS difference. We have split the data into 6 regions defined by 3 latitude bands and 2 longitude bands because the TES a priori is different in these 6 regions (see Fig. 1). This ensures that the variability in the TES data is from TES rather than from the TES a priori. We also consider only the data for which the DOF between 500-hPA-

tropopause is equal or larger than 1 to ensure that TES has sensitivity.

3.2.1 Sensitivity studies to the lightning distribution over the United States

In S_{base}, the lightning activity in the model is scaled to OTD/LIS climatology on a regional and monthly basis to realistically distribute the 6 Tg N/yr of NO produced by lightning over the globe and throughout the year. This means that the lightning is uniformly scaled over the United States, but the relative distribution of lightning intensities within the United States still relies on Price and Rind (1992) and the convection simulated by GEOS4. However, as demonstrated by Boccippio (2002), the parameterization of Price and Rind (1992) cannot fully represent the regional variability of lightning activity. In addition, several studies pointed out that GEOS4 meteorological dataset has some biases, in particular, cloud top height (Hudman et al., 2007). As a result, the correlation between daily lightning activity values in the S_{base} simulation and observed by NLDN and LRLDN ranges only between 0.3 and 0.5 over the US (not shown). Therefore we test whether imposing the distribution of lightning intensities according to observations can explain part of the discrepancy between model and observations. In the simulation S_{NLDN}, lightning flashes in GEOS-Chem are scaled on a daily basis to the NLDN and LRLDN observations for each particular grid box where lightning is simulated in the GEOS-Chem model (see Sect. 2.2). We observe that the correlation between TES and GEOS-Chem ozone changes only slightly in the S_{NLDN} simulation compared to S_{base} (Fig. 7). This suggests that even though the model does not reproduce the relative lightning intensities observed by NLDN, this cannot account for the difference between TES and GEOS-Chem. Furthermore, we see that the correlation between TES and GEOS-Chem is only slightly sensitive to the presence of NO_x from lightning (S_{noligh}). This leads to look at the correlation between TES and the simulation without surfaces sources (S_{noanth}) as well as the simulation $S_{noligh+noanth}$ for which lightning and anthropogenic sources over North America have been turned off. We see that the correlation is only slightly sensitive to the absence of NO_x from lightning (S_{noligh}) or from surfaces sources (S_{noanth}). But when these 2 sources are omitted ($S_{noligh+noanth}$),

i.e. ozone precursors do not emanate from convective grid box, the model fails capturing the ozone variability exhibited by TES. As seen in Fig. 7, only one of these sources is necessary to reproduce most of the observed variability, as they both have the same spatial distribution, linked to the spatial distribution of the convective activity. Figure 8 presents the probability of having a non-null daily lightning activity in a particular grid box when NLDN observes lightning activity in this same grid box. We see that the model and observations agree on the presence of lightning activity in more than 60% of the time for most of the grid boxes over the USA in July 2006, suggesting that daily pattern of convective activity occurrence is correctly reproduced by the model. This is likely due to the fact that GEOS-Chem runs with assimilated meteorology. To conclude, the model's ability to reproduce the location of the ozone enhancements seen by TES is due to the fact that this model reproduces the pattern of the occurrence of convective events and their associated lightning on a daily basis during the summer of 2006, even though it does not well represent the relative distribution of lightning flash intensities.

3.2.2 Sensitivity studies to the NO_x production per flash

We investigate the sensitivity of our results to the lightning NO_x production/Flash. We use a production of 520 molecules of NO/Flash very close to the one found by DeCaria et al. (2005) (460 mol/Flash) and the mean value found by Ott et al. (2008). This updated value was also used by Hudman et al. (2007) (500 mol/Flash), who found better agreement between the GEOS-Chem NO_x field and measurements from the ICARTT campaign. In our case, the bias between GEOS-Chem and TES is reduced by about 40%, it now ranges between −18 to −6 ppbv, compared to between −22 ppbv to −12 ppbv previously (Fig. 7). This is still larger than the bias between sondes and TES at this level of −5 ppbv (see Fig. 3). This simulation is also compared to IONS ozonesondes. The difference between IONS and GEOS-Chem decreases by 10 ppbv when the production per flash is doubled. However, the correlation between the TES and GEOS-Chem field does not improve. A closer look at scatter plots of TES vs. GEOS-Chem (not shown) indicates that the ozone in the model is scaled up every-

where. As a result, the model now slightly overestimates the low ozone values not influenced by recent lightning emissions. Indeed, with the increased NO_x production/Flash, lightning occurring in midlatitudes in Europe and Asia increase the ozone background in the upper troposphere in the US.

4 Conclusions

We find ozone enhanced layers downwind of lightning flashes in TES data over the United States in summer 2006 using lightning observations from NLDN and the HYSPLIT model. The global chemistry transport model GEOS-Chem with lightning activity calculated with the Price and Rind (1992) parameterization, scaled to OTD/LIS climatology, and with a production of 260 NO mol/Flash captures the ozone enhancements seen by TES and a sensitivity study performed with this model confirms the influence of the NO_x lightning emissions on these enhancements. However, we find that the model generally underestimates these ozone enhancements and the differences between TES and GEOS-Chem are larger than the TES observational errors.

We tested whether this discrepancy between TES and model could be due to a deficiency of the lightning parameterization. Two sensitivities studies, one to the distribution of the lightning source and the other one to the strength of the source, have been performed. We find that imposing the NLDN lightning distribution in the model on a daily basis does not change the location of the ozone enhancements from lightning in the model, and only slightly changes the magnitude of these enhancements as more lightning are observed in 2006 in NLDN than in OTD/LIS climatology. We show that the model's ability to reproduce the location of the enhancements is due to the fact that this model reproduces the pattern of the occurrence of the convective events on a daily basis during the summer of 2006, even though it does not well represent the relative distribution of lightning flash intensities.

We tested whether increasing the NO production per flash would reduce the discrepancy between TES and the GEOS-Chem model. For this test, we used the value of

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520 NO mol/Flash, for lightning occurring in midlatitudes, that better agrees with the values proposed by the recent studies of DeCaria et al.(2005) and Ott et al. (2008). At 300 hPa, the mean bias between TES and the original GEOS-Chem is between −22 and −12 ppbv depending the latitude band. With the increase NO/Flash production, the bias is now between −18 and −6 ppbv. This is still larger than the bias between sondes and TES at this level of −5 ppbv. However, this improvement should be considered with caution for two reasons. Firstly, the ozone in the GEOS-Chem simulations is biased low in the upper troposphere by 15–30 ppbv compared to the ozonesondes with the bias increasing with altitude and latitude, suggesting the stratospheric contribution to tropospheric ozone is underestimated in the model. Secondly, although the bias between TES and GEOS-Chem decreases in the simulation with the increased NO/Flash, the correlation does not improve. One explanation for this is that GEOS-Chem now overestimates the ozone background as compared to TES. Other factors that could be impacting the bias and correlations between TES and GEOS-Chem are deficiencies in the efficiency of the convective transport of pollution (Folkins et al., 2006), or in the emissions of the ozone precursors at the surface in the model. For the latter, it is important to note that NO_x surface sources from NEI99 and soil emissions had not been updated from the more recent estimates (Hudman et al. 2007; Jaegle et al. 2005) in the present study. However this update would likely lead to less ozone production.

Since TES data provide direct measurements of ozone enhanced layers from lightning, they constitute a valuable dataset for characterizing ozone production from lightning NO_x emissions. TES can provide a unique capability to campaigns focused on understanding the influence of convection and lightning on the chemistry of the upper troposphere over the United States, such as the Deep Convective Clouds and Chemistry experiment (DC3) planned for the summer 2011. In this framework, TES ozone profiles could be used to constrain model calculations in conjunction with additional data, particularly NO_x and OH concentrations in the upper troposphere collected during the campaign. This campaign will also involve different chemistry transport models,

including global and regional models, which could help prioritize limitations of the models, as indicated by our findings, in terms of treatment of the stratosphere, convection, and model resolution and their impacts on the interpretation of the data. In particular, given that the IONS sondes reveal stratospheric influences often are important (Thompson et al., 2007a,b, 2008; Yorks et al., 2008) when upper tropospheric ozone is elevated over the summertime US, the treatment of the stratospheric ozone flux in global models merits further investigation.

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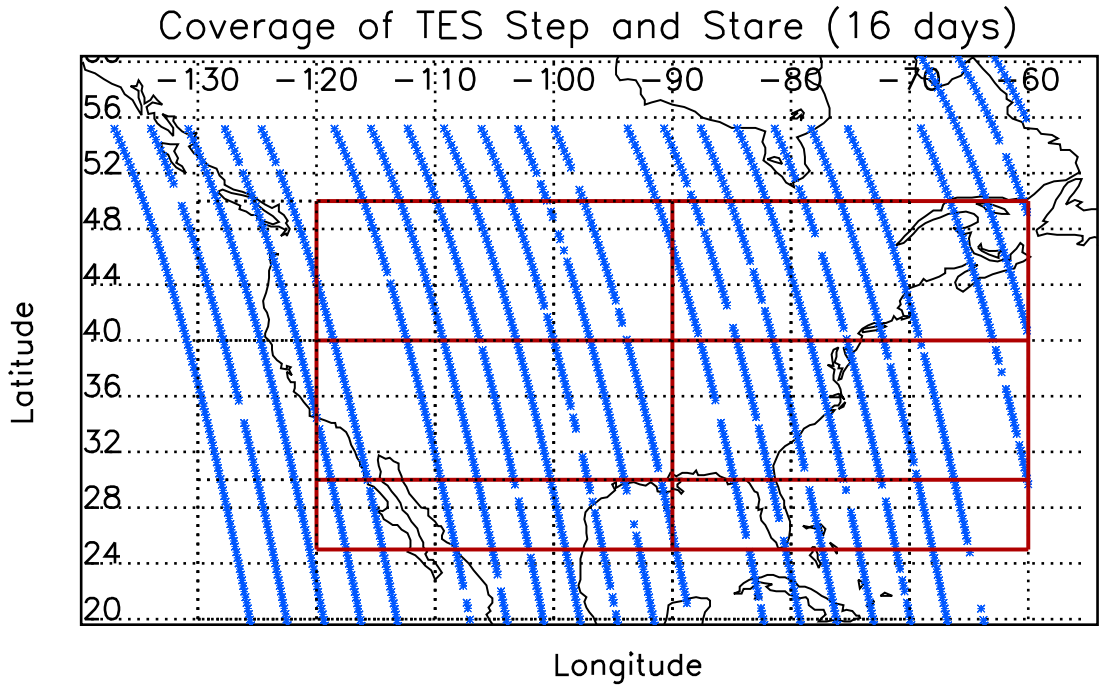

Fig. 1. Coverage of the TES Step and Stare for a 16 days period in July 2006 (blue). The different regions that are distinguished for the statistics in Fig. 7 are presented in red.

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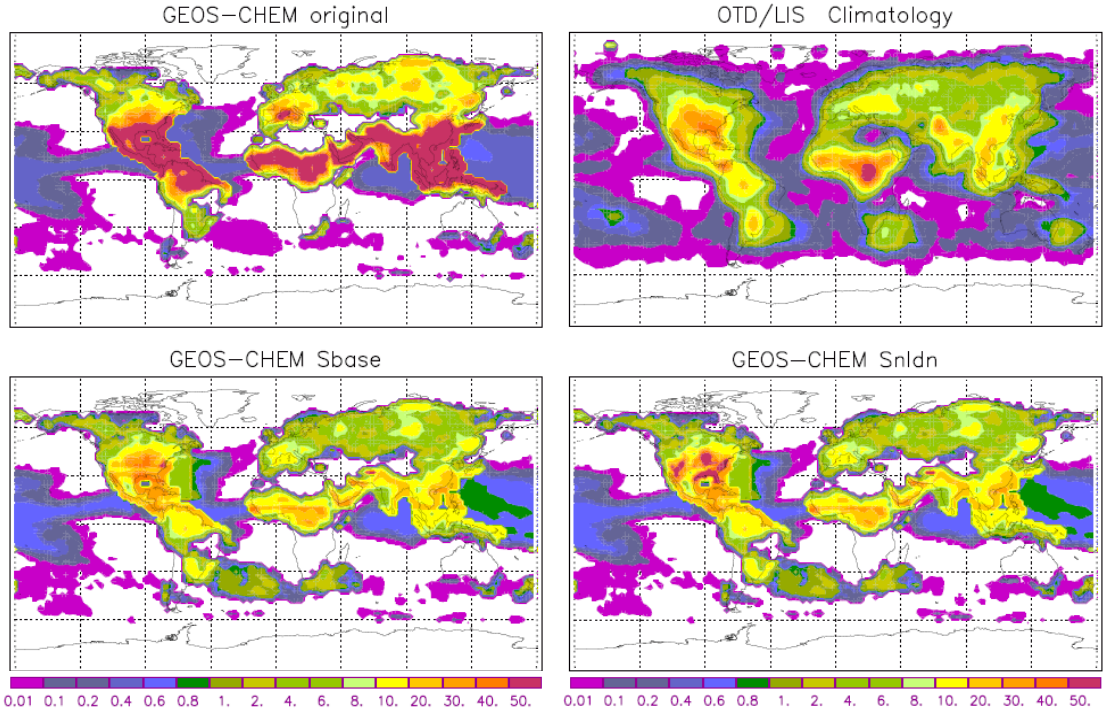

Fig. 2. Top: July Monthly mean Lightning flash densities simulated by GEOS-Chem for 2006 (left) and derived from 5 years of OTD and LIS observations (see text for more details). Bottom: July Monthly mean Lightning flash densities simulated by GEOS-Chem for 2006, when GEOS_Chem is scaled to OTD/LIS on monthly basis simulation Sbase (left) and scaled to NLDN observations on a daily basis (right).

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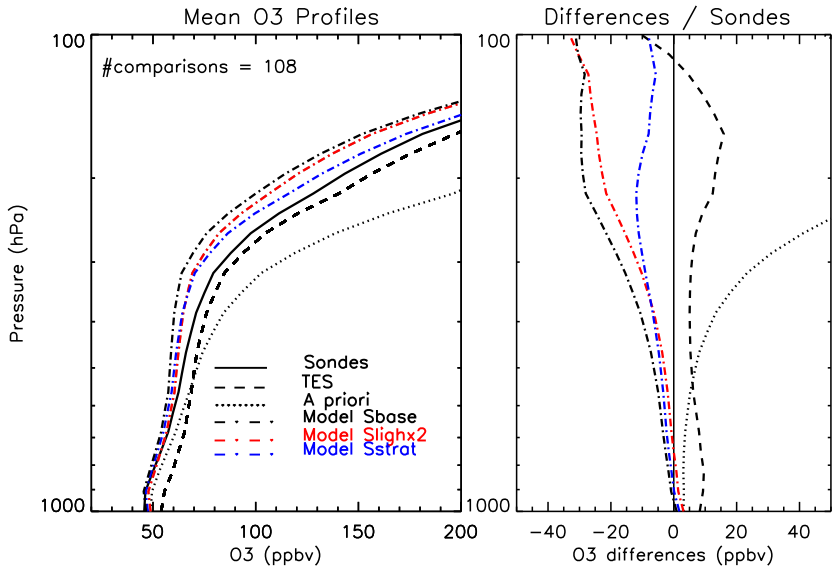

Fig. 3. Left: comparison between the mean ozonesondes profile from the IONS project (black), mean TES profile (black dashed line), mean TES apriori profile (black dotted line) and mean GEOS-chem profiles for the S_{base} simulation (black dashed-dotted line), S_{strat} simulation (green dashed-dotted line), S_{lighx2} simulation (red dashed-dotted line). Right : Mean difference between ozonesondes measurements and respectively TES measurements (black dashed line), mean TES apriori profile (black dotted line), mean GEOS-chem profile for the S_{base} simulation (black dashed-dotted line), S_{strat} simulation (green dashed-dotted line), S_{lighx2} simulation (red dashed-dotted line).

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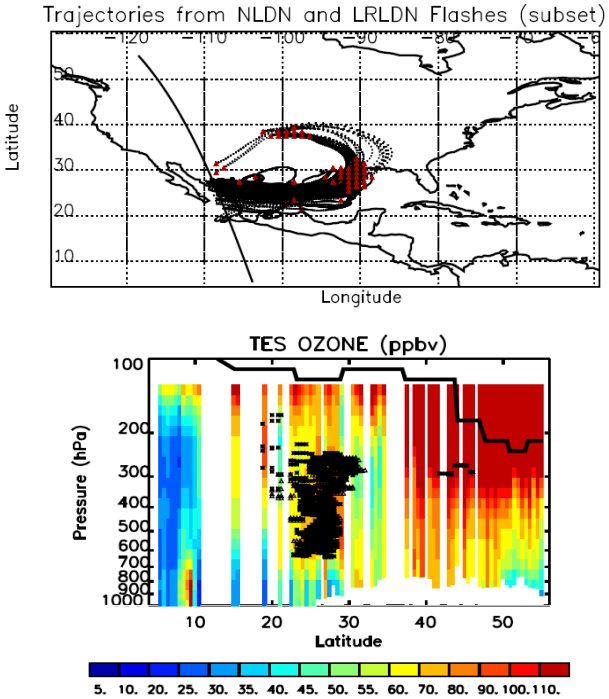

Fig. 4. Top: Five-day forward trajectories initialized from the location and time of the 1×1 gridded and hourly averaged NLDN and LRLDN data and intersecting this particular TES track within ±1 degree latitude, ±1 degree longitude and ±1 h are shown. Bottom: cross section pressure-latitude of mean ozone volume mixing ratio retrieved by TES along the same TES track that shown above. The black symbols represent the trajectories that intersect this particular TES track within ±1 degree latitude, ±1 degree longitude and ±1 h.

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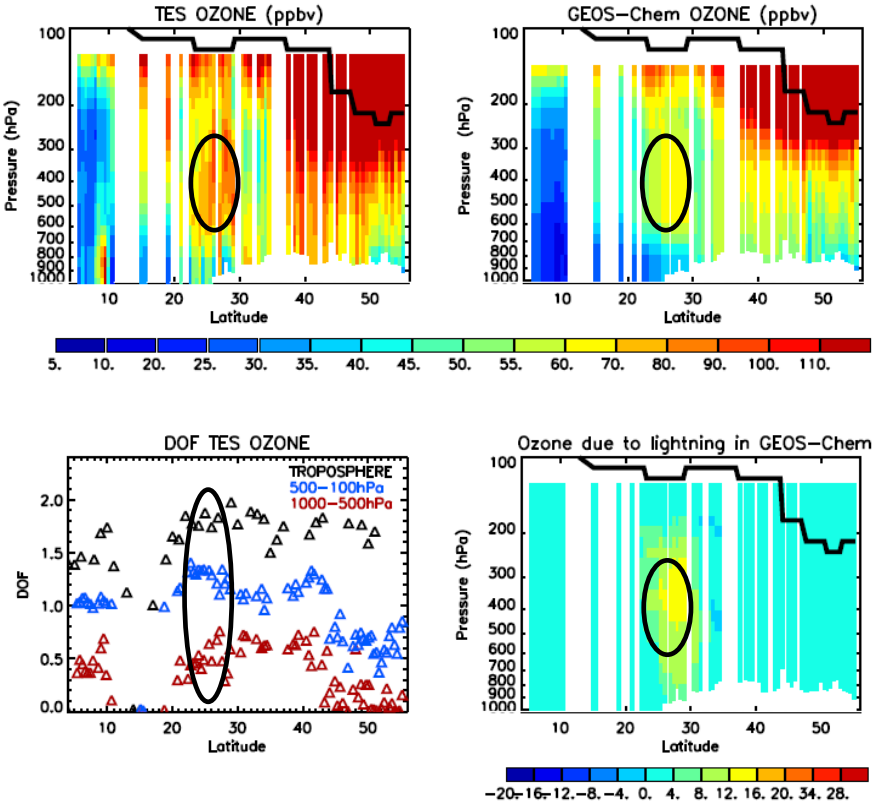

Fig. 5. Top: cross section pressure-latitude of mean ozone volume mixing ratios retrieved by TES (left) and simulated by GEOS-Chem (right) along this particular TES track. Bottom: latitudinal variation of the DOF for TES ozone retrievals for the whole troposphere (black triangle), the surface-500 hPa region (red triangles), the 500 hPa–100 hPa region (blue triangles) for the same TES track (left) and cross section pressure-latitude of the difference in ozone between the simulation S_{base} and $S_{nolight}$ (right). The black circle represent the area downwind of the recent lightning events as show in Fig. 4.

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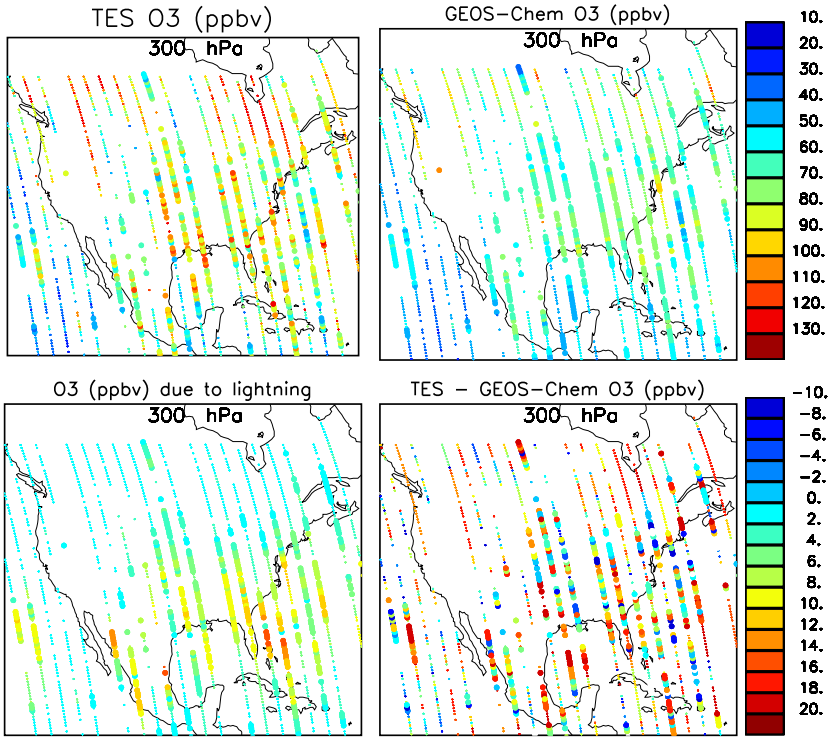

Fig. 6. Top: Ozone mixing ratios over the North America averaged over the 250 and 350 hPa vertical domain retrieved by TES (left) and simulated by GEOS-Chem (right). Bottom: Difference in ozone between the simulation Sbase and Snoligh (left), difference between TES and GEOS-Chem Sbase mean ozone mixing ratios (right). Large filled circles represent the recently influenced by lightning as found by the analysis using HYSPLIT and NLDN and LRLDN. Note that only the measurements taken between the 4 and 18 July 2006 are shown here on the map to avoid the overlapping of the symbols.

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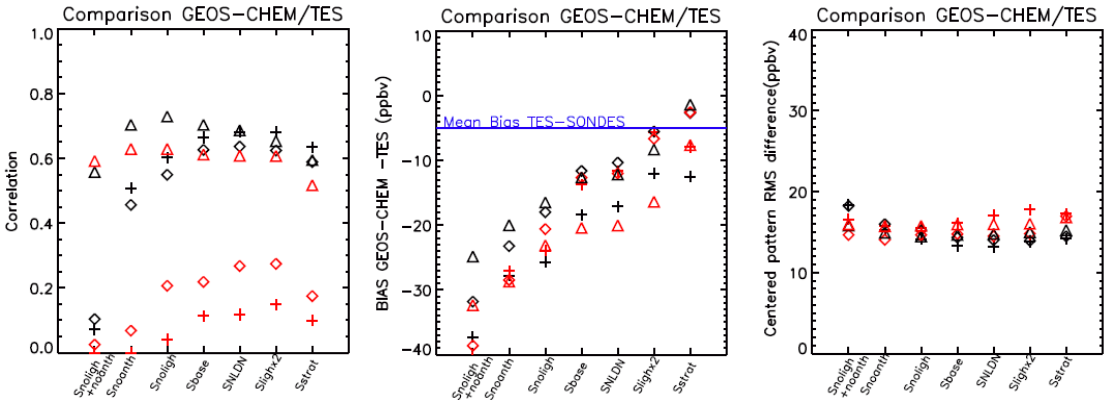

Fig. 7. Comparison between TES ozone and GEOS-Chem ozone at 300 hPa in terms of correlation (left), mean bias (middle), centered pattern RMS difference (right) for 6 different regions and the $S_{\text{noligh+noanth}}$, S_{noligh}, S_{base}, S_{NLDN}, S_{lighx2}, S_{strat} GEOS-Chem simulations (see text for definition of these simulations). The red symbols are for the 60–90 W longitude band, black symbols are for the 120–90 W longitude band. Triangle are for the 40–50 N latitude band, diamonds for the 30–40 N latitude band, plus for the 25–30 N latitude band. Distinction is made between these different regions because TES a priori varies. The different regions are presented in Fig. 1.

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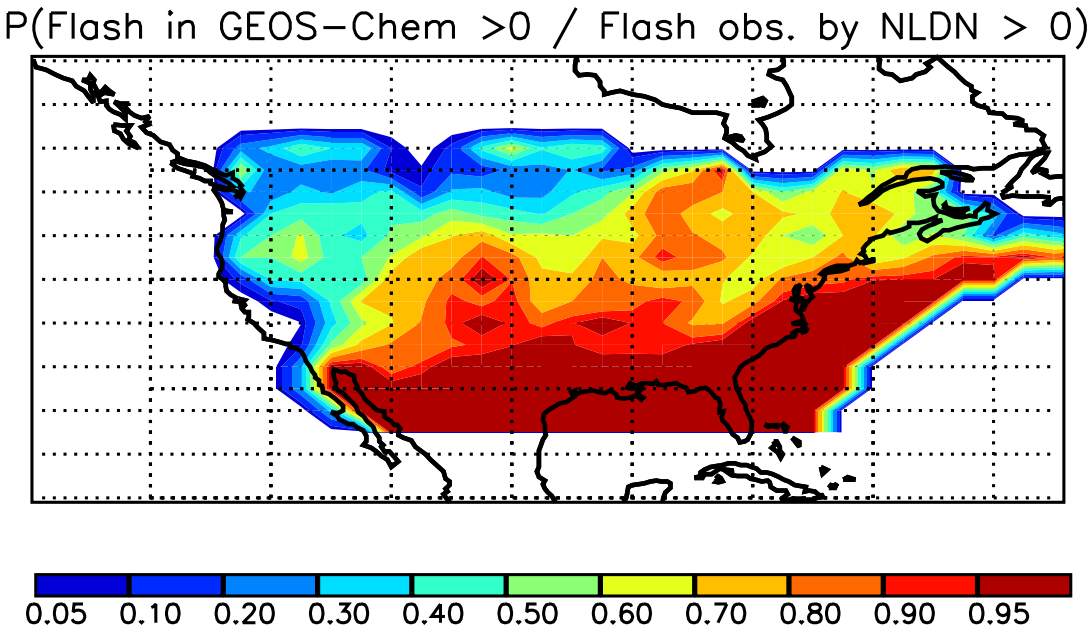

Fig. 8. Probability of having a non null daily lighting activity in the GEOS-Chem model when NLDN observes lightning activity in the same grid box and on the same day. This calculation is for July 2006. NLDN and LRLDN were averaged on the grid box of the GEOS-Chem model.

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