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Interactive Comment

Interactive comment on "The effect of lightning NO_x production on surface ozone in the continental United States" *by* B. Kaynak et al.

B. Kaynak et al.

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First we would like to thank Anonymous Referee 1 for his/her insightful comments and questions.

Summary of the Paper: This paper addresses an important science question that what is the effect of lightning NOx emissions on the surface ozone, in particular if they can affect the distribution of regional high ozone events. The paper is well organized, easy to read, and the team's knowledge in this area is well reflected. The main focus of the analysis presented in the paper is if the addition of lightning NOx production in air quality modeling would affect background ozone leading to a change in the air pollution control strategy. Because of its policy implication, one should consider robustness of the presented results, in particular how sensitive is the results to the model configuration used. It is well known that modeled ozone production by NOx emissions is very





sensitive to the resolution of the model. An excessive ozone production is estimated if the NOx emissions are dispersed in a large grid box initially because biogenic VOCs are already well dispersed. Because physical dimension of lightning is extremely narrow and intermittent, I suspect if the present grid models are capable of quantifying such effects properly.

Even though individual lightning flashes are narrow, during the thunderstorm with hundreds of flashes, the overall affect is more regional. Further, storms generate considerable mixing as well, further dispersing the NOx produced from a single flash. The general horizontal size of convective storms that produce lightning is larger than our grid size of 36 km. Also, as shown by Cohan et al ¹ while the individual sensitivities of ozone to NOx when using a fine grid do vary, the average over the fine grids to a corresponding larger grid, is effectively captured at the larger grid. Additionally, the grid size in this study is still much smaller than global scale models in which lightning emissions are currently included.

Here are the answers to the following specific questions of Reviewer 1 given in italic:

(1) Wouldn't the impact of lightning production of NOx behave more like an addition of NOy species in the atmosphere? That means that direct addition of NO and NO2 into the photochemical system with 36-km grid resolution may interfere with the radical balance expected in the grid model, thus affecting reactivity of the system. NOy can be transported longer distance and also subject to direct deposition (and washout) as well as involving in the heterogeneous reactions.

This question is an extension of the above issue in regards to scale, and the role of having a large number of lightning flashes and increased mixing are still of importance. The addition of NOx to the atmosphere will impact the radical cycling in that area, both in the model system as well as in the actual atmosphere. At issue is how well the model can represent the chemical and physical processes taking place. As noted above, studies show that the sensitivities developed using a coarser grid capture the

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average sensitivities found using a finer (4 km) grid. What the reviewer appears to be saying is that the NOx will be more rapidly converted to other species, presumably PAN and HNO3 due to the larger grid. Formation of PAN in the higher altitudes is more dependent upon the availability of peroxyacetyl radicals. Formation of HNO3 is dependent upon the radical availability. Again, however, we find little difference when doing 4 km simulations, particularly when the source is so spread out like a storm.

(2) Spatial and temporal allocation of cloud and precipitation related process is more an art form than an exact modeling. More often than not, we are forced to implement such process as a phenomenological parameterization than an actual physical processor. Although the authors explained clearly what they have done to distribute the NOx emissions from the satellite-estimated IC/CG lightning information, I see a few issues with the current approach. First, have you checked how well the MM5 simulated cloud, precipitation, and moisture fields correspond to the satellite observed cloud and precipitation? Second, the paper reports that it is rare that the satellite observed IC/CC lightning NOx could not be allocated to the target or adjacent cells. Wouldn't it because a very low threshold value (not described in the paper, but I assume "0" cloud fraction) is used to declare there is no "modeled" cloud to allocate lightning NOx emissions? Because the NOy species will go through heterogeneous processes in air quality models, distribution bias of cloud/precipitation must be accounted for.

One clarification for the possible misunderstanding here on raw lightning data is it is not obtained from a satellite but from a set of ground based radar stations (National Lightning Detection Network). The data only consists of the time and location of GC and does not have observed cloud and precipitation. This is why the cloud information from MM5 and lightning flash locations from NLDN are first compared and then combined to prepare an emissions inventory. We do evaluate how well MM5 does in regards to simulating clouds where lightning is found, as well as winds and precipitation. Considering the size of the convective events, two or more grids likely fall partly inside of the event. 8, S4301-S4305, 2008

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Before this study, lightning emissions are used commonly in global scale models which have significantly lower horizontal resolution than our regional air quality model. A parameterization is used in that case which highly depends on the accuracy of the meteorology model used. In our case, real time measurements are combined with meteorological model outputs. It is expected to have cases when the simulated meteorology does not coincide with the real-time lightning events. Nudging is also performed on MM5 every 3 hours for winds, temperature and humidity. The procedure we used in this study has some uncertainties just like the alternative method used (parameter-ization according to simulated meteorological parameters), but we believe this is the closest approximation we can get with the current information available.

As indicated by the reviewer, this could be an issue for heterogeneous processes in air quality models, but this is an issue that we believe will be minor compared to the greater uncertainties in the assumptions for production of NO from lightning. The model does include heterogeneous reactions of oxidized nitrogen species.

(3) Again, is the vertical resolution '13 layers' enough to represent the effect of lightning NOx emissions on ozone assessment? Can we resolve the different effects of IC/CC discharge (although the same emission rates are used in this paper) vertically with this model resolution?

As indicated in question 2, compared to global scale models which handled the lightning NO process previously, the vertical resolution is quite good. The effect on the ground level ozone is investigated in our study and lower layers have even higher resolution in the model. In addition, the usual configuration of CMAQ of 9 vertical layers is acceptable in most studies, but we used 13 to increase the accuracy throughout the vertical column (there is very little difference when going from 9 to 13 layers). These 13 layers cover the lower 16 km which is higher than the lightning NOx is distributed.

The frequencies for occurrences are different, but the production process is fairly similar in IC and CG flashes. The shape of the vertical profile given in Fig. 2 already takes

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IC/CG discharge occurrences into account.

(4) The paper uses O3-NOz plot to estimate the effects of lightning NOx on the background ozone (Figure 11). The ozone-NOz plot is supposed to represent the number of moles of ozone produced for each mole of NOx reacted. Usually, the background ozone (which varies significantly from place to place) concentrations are initially subtracted from the maximum values in the scatter diagram to represent the ozone production rate. The scatter in Figure 11 is too large to justify estimation of both the offset as the background values and slope to represent the ozone production rates.

We do not have the background ozone concentration information for different places. Obtaining this information adds ambiguity because the background O3 in different places will be different in the base case and lightning case. One alternative is to calculate the ozone production rates from the lightning, which can be derived from (O3 light -O3 base)/(NOz light -NOz base). This approach yields 1.2 ppb increase in background ozone and 5.56 for OPE. This is now included in the text.

1. Cohan, D.S., Y.T. Hu, and A.G. Russell, Dependence of ozone sensitivity analysis on grid resolution. Atmos. Environ., 2006. 40(1): 126-135.

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