Response to Reviewers

ACPD Manuscript doi:10.5194/acpd-10-13029/2010
Title: Interannual variability in soil nitric oxide emissions over the United States as viewed from space

We would like to thank both reviewers for their useful comments and suggestions.

Response to Reviewer #1 (Responses in BOLD, additions to test in *BOLD ITALICS*)

1) The authors do a very good job at comparing the interannual variability of OMI NO_2 columns with that of soil NO_x emissions, lightning, precipitation, temperature. However, I am surprised that they do not show the predicted NO_2 column interannual variability as calculated by the GEOS-Chem model – driven by their soil NO_x emissions. I realize that the model has a lower resolution than the observations, however it seems that by not showing or even discussing this in the paper, the authors miss an important part of the picture. This is particularly important as they go on to examine the impact of the modeled soil NO_x on ozone using the GEOS-Chem model. I thus suggest that the authors include another panel in Figure 3 showing the mean anomalies in column NO_2 calculated with the model.

As suggested, we have added this plot to figure 3a and add the following discussion to Section 4:

"In the GEOS-Chem model, however, the June NO_2 tropospheric column anomalies are incorrectly governed by lightning NO_x emissions. The lightning parameterization currently assumes ~20% of lightning NO_x emissions are placed below 1km [Pickering et al., 1998]. More recent observations, however, suggest most lightning NO_x originates and remains in the middle and upper troposphere, with only a small percentage (~2%) found near the surface [Ott et al., 2010]. Since NO_x is primarily NO at higher altitudes, it does not impact the tropospheric NO_2 anomalies, consistent with OMI anomalies shown in Figure 3a. GEOS-Chem modeled tropospheric NO_2 anomalies for a simulation without lightning are consistent with OMI. This anomaly disappears when S_{NOx} emissions are removed, adding further support to the conclusion that soil emissions are responsible for this variability and demonstrating that variations in transport of urban emissions are not responsible (Figure 3a)."

and

"Figure 3 shows the OMI NO_2 monthly mean columns and deviations from the June 2005-2008 mean for OMI NO_2 columns (standard product), temperature, precipitation, lightning counts, S_{NOx} emissions, and GEOS-Chem NO_2 columns (run without lighting emissions) for regions with mean June OMI NO_2 column > 1.25×10^{15} molec cm^{-2}."

2) Similarly they could add a line indicating the model NO2 column timeseries on figure 5 – or at least discuss how the model compares to the observed column.

We have added this plot to figure 5. Additionally we added the following text to the discussion to Section 5:

"The GEOS-Chem simulation with NARR S_{NOx} predicts a tropospheric NO_2 column mean (standard deviation) of 1.09 x 10^{15} molec cm^2 s^{-1} (0.28 x 10^{15} molec cm^2 s^{-1}), ~ 40% lower than the retrievals (Figure 5). The model captures the peaks soil NO_x pulses and the largest variability in the modeled column is due to S_{NOx}, suggesting that S_{NOx} rather than changes in anthropogenic emissions resulting from increased energy use, or anthropogenic transport leads to the June anomaly and NO_2 peaks."

3) Page 13038 lines 10-13. The authors found that satellite retrievals are affected by the subtraction of the stratospheric component in the vicinity of a storm. This is somewhat worrisome as it seems that some of the observed variability could thus be an artifact. Do they have reason to believe that the DOMINO product it better or worse than the standard product? I suggest that they elaborate on this point in the text.

We agree that some of the observed variability might be aliasing of the stratosphere into the troposphere. However the preponderance of evidence suggests that the main factor governing the variability we describe is soil NO_x emissions. As far as we are aware all other comparisons of the stratospheric aspects of these two retrievals have not found statistically significant differences. Although DOMINO is capable of resolving some large stratospheric intrusions and the standard product is not, we are unsure whether that implies either product represents storm related variations in the stratospheric NO_2 column accurately at the spatial scale of an OMI pixel. Properly looking at this issue would require a global, comprehensive study of storm dynamics and stratospheric intrusions into the troposphere along with detailed comparison of the two retrievals and also one with a much higher resolution model than TM4 a scope of study beyond what we can offer in this paper. We add the following discussion, reiterating this uncertainty and need for validation in the text: The following has been added to Section 3

"Both products begin with the same NO_2 slant column densities, determined using a non-linear least squares fit on the ratio of measured earthshine radiance to solar irradiance spectrums in the 405-465 nm window. Differences in the resulting tropospheric NO_2 vertical column densities arise from differences in the methodology used to calculate the stratospheric component of the slant column and the tropospheric air mass factor, which converts slant column to vertical column densities."

"In Section 5, we show there can also be large differences between the retrievals on daily timescales in the vicinity of storm systems due to differences in the stratospheric

subtraction. For the DOMINO product, the stratospheric NO_2 field is estimated by assimilating NO_2 slant columns into the TM4 chemistry-transport model. For the standard product, the stratospheric NO_2 field is determined by masking regions where tropospheric NO_2 columns are high. The remaining areas are used to generate a smoothed, interpolated stratospheric field using planetary wave-2 analysis in 9° wide zonal bands. A global, comprehensive validation of the stratospheric component vicinity of storms is warranted to reduce uncertainty in the individual retrievals both with respect to soil NO_x and with respect to other studies interested in lightning."

In addition, this point is reiterated in Section 5:

"The S_{NOx} model predicts four S_{NOx} pulses between May-July, each corresponding to peaks in the DOMINO OMI NO_2 column, however not uniformly identified in the standard product retrieval. Our detailed analysis shows the primary difference between the appearance of peaks in these retrievals is a result of the method each uses for stratospheric subtraction and that the differences are more important in the vicinity of storm systems. Future improvements in an analysis of S_{NOx} pulses will require more comprehensive validation of the stratospheric and tropospheric NO_2 products in the vicinity of storms."

4) What is the time period shown in Figure 2?

The time period has been added to the axis of plots and to the caption as follows:

"Simulated contribution of soil NOx emissions to the 2006 monthly means (left) and standard deviations (right)"

and to the text

"Figure 2 shows the soil column is predicted to comprise between 15-40% of the total tropospheric NO_2 column between May-July 2006"

<u>Response to Reviewer #2 (Responses in BOLD)</u>

1) Page 13037 line 6-8 : The same storms that cause extra lightning NOx, can also contribute to a more than typical transport of anthropogenic NO2 to the region of interest. Especially if the wind direction is atypical in the month of interest, this can lead to an extra anthropogenic signal due to transport. The blue spots in Figure 4 at isolated sources of NOx seems to point to this (more NO2 blown out of the city).

The issues referred to by the reviewer are relevant in the near field of large cities and power plants, but do not seem to have a significant effect on the large spatial scale of the anomalies we discuss. To address this concern we have added to the analysis comparisons to GEOS-Chem columns (Figure 3b and Figure 5). The GEOS-Chem model using NARR soil NO_x emissions captures and attributes the

timing of the NO2 peaks (Figure 5) as well as the anomaly in June 2006 (Figure 3b) to soil NO_x. In a model that does represent the transport but has no soil or lightning NO_x there is little variation between June 2006 and the other years. We now articulate this point in the text as follows:

In Section 4 (in combination with #1 from Reviewer 1):

"In the GEOS-Chem model, however, the June NO_2 tropospheric column anomalies are incorrectly governed by lightning NO_x emissions. The lightning parameterization currently assumes ~20% of lightning NO_x emissions are placed below 1km [Pickering et al., 1998]. More recent observations, however, suggest most lightning NO_x originates and remains in the middle and upper troposphere, with only a small percentage (~2%) found near the surface [Ott et al., 2010]. Since NO_x is primarily NO at higher altitudes, it does not impact the tropospheric NO_2 anomalies, consistent with OMI anomalies shown in Figure 3a. GEOS-Chem modeled tropospheric NO_2 anomalies for a simulation without lightning are consistent with OMI. This anomaly disappears when S_{NOx} emissions are removed, adding further support to the conclusion that soil emissions are responsible for this variability and demonstrating that variations in transport of urban emissions are not responsible (Figure 3a)."

And in section 5:

"The GEOS-Chem simulation with NARR S_{NOx} predicts a tropospheric NO_2 column mean (standard deviation) of 1.09 x 10^{15} molec cm^2 s^{-1} (0.28 x 10^{15} molec cm^2 s^{-1}), ~ 40% lower than the retrievals (Figure 5). The model captures the soil NO_x pulses and the largest variability in the modeled column is due to S_{NOx}, suggesting that S_{NOx} rather than changes in anthropogenic emissions resulting from increased energy use or anthropogenic transport leads to the June anomaly and NO_2 peaks."

2) Page 13037 lines 13-20 : As pointed out before by several authors on NO2 monitoring, the fact that June 2006 was warmer than usual can also lead to more use of air conditionings and therefore higher energy usage and more NOx emissions.

Presumably, higher temperatures would be associated with air conditioners powered by powerplant emissions in the region. We looked at the powerplant emission data for 2005-2008 over the Great Plains and found that there has been a decreasing trend in Junes since 2005, so this could not account for the 2006 anomaly anomaly. We address this in the text as follows:

"The GEOS-Chem simulation with NARR S_{NOx} predicts a tropospheric NO_2 column mean (standard deviation) of 1.09 x 10^{15} molec cm^2 s^{-1} (0.28 x 10^{15} molec cm^2 s^{-1}), ~ 40% lower than the retrievals (Figure 5). The model captures the soil NO_x pulses and the largest variability in the modeled column is due to S_{NOx}, suggesting that S_{NOx} rather than changes in anthropogenic emissions resulting from increased energy use or anthropogenic transport leads to the June anomaly and NO_2 peaks.
"

And additionally add the following text to section 4:

"The anomaly is not due to an increase in energy use associated with warmer temperatures. There has been a decreasing trend in powerplant emissions in the month of June over the Great Plains of -4% from 2005-2006, -19% from 2006-2007, and constant from 2007-2008 (data available at ftp://ftp.epa.gov/dmdnload/emissions/)."

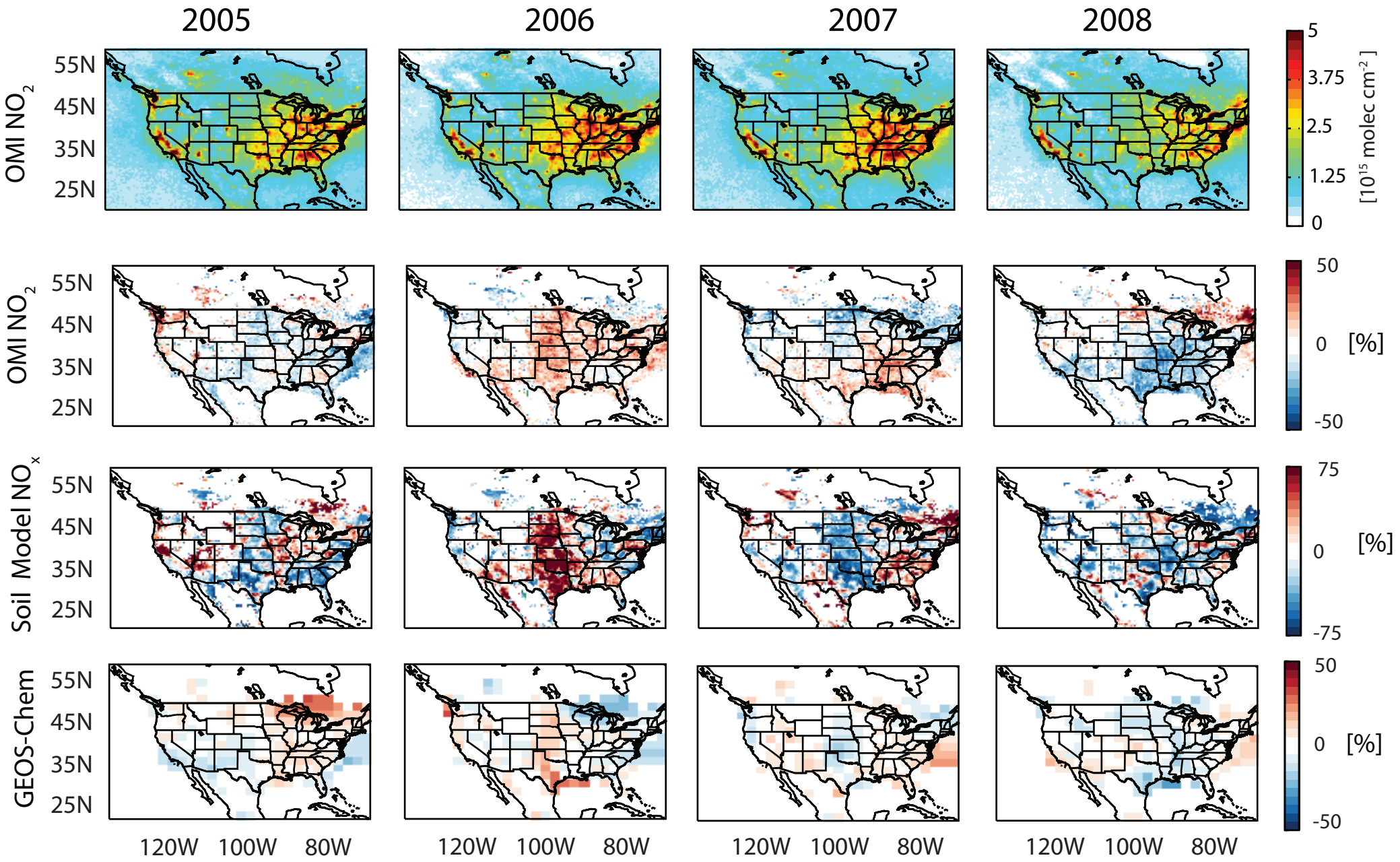

Figure 3a June mean OMI NO_2 column densities (top) are compared to mean anomalies for OMI NO_2 (2nd row) and Soil Model NO_x emission (3rd row), GEOS-Chem NO2 column densities (run without lightning) calculated as difference with June 2005-2008 mean. Only regions with mean June OMI NO_2 column > 1.25×10^{15} molec cm^{-2} are shown in anomaly plots. Color bar saturated at high and low end.

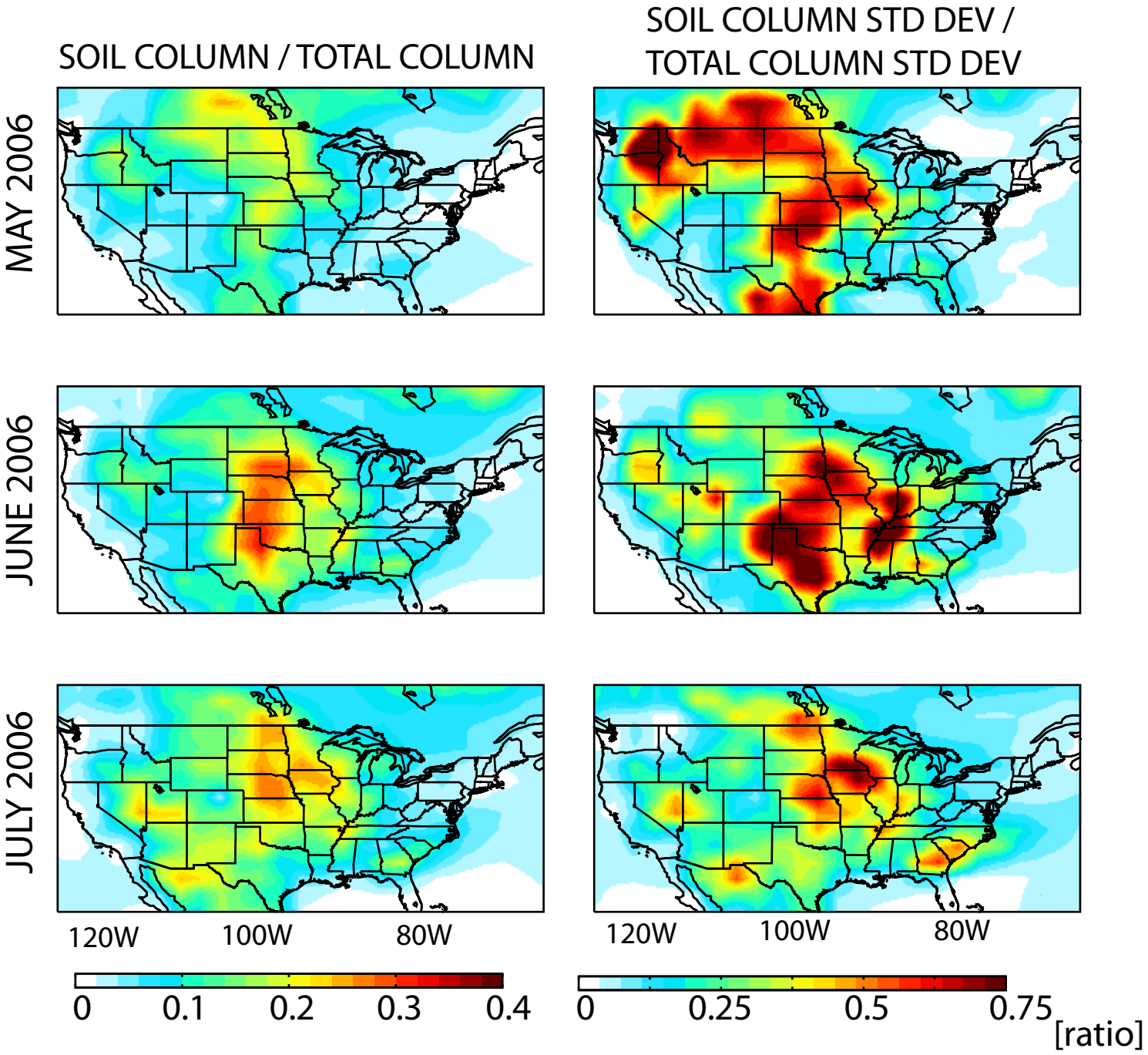

Figure 2 Simulated contribution of soil NO_x emissions to the 2006 monthly mean (left) and standard deviation (right) in tropospheric NO_2 column over the United States using GEOS-Chem. The soil column is defined as the difference in the troposphereic NO_2 column between a simulation with and without soil NO_x emissions over the region 20-65N, 135-70W.

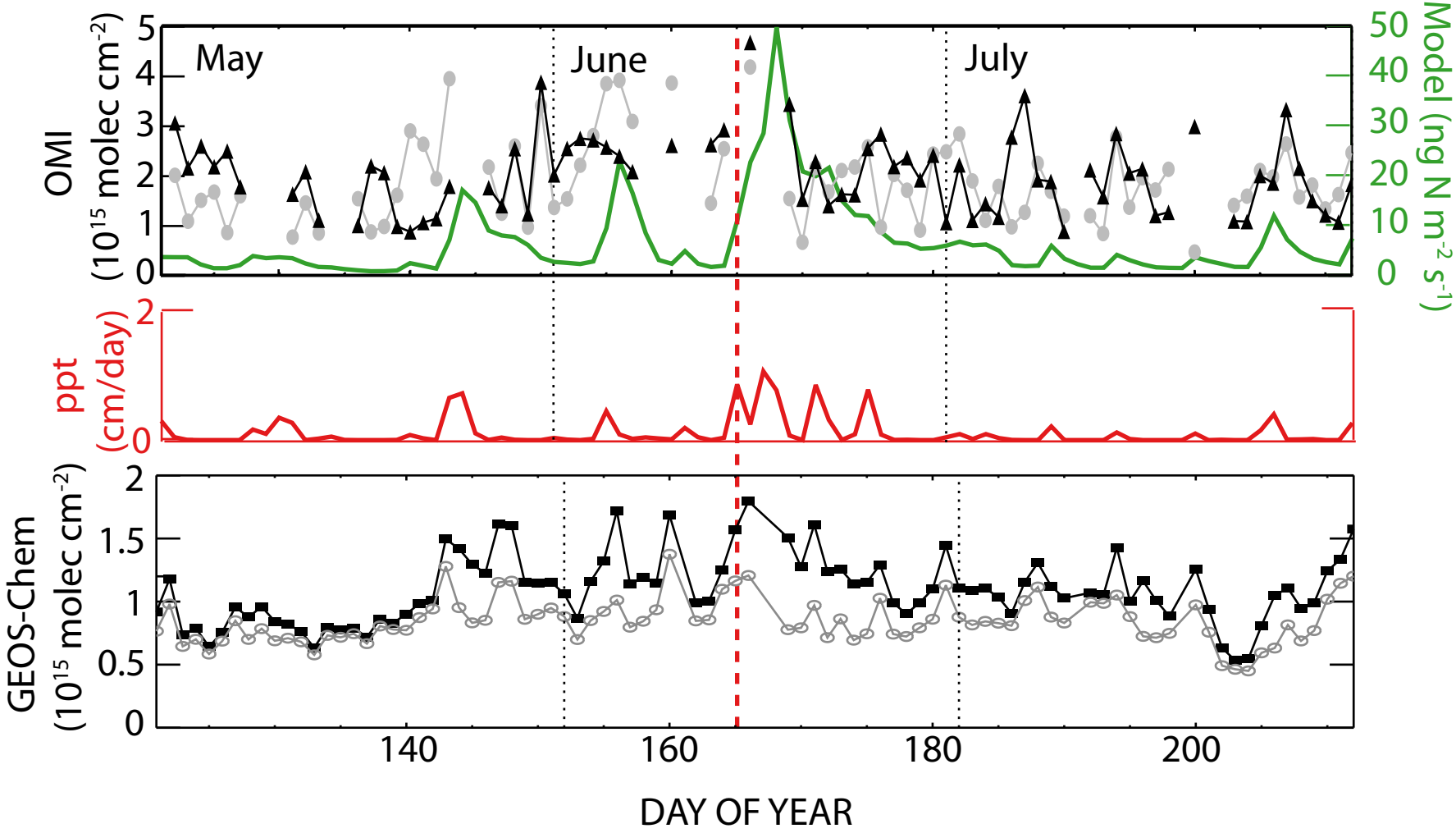

Figure 5 May - July 2006 timeseries of soil NO pulsing events over rural South Dakota (43-45N, 98.75 - 96.25W). OMI NO_2 column densities (top) for the Standard Product (black triangles) and DOMINO Product (grey circles) are compared with predicted soil NO emissions (top, green), precipitation (bottom, red), and GEOS-Chem predicted NO_2 column densities with (black squares) and without (grey open circles) soil NO_x emissions.