Evidence of lightning NO_x and convective transport of pollutants in satellite observations over North America

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Received 7 September 2004; revised 24 November 2004; accepted 17 December 2004; published 19 January 2005.

[1] Column observations of NO_2 by GOME and CO by MOPITT over North America and surrounding oceans for April 2000 are analyzed using a regional chemical transport model. Transient enhancements in these measurements due to lightning NO_x production or convective transport are examined. Evidence is found for lightning enhancements of NO_2 over the continent and western North Atlantic and for convective transport enhancements of CO over the ocean. The two independent satellite measurements show consistent enhancements related to convective events. Model results suggest that the enhancements are particularly large in the lower troposphere due to convective downdrafts of lightning NO_x and shallow convection of CO, implying that low-altitude aircraft in situ observations are potentially critical for evaluating the model simulations and validating satellite observations of these transient features. **Citation:** Choi, Y., Y. Wang, T. Zeng, R. V. Martin, T. P. Kurosu, and K. Chance (2005), Evidence of lightning NO_x and convective transport of pollutants in satellite observations over North America, *Geophys. Res. Lett.*, *32*, L02805, doi:10.1029/2004GL021436.

1. Introduction

[2] Convective outflow is an important pathway for ventilating pollutants from the boundary layer to the free troposphere; subsequent transport of these pollutants has significant ramifications for hemispheric and global air quality. The effects of such processes over North America have been previously investigated using 3-D chemical transport simulations and surface and aircraft observations [e.g., *Thompson et al.*, 1994; *Horowitz et al.*, 1998; *Liang et al.*, 1998; *Park et al.*, 2004; Q. B. Li et al., Outflow pathways for North American pollution in summer: A global three-dimensional model analysis of MODIS and MOPITT observations, submitted to *Journal of Geophysical Research*, 2004, hereinafter referred to as Li et al., submitted manuscript, 2004]. However, in situ observations of convective outflow are limited because of the sporadic nature of convection and aircraft operational difficulties. Recent advancement in satellite observations could potentially provide additional constraints on model simulated convective outflow.

[3] Satellite observations of trace gases and aerosols have been used to detect forest fire plumes [*Thomas et al.*, 1998; *Spichtinger et al.*, 2001; *Lamarque et al.*, 2003]. In comparison, convective outflow is more difficult to detect due in part to cloud interference. Li et al. (submitted manuscript, 2004) showed that despite this interference, satellite observations of CO and aerosol optical depth are useful for mapping convective outflow from North America to the western North Atlantic. In addition, indications were found for lightning activity in the monthly/seasonally averaged NO_2 columns over the tropical Atlantic observed by the Global Ozone Monitoring Experiment (GOME) [*Richter and Burrows*, 2002; *Edwards et al.*, 2003].

[4] In this work, we make use of GOME NO_2 observations and Measurements of Pollution In The Troposphere (MOPITT) observations of CO to evaluate the simulations using a regional chemical transport model (RCTM). Both NO_2 and CO are good chemical tracers for convection. Lightning during convection provides a major source of NO_x ($NO+NO_2$) in the free troposphere [e.g., *Price and Rind*, 1993].

[5] We analyze model simulations and satellite observations for April 2000 because of frequent cyclonegenesis and convective events over North America during the period. The analysis is carried out on a daily basis to emphasize the transient nature of convection. We conduct two model simulations with and without lightning NO_x production and compare these results with GOME observations. Carbon monoxide has much higher concentrations near the surface due to combustion and industrial emissions over North America. To test the effects of convection on CO concentrations, we conduct a sensitivity simulation in which convective transport of CO is turned off and compare the standard and sensitivity simulations in light of MOPITT observations.

2. GOME and MOPITT Retrievals

[6] The retrieval of tropospheric NO_2 columns from GOME measurements and its uncertainty are calculated using the algorithms by *Martin et al.* [2002]. The retrieval uncertainties are due to spectral fitting, spectral artifact from the diffuser plate, the removal of stratospheric column, and air mass factor calculation. The MOPITT CO columns are obtained from the data pool at NASA Langley Atmospheric Science Data Center (ASDC). Only MOPITT retrievals with an a priori fraction <50% were used. The uncertainty of CO columns as reported with the data is about 2×10^{17} molecules cm^{-2} in this work. When

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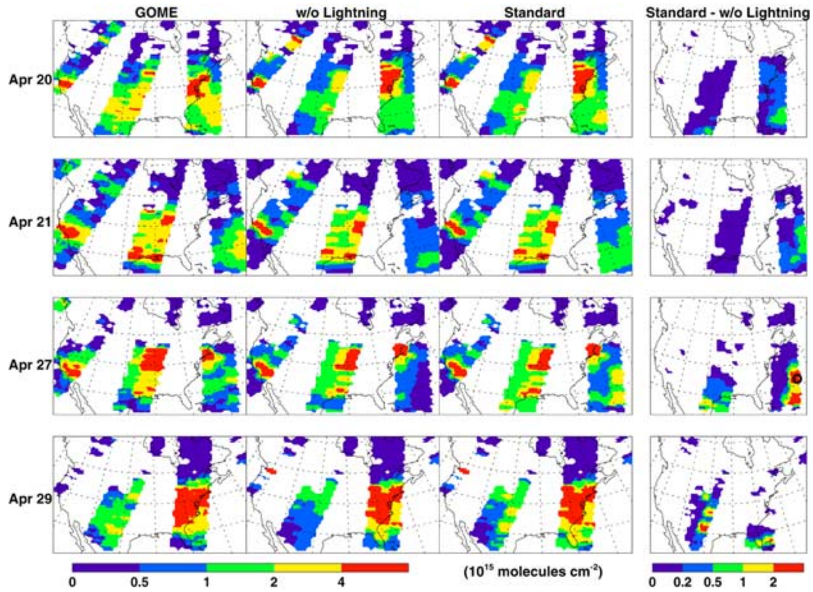

Figure 1. GOME retrieved and the corresponding RCTM simulated tropospheric NO_2 vertical column on April 20, 21, 27, and 29, 2000. GOME columns less than the spectral fitting uncertainties are not included. Simulations with and without lightning NO_x production are shown. The last column shows the simulated lightning NO_2 enhancements.

comparing to the observations, simulated CO results were processed using the MOPITT retrieval averaging kernel described by *Deeter et al.* [2003] and *Emmons et al.* [2004]. The horizontal resolutions of GOME and MOPPIT are 40 × 320 and 22 × 22 km^2, respectively.

3. Model Descriptions

[7] The RCTM has a horizontal resolution of 70 km with 20 vertical layers below 100 hPa. The National Center for Atmospheric Research/Penn State MM5 was used to simulate the meteorological fields using four dimensional data assimilation with the National Center for Environmental Prediction reanalysis, surface, and rawinsonde observations. Most meteorological variables were archived every 30 min except those related to convection, which were archived every 2.5 min. The horizontal domain of MM5 has 5 extra grids beyond that of the RCTM on each side to minimize potential transport anomalies near the boundary. As in the work by *Zeng et al.* [2003], spring 2000 simulations using the global GEOS-CHEM model [*Bey et al.*, 2001] provide the initial and boundary conditions for trace gases. The regional simulations were spun up in the last week of March.

[8] The RCTM was updated from the previous model by *McKeen et al.* [1991]. The transport scheme by *Walcek* [2000] was adopted. Twenty-four chemical tracers describing tropospheric O_3 chemistry [*Bey et al.*, 2001] were transported. The convective scheme by *Grell* [1993] was implemented to be consistent with the meteorological model. The photochemistry module, and the algorithms for dry and wet deposition and emissions from vegetation and soils are adopted from GEOS-CHEM [*Bey et al.*, 2001, and references therein]. Biogenic emissions of hydrocarbons are limited to the regions south of 30°N in April. The monthly mean leaf area index distribution was derived from the Advanced Very High Resolution Radiometer data by *Bonan et al.* [2002]. Emission inventories for combustion and industrial sources were also taken from GEOS-CHEM [*Bey et al.*, 2001] except that fossil fuel NO_x and CO emissions over the United States were taken from the 1999 US Environmental Protection Agency National Emission Inventory. The lightning NO_x algorithm is described in Appendix A. Cloud-to-ground lightning flashes in the model are constrained by the observations from the National Lightning Detection Network (NLDN).

4. Results and Discussion

4.1. Is Lightning NO_x Evident in GOME Observations?

[9] Monthly mean simulated tropospheric NO_2 column compares well with GOME observations (not shown). We find a correlation coefficient of 0.95 with little mean bias (−3%). Our main goal in this work is to determine if transient convection features such as lightning NO_x production can be detected in GOME observations. Large convective or cyclonegenesis events were simulated on April 7–10, 14–16, 18–22, 25–27, and 29–30. Generally we find corresponding NO_2 column enhancements associated with these events. We show 4 specific days of April 20, 21, 27, and 29 (Figure 1).

[10] The model simulations with lightning NO_x are clearly in much better agreement with the observations. All four cases show various degrees of lightning enhancements over the western North Atlantic. The April 29 case also shows significant continental enhancements from western Texas to Kansas. The lightning signals are more difficult to detect over the continent because of surface emissions. The standard model underestimates NO_x concentrations over the western North Atlantic on April 20 and 21 but tends to overestimate on April 27.

[11] The lightning enhancements are 0.5–1 × 10^{15} molecules cm^{-2} on April 20 and 21 and >1 × 10^{15} molecules cm^{-2} on April 27 and 29. Following *Martin et al.* [2002], we estimate the uncertainties of GOME NO_2

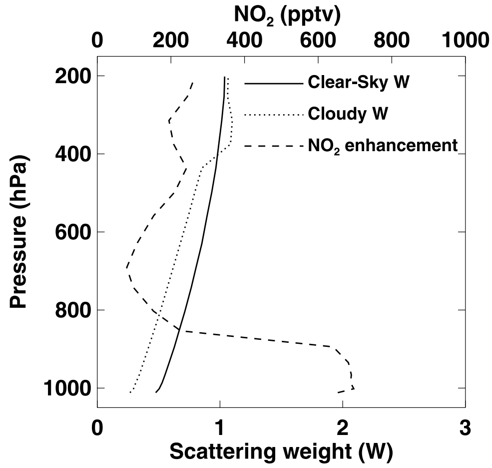

Figure 2. Scattering weights under clear and cloudy sky conditions for the standard model NO_2 simulation on April 27 over the grid box indicated by the black circle in Figure 1. Also shown as a function of pressure is the simulated enhancement of NO_2 due to lightning NO_x production. The scattering weight represents the sensitivity of backscattering radiance measured by GOME to NO_2 concentrations at a given level.

vertical columns to be 50–100% of the lightning enhancements simulated in the model. The relative uncertainties are at the high end for April 20 and 21, when the model underestimates lightning NO_x enhancements over the western North Atlantic. Satellite observations with an improved spatial coverage and lower uncertainty than GOME should provide better quantitative constraints on lightning production of NO_x.

[12] We select a grid box with large lightning NO_x enhancements on April 27 to illustrate the altitude dependence of lightning NO_x contribution to NO_2 column (Figure 2). The NO_2 lightning enhancements are in the lower and upper troposphere, corresponding the "C"-shaped NO_x profile by *Pickering et al.* [1998]. The upper tropospheric NO enhancement is far more prominent than that of NO_2 because the NO/NO_2 ratio increases with decreasing temperature. The large enhancement in the lower troposphere is due to convective downdrafts. The contribution to the column enhancement by NO_2 above 600 hPa is larger than in the lower troposphere (below 850 hPa) in part because of the larger scattering weight at higher altitude (Figure 2).

4.2. Signals of Convective Transport in MOPITT CO Observations

[13] Simulated monthly mean CO column also compares well with MOPITT observations (not shown). We find a linear correlation coefficient of 0.88 with little mean bias (−2%). As in the previous section, we select 3 cases on April 20, 21, and 27 to illustrate the effects of convection on CO column concentrations (Figure 3). The effects are found over the ocean because the only difference between standard and sensitivity simulations is the convective transport of CO. Its effect is to lift CO, emitted from the surface, into higher altitude, where wind speeds are higher than near the surface. As a result, higher-altitude CO lifted by convection is carried over the ocean faster in the more westerly flow compared to that near the surface, creating the enhancements seen in Figure 3. The April 29 case is not shown because the convection is mostly limited to land (Figure 1).

[14] The simulations without convective transport of CO clearly underestimate CO columns over the western North Atlantic. The standard simulations agree much better with the observations. The model tendencies to underestimate CO enhancements on April 20 and 21 but overestimate CO enhancements on April 27 are consistent with the results for lightning NO_x enhancements. The simulated CO column enhancements are above the MOPITT retrieval uncertainty of 2×10^{17} molecules cm^{-2}.

[15] The altitude dependence of the CO enhancement contribution for a selected grid box on April 27 is shown in Figure 4. The profile with convective transport of CO compared to that without is much higher at 300 hPa and 700–980 hPa but is lower near the surface due to the redistribution of CO by convective transport. The 700–980 hPa enhancement due to shallow convection is much larger than that at 300 hPa due to deep convection. This result is consistent with our finding that mass fluxes of

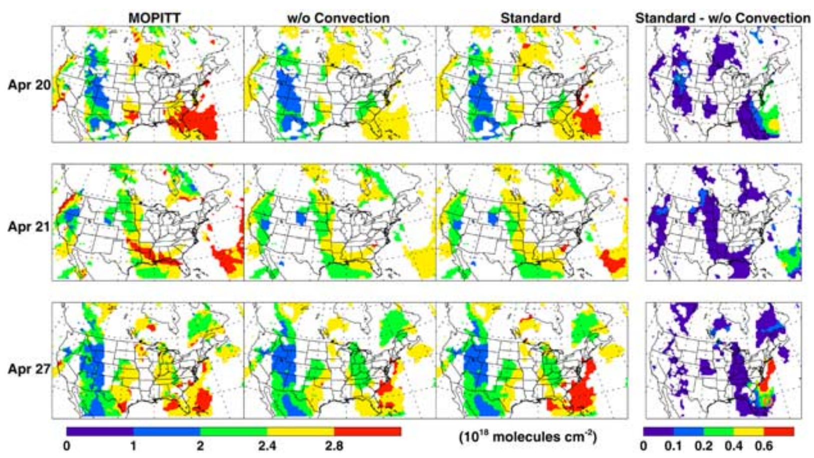

Figure 3. Same as Figure 1 but for MOPITT retrieved and RCTM simulated CO column on April 20, 21, and 27, 2000. The model results with and without convective transport have been processed with the MOPITT averaging kernel.

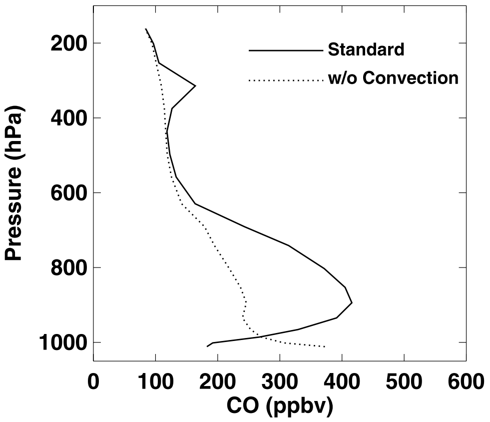

Figure 4. Simulated CO concentrations with and without convective transport as a function of pressure on April 27 over the grid box indicated by the orange circle in Figure 3.

shallow convection are much larger than deep convection in MM5 simulations during this period.

5. Conclusions

[16] Chemical tracer distributions are strongly affected by convective transport and, in the case of NO_x, lightning production. We show that column observations of NO_2 by GOME and CO by MOPITT can be used to identify these transient features when used in combination with 3-D chemical transport model simulations. The two independent measurements show consistent convection related enhancements in terms of geographic location and model bias. While the middle and upper tropospheric contribution from lightning NO_2 to the column enhancements is more important, the major contribution to CO column enhancements is from the lower troposphere. The model results indicate large enhancements in the lower atmosphere of lightning NO_2 (due to convective downdrafts) and transported CO (due to shallow convection), suggesting that low-altitude aircraft in situ observations can potentially provide valuable and critical observations for evaluating model simulations and validating satellite observations.

Appendix A: Lightning NO_x Parameterization

[17] We parameterize the lightning NO_x production rate as a function of meteorological variables so that this emission is consistent with model dynamics. In our work, we experimented with cloud top height [*Price and Rind*, 1993], convective mass flux [*Allen and Pickering*, 2002], and convective available potential energy (CAPE). We found that the parameterization with CAPE produces a similar but better lightning flash distribution than cloud top height when compared with NLDN observations and that CAPE is a better variable for parameterizing lightning flashes than convective mass flux over the southern part of North America and the western Atlantic. To take advantage of the distribution difference between CAPE and convective mass flux, both variables are used in the parameterization (up to the 4th order including cross terms). Two parameterizations are created separately for land and ocean. The intracloud (IC) to cloud-ground (CG) flash ratio is calculated following *Wang et al.* [1998]. We assume that IC and CG flashes have the same energy [*Ott et al.*, 2003]. The rate of NO produced per unit energy is that of *Pickering et al.* [1998]. Lightning NO_x is distributed vertically following the mid latitude profile by *Pickering et al.* [1998].

[18] **Acknowledgments.** We thank Dale Allen and Louisa Emmons for helping us analyze MOPITT CO data, Daniel Jacob and Robert Yantosca for providing GEOS-CHEM model and data, Mian Chin for providing aerosol optical depth data, and Kenneth Cummins for providing the NLDN effective detection efficiency. The GEOS-CHEM model is managed at Harvard University with support from the NASA Atmospheric Chemistry Modeling and Analysis Program. This work was supported by the NSF Atmospheric Chemistry Program and NASA ACMAP program.

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