IMPROVING SOURCES OF STRATOSPHERIC OZONE AND NOy AND EVALUATING UPPER LEVEL TRANSPORT IN CAMx

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1. INTRODUCTION

As the National Ambient Air Quality Standard (NAAQS) for ozone becomes more stringent, understanding regional ozone transport becomes increasingly important. Because their lifetimes are relatively long in the upper troposphere, ozone and a substantial fraction of total oxidized nitrogen (NOy) can be transported for long distances and potentially mixed downward into the planetary boundary layer, where they can influence surface ozone concentrations. It is therefore necessary that regional air quality models used for ozone air quality planning accurately simulate background sources of ozone, including the transport and fate of ozone and NOy in the upper troposphere and lower stratosphere.

Leading up to this work, modeling with the Comprehensive Air quality Model with extensions (CAMx) exhibited under estimates of NOy in the free troposphere above 8 km relative to regionally-averaged aircraft data from the INTEX-A field experiment (ENVIRON, 2012). A similar low bias for NOx in the upper troposphere occurs in other global and regional models (Fang et al., 2010; Hudman et al., 2007; Allen et al., 2012), and the addition of aircraft and lightning NOx emissions reduces this bias but does not eliminate it. Past sensitivity tests performed with CAMx for Texas (ENVIRON, 2013) showed that stratosphere-to-troposphere transport is an important source of upper tropospheric NOy and ozone that must be represented correctly in order to accurately model their respective vertically integrated column masses, which is critical for model/satellite inter-comparison.

Here we describe a set of modeling analyses using CAMx to improve our understanding of the transport of stratospheric ozone and NOy into the troposphere and its effect on surface ozone in Texas during the summer of 2006. We improved the CAMx simulation of the upper troposphere by implementing top boundary conditions derived from a global model, so that explicitly defined stratospheric concentrations of NOy and ozone are advected into the top layer of CAMx. Results were compared against routine ozonesonde data available from four launch sites in the continental U.S. We used the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT; Draxler and Hess, 1997; Draxler and Rolph, 2013) and CAMx models in tandem to study the origin and fate of air transported from the stratosphere to the troposphere. Sensitivity tests evaluated the effect of layer collapsing versus using all layers available from the driving meteorological model.

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2. MODELING APPROACH

We modified an existing CAMx pre-processor to extract time- and space-varying top boundary conditions along with the usual lateral boundary conditions from gridded chemistry fields generated by the GEOS-Chem global chemistry-transport model (Bey et al., 2001). We then modified CAMx v6.1 (ENVIRON, 2014) to read the new top boundary conditions in lieu of the original scheme that internally set top boundary conditions equal to top layer concentrations during the model run (referred to as "zero gradient" method). Figure 1 shows examples of episode-average top boundary conditions for ozone and three NOy species.

CAMx was run for three different configurations using the Texas Commission on Environmental Quality (TCEQ) June 2006 modeling database (TCEQ, 2014). The first two employed the standard 28 vertical layer structure defined by TCEQ. CAMx was run with the original "zero gradient" top boundary conditions, and again with the top boundary conditions extracted from a GEOS-Chem simulation of this period. The third run was identical to the second, except that the CAMx vertical layer structure matched the 38 layers between the surface and ~100 mb from the Weather Research and Forecasting (WRF, Skamarock et al., 2005) model used to supply meteorological input fields. All runs were conducted on a 36/12/4 km nested grid system, where the 4 km grid covered the entirety of

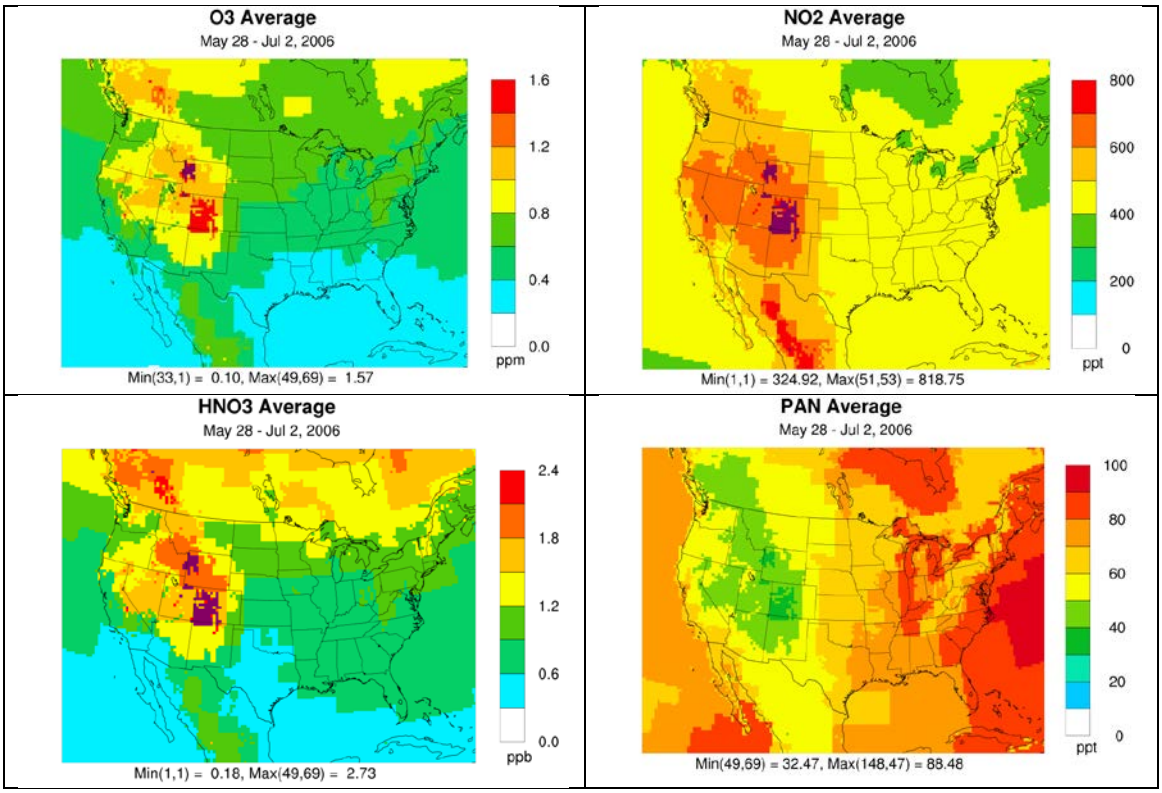

Fig. 1. June 2006 mean spatial distribution of top boundary conditions: ozone (upper left), NO_2 (upper right), HNO_3 (lower left) and PAN (lower right). Concentration units are noted on each plot. Effects from higher model top over elevated terrain as a result of the terrain-following layer structure are evident over the western U.S.

eastern Texas. CAMx surface and aloft model performance for ozone and NOy against rural CASTNET sites (EPA, 2014), ozonesondes and aircraft profiles were compared from all three runs.

All CAMx scenarios were driven by boundary conditions derived from GEOS-Chem version 9-01-03, which was run for the years 2005 and 2006 using meteorology from the Goddard Earth Observing System Model, Version 5 (GEOS-5). The 2005 year was a spinup period and was discarded.

3. RESULTS

3.1 Top Boundary Conditions

According to space- and time-averaged aircraft profiles over North America from the July – August 2004 INTEX-A field study (Singh et al. 2006; 2007), implementation of the new top boundary condition in CAMx improved model performance in the upper troposphere for ozone and NOy species that make up the bulk of the CAMx NOy budget (NO_2, HNO_3 and PAN). The simulation of NO_2 in the upper troposphere is sufficiently improved relative to previous modeling performed for TCEQ that the CAMx model is now ready to be used in column-integrated comparisons with satellite NO_2 column retrievals. Figure 2 compares the average INTEX-A aircraft profile against CAMx profiles averaged over June 2006 and over the 36 km grid shown in Figure 1.

GEOS-Chem and CAMx ozone profiles were compared to measured 2006 ozonesonde profiles at four locations across the U.S (Tarasick et al., 2010): Trinidad Head (coastal California), Boulder (Colorado), Huntsville (Alabama), and Rhode Island. An example of June-averaged ozone profiles at Huntsville is displayed in Figure 3, which shows that GEOS-Chem and CAMx with 38 layers performed quite well, while CAMx with 28 layers exhibited diffusion problems in the upper troposphere for both the original "zero gradient" and GEOS-Chem top boundary conditions.

However, GEOS-Chem's performance in simulating ozone in the upper troposphere and lower stratosphere during the June 2006 episode was inconsistent. For some locations, the model tracked the observed vertical ozone profile very well, but for other locations, GEOS-Chem diverged sharply from the observed profile, and this affected the CAMx simulation of the upper troposphere and

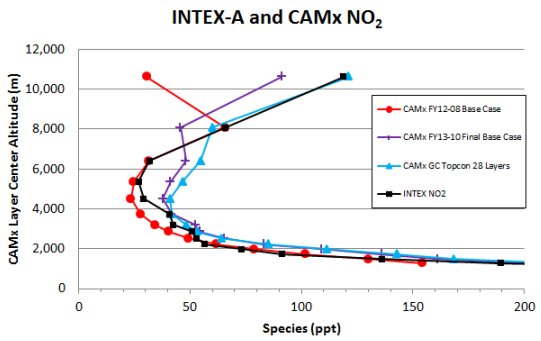

Fig. 2. Comparison of space- and time-averaged NO_2 profiles on the CAMx 28 layer vertical grid structure: July-August 2004 observed INTEX-A fight data (black), June 2006 CAMx using the original "zero gradient" top boundary conditions (red), CAMx with additional sources of NOx including lightning, aircraft, and specified NO_2 top boundary conditions (purple), and CAMx using top boundary conditions from GEOS-Chem (blue).

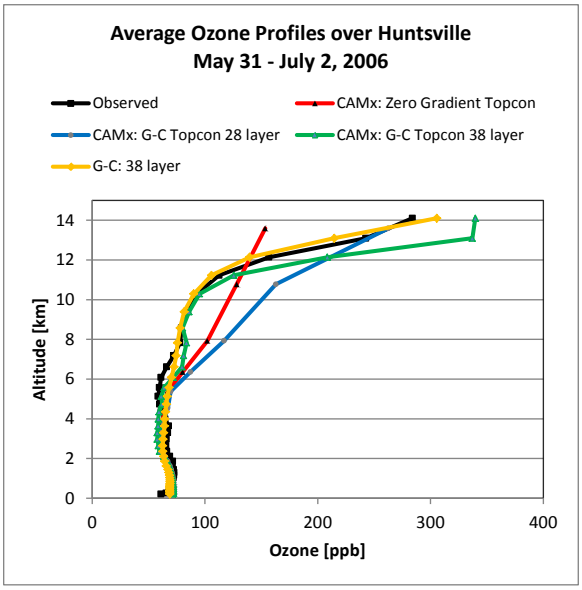

Fig. 3. Comparison of June 2006 average ozone profiles over Huntsville, Alabama: observed (black), GEOS-Chem on the CAMx 38 layer structure (yellow), CAMx "zero gradient" top boundary with 28 layers (red), CAMx GEOS-Chem top boundary with 28 layers (blue), and CAMx GEOS-Chem top boundary with 38 layers (green).

lower stratosphere via the CAMx top boundary condition. The standard version of GEOS-Chem used here has a detailed simulation of tropospheric chemistry but only a simple, linearized treatment of stratospheric chemistry. A newly-available version of GEOS-Chem (Eastham et al., 2014) contains a Unified Chemistry Extension (UCX) that explicitly simulates the chemistry of the stratosphere, providing a more realistic simulation of ozone and NOy variability in the stratosphere as well as stratosphere-troposphere exchange. Use of GEOS-Chem UCX for generating boundary conditions should be investigated to determine whether UCX would provide a better top boundary condition for CAMx than the standard version of GEOS-Chem.

During the June 2006 episode, model performance for surface layer ozone was not greatly affected by the new top boundary condition (not shown). The effects on surface ozone were relatively small and intermittent at low elevation sites, especially in the eastern U.S. At East Texas monitoring sites within the 4 km grid, maximum differences in ground level 1-hour average ozone between the two runs were ~5 ppb. Effects were only slightly more pronounced at high elevation monitoring sites in the western U.S. Meteorological patterns during June 2006 were rather quiescent, which is typical of periods of elevated boundary layer pollution. Greater impacts to surface concentrations from the new top boundary conditions, especially over the intermountain west, would be expected during winter and spring seasons when deeper vertical circulations occur with transient storm systems (Emery et al., 2011, 2012).

Overall, the implementation of the top boundary condition from GEOS-Chem improved the CAMx simulation aloft and we recommend that global modeling results be used to develop both lateral and top boundary conditions for future regulatory modeling.

3.2 Vertical Resolution

Increasing the vertical resolution of CAMx in the upper troposphere also significantly improved the simulation of the upper tropospheric ozone profile. Because the tropopause region is characterized by sharp vertical gradients in ozone, increased model resolution in this region is necessary to simulate ozone accurately and to reduce numerical diffusion. However, the additional vertical resolution in the upper troposphere had only a small and intermittent effect on surface ozone. These findings indicate that increased model resolution is needed for applications that focus on the upper troposphere (e.g. simulation of a stratospheric ozone intrusion). For such applications, the CAMx model should use all available meteorological model layers and not use layer collapsing of meteorological model layers into CAMx layers, as is often done to increase computational efficiency. However, the layer collapsing applied in the TCEQ's June 2006 modeling does

not compromise the CAMx model's ability to reproduce observed surface ozone concentrations and is appropriate for air quality planning applications focused on ground level ozone.

3.3 Transport of Stratospheric Parcels

We identified periods of influx from the stratosphere during the June 2006 episode within the continental modeling grid. Each stratosphere-to-troposphere exchange event was diagnosed using the CAMx vertical velocity field and NOy and ozone concentrations in the top layer of CAMx. For each of these events, we used HYSPLIT back trajectories driven by CAMx three-dimensional winds (with and without layer collapsing) and by external meteorological datasets to determine whether transport from the stratosphere to the troposphere was diagnosed consistently. The external datasets included three-dimensional wind fields from WRF, the Eta Data Assimilation System (EDAS), and a third set that included only the horizontal winds from WRF for which vertical velocity was diagnosed by HYSPLIT (WRF_noVV).

The WRF and CAMx back trajectories were consistent in showing downward vertical motion preceding each stratosphere-to-troposphere transport event. Overall, there was reasonable agreement between the CAMx and WRF trajectories; Figure 4 shows an example set of trajectory comparisons for June 2, 2006. This indicates that CAMx internally diagnoses vertical velocities that are generally consistent with those of WRF for model configurations with and without layer collapsing. There were some days when the CAMx and WRF trajectories diverged more than indicated in Figure 4. The reasons for this are not clear, but these differences were not found to be caused by differences in the WRF and CAMx model vertical structures. Thus, they are more likely related to differences in numerical schemes.

Although the EDAS, WRF and CAMx trajectories were generally similar in showing downward motion preceding the stratosphere-to-troposphere transport events, the EDAS trajectories sometimes showed significant differences in vertical and horizontal motion from the WRF and CAMx trajectories. The reasons for these differences were again not clear and it is not apparent which among the EDAS and WRF/CAMx trajectories are better representations of the actual state of the upper atmosphere on these days.

The WRF_noVV trajectories exhibited significant and unexpected differences in vertical motion from the other trajectories on most days, such as frequent vertical oscillations and rapid rising motion in the stratosphere, contrary to the strong static stability of the stratosphere that inhibits such motion. Figure 4 shows a clear example of this behavior. The HYSPLIT vertical velocity calculation is not described in detail in the model's documentation and the HYSPLIT source code is not publicly available. Therefore, we were not able review how HYSPLIT calculates vertical velocity internally via the default divergence approach. We note that HYSPLIT developers recommend providing HYSPLIT with full three-dimensional wind fields whenever possible.

4. CONCLUSION

Overall, this project marks progress toward understanding the stratospheric contribution to background ozone in Texas and developing the capability to model stratospheric ozone intrusions. In this project, we improved the simulation of the upper troposphere in CAMx via more realistic top boundary conditions and verified that CAMx winds in the upper troposphere are consistent with those of WRF and the EDAS analysis. Recommendations for future work are given below:

- Global modeling should be used to develop both lateral and top boundary conditions for regional regulatory modeling.
- The GEOS-Chem stratospheric chemistry extension UCX should be tested to determine whether it improves CAMx performance in the upper troposphere through more realistic top boundary conditions.
- The HYSPLIT internal diagnostic vertical velocity option should not be used for generating trajectories.

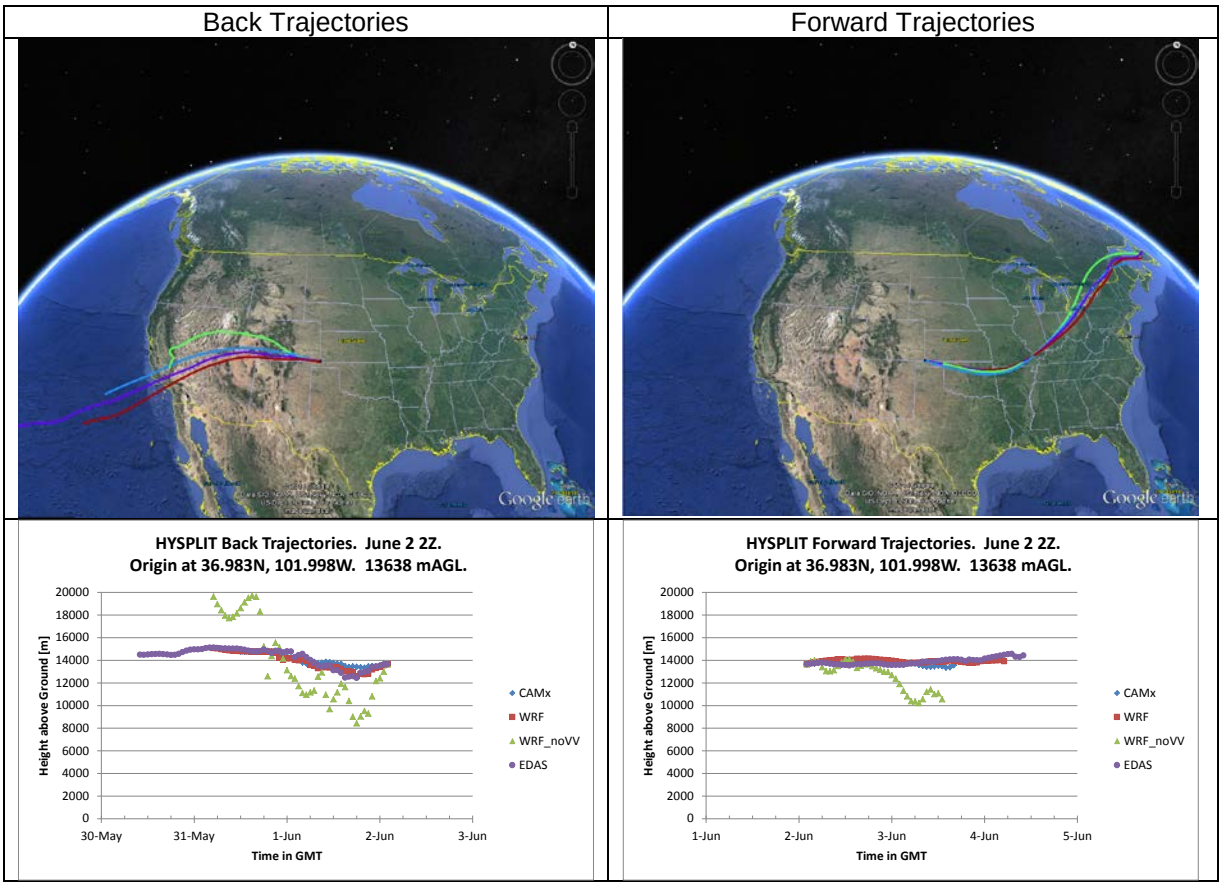

Fig. 4. June 2 0200Z trajectory origin over southeastern Kansas. Trajectory colors: blue = CAMx 28 layer, red = WRF, Green = WRF without vertical velocity input to HYSPLIT, Purple = EDAS. Left panels are back trajectories; right panels are forward trajectories.

8. REFERENCES

Allen, D.J., K.E. Pickering, R.W. Pinder, B.H. Henderson, K.W. Appel, A. Prados, 2012: Impact of lightning-NO on Eastern United States photochemistry during the summer of 2006 as determined using the CMAQ model. *Atmos. Chem. Phys.*, 12, 1737-1758.

Bey, I., D.J. Jacob, R.M. Yantosca, J.A. Logan, B.D. Field, A.M. Fiore, Q. Li, H.Y. Liu, L.J. Mickley, M. Schultz, 2001: Global modeling of tropospheric chemistry with assimilated meteorology: model description and evaluation. *J. Geophys. Res.*, 106, 23073–23096.

Draxler, R.R. and G.D. Hess, 1997: Description of the HYSPLIT_4 modeling system. NOAA Tech. Memo, ERL ARL-224, NOAA Air Resources Laboratory, Silver Spring, MD, 24 pp.

Draxler, R.R. and G.D. Rolph, 2013: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website, NOAA Air Resources Laboratory, College Park, MD, http://ready.arl.noaa.gov/HYSPLIT.php.

Eastham, S., D.K. Weisenstein, S.R.H. Barrett, 2014: Development and evaluation of the unified tropospheric-stratospheric chemistry extension (UCX) for the global chemistry-transport model GEOS-Chem. *Atmos. Env.*, 89, 52-63, http://dx.doi.org/10.1016/j.atmosenv.2014.02.001.

Emery, C., E. Tai, G. Yarwood, R. Morris, 2011: Investigation into approaches to reduce excessive vertical transport over complex terrain in a regional photochemical grid model. *Atmos. Environ.*, 45, 7341-7351, doi:10.1016/j.atmosenv.2011.07.052.

Emery, C., J. Jung, N. Downey, J. Johnson, M. Jimenez, G. Yarwood, R. Morris, 2012: Regional and global modeling estimates of policy relevant background ozone over the United States. *Atmos. Environ.*, 47, 206–217, http://dx.doi.org/10.1016/j.atmosenv.2011.11.012.

ENVIRON, 2012: Evaluating TCEQ NOx Emission Inventories Using Satellite NO_2 Data: Final Report, WO 10365-FY12-08. Prepared for the TCEQ, August.

ENVIRON, 2013: Continuation on Use of Satellite Nitrogen Dioxide (NO_2) Data: Final Report. WO 10365-FY13-10, Prepared for the TCEQ. August.

ENVIRON, 2014: User's Guide, Comprehensive Air Quality Model with Extensions, Version 6.10, http://www.camx.com.

EPA, 2014: Clean Air Status and Trends Network website, http://epa.gov/castnet/javaweb/index.html.

Fang, Y., A.M. Fiore, L.W. Horowitz, H. Levy II, Y. Hu, and A.G. Russell, 2010: Sensitivity of the NOy budget over the United States to anthropogenic and lightning NOx in summer. *J. Geophys. Res.*, 115, D18312.

Hudman, R.C., D.J. Jacob, S. Turquety, E.M. Leibensperger, L.T. Murray, S. Wu, A.B. Gilliland, M. Avery, T.H. Bertram, W. Brune, R.C. Cohen, J.E. Dibb, F.M. Flocke, A. Fried, J. Holloway, J.A. Neuman, R. Orville, A. Perring, X. Ren, G.W. Sachse, H.B. Singh, A. Swanson, P.G. Wooldridge, 2007: Surface and lightning sources of nitrogen oxides over the United States: Magnitudes, chemical evolution, and outflow. *J. Geophys. Res.*, 112, D12S05, doi:10.1029/2006JD007912.

Singh, H.B., W.H. Brune, J.H. Crawford, D.J. Jacob, P.B. Russell, 2006: Overview of the summer 2004 Intercontinental Chemical Transport Experiment-North America (INTEX-A). *J. Geophys. Res.*, 111(D24S01), doi:10.1029/2006JD007905.

Singh, H.B., L. Salas, D. Herlth, R. Kolyer, E. Czech, M. Avery, J.H. Crawford, R.B. Pierce, G.W. Sachse, D.R. Blake, R.C. Cohen, T.H. Bertram, A. Perring, P.J. Wooldridge, J. Dibb, G. Huey, R.C. Hudman, S.Turquety, L.K. Emmons, F. Flocke, Y. Tang, G.R. Carmichael, and L.W. Horowitz, 2007: Reactive nitrogen distribution and partitioning in the North American troposphere and lowermost stratosphere. *J. Geophys. Res.*, 112(D12S04), doi:10.1029/2006JD007664.

Skamarock, W.C., J.B. Klemp, J. Dudhia, D.O. Gill, D.M. Barker, W. Wang, and J.G. Powers, 2005: A description of the Advanced Research WRF Version 2. NCAR Tech. Note NCAR/TN-468+STR, http://www.mmm.ucar.edu/wrf/users/docs/arw_v2.pdf.

Tarasick, D.W., J.J. Jin, V.E. Fioletov, G. Liu, A.M. Thompson, S.J. Oltmans, J. Liu, C.E. Sioris, X. Liu, O.R. Cooper, T. Dann and V. Thouret, 2010: High-resolution tropospheric ozone fields for INTEX and ARCTAS from IONS ozonesondes. *J. Geophys. Res.*, 115, D20301, doi:10.1029/2009JD012918.

TCEQ, 2014: Rider 8 Modeling Database website, http://www.tceq.texas.gov/airquality/airmod/rider8/rider8Modeling.

9. ACKNOWLEDGMENT

This work was sponsored by the Texas Commission on Environmental Quality.