Response to Comments by Anonymous Referee #1

Comment: In this article, the authors describe the hemispheric version of the Community Multiscale Air Quality (CMAQ) modeling system and present a variety of applications of the system for evaluation. In general, the paper is of good quality and should be published with minor revisions as detailed below, although it is a description paper of a new model version and does not contain any truly new science.

Response: We thank the reviewer for the overall positive assessment of our manuscript and for the constructive suggestions for improvements. Detailed below is our response to the specific reviewer comments and the changes incorporated in the revised manuscript. We believe the incorporation of the reviewer suggestions has helped improve the quality of the revised manuscript.

The extension of CMAQ to hemispheric scales required enhancements to both the model's structural and process attributes, to adequately represent the expanded space and time-scales. We do feel that the incorporation, synthesis, and systematic evaluation of these enhancements conveyed in this paper, does represent new modeling science.

Comment: I think the authors should make it clearer in the Introduction that Figure 1 is simply a characterization of the regional-scale CMAQ model and does not necessarily represent the what actually happens in the real atmosphere. CMAQ is known to be rather diffusive (Emery et al. 2011; Garcia-Menendez et al. 2010; Mathur 2008) and probably does not represent this transport very faithfully. Some readers might be fooled into believing that the fractions presented in Figure 1 are realistic, when they are probably not.

Response: Figure 1 illustrates the impact of lateral boundary conditions on the *simulated* free tropospheric and surface concentration variability. The discussion explicitly states that Figure 1 illustrates the influence of "LBC specification on simulated surface-level concentrations across a typical regional modeling domain covering the contiguous U.S.". The subsequent sentences then describe the model tracer species calculations that were used in constructing the Figure. The figure caption also states that these are model simulated concentrations from CMAQ.

The reviewer's somewhat philosophical question on "what happens in the real atmosphere?" speculates on whether this specific model characterization agrees with those from other models and measurements. The results illustrated in Figure 1 agree with the well-established conceptual understanding (built off both measurements and modeling analysis) of long-range transport where in source regions, pollutants within the boundary layer are convectively lofted to the free troposphere where they are intercontinentally transported over long distances by efficient winds and can subsequently be entrained to the surface (subsidence, cloud and boundary layer mixing) in receptor regions thereby regulating "background" concentrations of the receptor region. This conceptual view also implicitly suggests that surface background concentrations are likely influenced by free tropospheric values since that's where transport is more efficient, and that's what the model tracer concentrations suggest. Of course the extrapolation of the illustrated fractions to a specific atmospheric species would also require consideration of specific chemical and physical sinks for that species, and in our discussion of results we have attempted to be deliberate of that distinction. Nevertheless, the relative importance of specification of free-

tropospheric LBCs (representative of long-range pollutant) transport in any model characterization of "background" pollution cannot be diminished, and that's what the figure is intended to illustrate.

We reread the three papers the reviewer brought to our attention. Mathur et al. (2008) analyze the impact of long-range transport of pollution from the Alaskan fires through complementary analysis of CMAQ simulations, in-situ surface and aloft measurements, and satellite retrievals and show impact on surface concentrations at distant sites in the continental U.S. We are not aware of any suggestions of impacts of excessive diffusive transport characteristics in that analysis. Garcia-Menendez et al. (2010) describe the implementation of an adaptive grid methodology in CMAQ to better resolve horizontal concentration gradients in plumes as they undergo lateral transport, but relative to a fixed resolution coarser grid. Their results are applicable to any model that utilizes a uniform discretization, but do not necessarily point to a systematic issue in the CMAQ modeling system. Emery et al. (2011) analyzed vertical transport in the CAMx model, found excessive vertical transport in mountainous regions due to a combination of using a low-order advection scheme, coarse vertical resolution, and likely artifacts of translation of dynamical information between different vertical grid structures employed by the meteorological model and their CTM. Though they did not present any analysis of CMAQ based calculations, the discussion in their manuscript speculates that models such as CMAQ may exhibit similar characteristics. The extrapolation of the Emery et al (2011) analysis with a different modeling system to the current CMAQ tracer results presented here is not straightforward given the differences in model formulations employed in these tests (advection in Emery vs advection, turbulent and cloud transport, dry deposition and wet scavenging here), set-up (different grid resolutions and coupling with meteorological model both in terms of layer configuration and process representation) and since significant changes have occurred in the CMAQ modeling system over the past 6 years. We thus respectfully disagree with the reviewer's assertion that "CMAQ is known to be rather diffusive", but acknowledge that model inferences of impacts of vertical transport from advection, turbulent mixing, and cloud transport are influenced by the choice of horizontal and vertical grid structures, accuracy of numerical methods employed, and consistency in coupling of these aspects with the driving dynamical model (as also discussed in Section 2.1 of our manuscript).

Comment: The results of Figure 3 are fascinating. Increasing the vertical resolution from 35 layers to 44 layers substantially reduced ozone profiles in the lower atmosphere. This immediately begs the question as to what would happen if the number of layers was increased to 60 or 70! A pet peeve of this reviewer is that air quality and atmospheric chemistry models are not rigorously evaluated as numerical models. In any basic numerical modeling class, one is taught to increase grid resolutions until the solution converges to a consistent result. This is _never_ done in 3-D atmospheric chemistry modeling! How much different would the results be if this simple numerical procedure was carried out? My guess is quite different.

Response: The reviewer raises a pertinent issue on the optimal vertical grid structure that should be employed in atmospheric chemistry models. As indicated by Figure 3, the associated discussion in Section 2.1, and our response to the previous comment, more attention needs to be devoted to the vertical grid structure employed by the models. The 44-layer structure depicted in Figure 2 was judiciously designed to provide higher resolution above the boundary layer (nominally at altitudes

> 2km) and near the tropopause to resolve the sharp gradients in O_3 mixing ratios. Clearly, employing 60 or 70 layers will improve the resolution further, but greater impacts will be seen if the additional layers are deployed near the tropopause. We whole heartedly agree with the reviewer that rigorous modeling protocols need to be developed to define appropriate layer configurations and vertical and horizontal grid resolutions for specific model applications, and numerical convergence tests that establish the order of accuracy of the models be conducted on a more routine basis. Pragmatically, this is a challenging endeavor as it is not only application-dependent (for instance, an optimal layer configuration designed specifically for resolving stratosphere-troposphere exchange cases may be different from one for resolving boundary layer venting) and also because some parameters (e.g., mixing height, definition of cloud base and top, representation of wind-shear) depend on the discrete layer structure. We echo the reviewer's concerns in the Summary and Concluding Remarks section by re-emphasizing the need for additional model sensitivity simulations to assess the impact of different "model vertical extent and vertical grid resolution" (see page 21).

Comment: In the description of the model, the authors in several cases describe what is in the version being presented in this paper, but also describe improvements that are or have been worked on. Examples of this include ... (i) seven NTR species rather than 1; (ii) the marine environment chemistry and deposition; (iii) windblown dust parameterization; and, (iv) the ozone-PV parameterization. In these discussions, it's not always clear what's included in this model version used in this paper and what's just an "advertisement" of the improvements to come in the future.

Response: Since the manuscript describes the extension of the CMAQ modeling system from the traditional regional scale to the expanded hemispheric scale, we have outlined the process representations in CMAQ and how they were enhanced for the extended space and time scales for hemispheric applications. This manuscript is an overview of a modeling system that has evolved over several years, so results are presented from model applications that spanned the development period and contributed to the extension of the CMAQ system. For instance, the initial constant PV-scaling approach helped in the conduct of the multi-decadal simulations. The analysis and availability of the fields from these simulations then enabled the development of a more robust space and time varying PV-scaling parameterization. Similarly, the analysis of NTR species and the initial simple approach to modulate its atmospheric lifetime prompted the investigation of the expanded NTR scheme as well as examination of the RACM2 mechanism.

We agree with the reviewer that it is important to clearly convey what process state is being examined and how the model configurations across the different applications may differ. To address the reviewer's concern we reviewed the descriptions of the model configuration in each section to ensure that the model/process state used in the specific runs is stated clearly.

Comment: For Figure 6c, the authors make the dubious statement ... "The comparisons in Figure 6c further show that CMAQ captures the SO4 enhancements in the free troposphere associated

with this episodic event." In looking at this figure, I find it very hard to not laugh out loud when reading this sentence! The observed and modeled SO4 values are of the same general magnitude, but don't seem to be correlated at ALL. I think the authors should be more truthful in their comments about this figure.

Response: The discussion was intended to convey the elevated SO_4^{2-} concentrations at 4-6 km altitudes measured by the C-130 aircraft and previously attributed to long-range transport from Asia. The modeled transport patterns in Figure 6b also suggest the influence of long-range transport. We agree that the model does not do a good job at capturing the space-time variability indicated in the measurements; the coarse (108 km) grid resolution is a likely contributor to such discrepancies. To address the reviewer's concern, in the revised manuscript (Page 13; lines 9-12) we have reworded the discussion as follows: "As illustrated in Figure 6c, SO_4^{2-} levels >1 $\mu g\ m^{-3}$ were often measured in the free troposphere. Both the observations and model show these enhanced SO_4^{2-} levels at altitudes of 4-6 km, which in conjunction with the large scale simulated SO_4^{2-} distributions in Figure 6b suggest that CMAQ captures the SO_4^{2-} enhancements in the free troposphere associated with this episodic event. Some discrepancies in space-time matched model and observed concentrations are also apparent in Figure 6c, which likely result from the relatively coarse (108 km) horizontal grid resolution employed in the model calculations".

Comment: It is admirable that the authors have implemented the RACM2 chemical mechanism into the hemispheric version of CMAQ, which is likely more suitable than CB05 for larger domain applications. However, the authors should give thought to taking the next step and implementing a mechanism that is even more applicable for domains containing regions remote from major sources. Both CB05 and RACM2 were designed for regional-scale applications where NOx concentrations are relatively large compared to values found in the remote troposphere.

Response: We agree with the reviewer that representation of chemistry of the remote troposphere is an important area of future research. Further evaluation of the current mechanisms options in CMAQ for the remote troposphere is an area of current and future research. In the manuscript discussions we acknowledge the need to represent the chemistry of longer-lived species (e.g., acetone) that are important for chemistry of the upper troposphere (page 21; lines 21-25) and also additional analyses of NO_y partitioning and HO_x predictions from the current and any additional chemical mechanism (Page 16; lines 3-4).