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Interactive comment on “Improved simulation of tropospheric ozone by a global-multi-regional two-way coupling model system” by Y.-Y. Yan et al.

Anonymous Referee #2

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Major Comments

This manuscript presents comparisons of a global-to-regional nested version of the GEOS-Chem model with a suite of in situ and satellite observations for the year 2009. The authors claim that due to the “coupling” effects, the two-way nested modeling system significantly improves simulations of tropospheric ozone and related tracers upon the global model alone. While the efforts to build a new model should be appreciated, it is not clear whether there are in fact any improvements from the so-called two-way “coupling effects”, based on the analysis presented in the current manuscript. I agree with the other referee that the revised article will fit better in GMD (as opposed to ACP) if the authors could appropriately address the following concerns:

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1. Improved simulation of ground-level ozone by the nested model over polluted regions in US and Europe is mainly due to the effect of higher-resolution emissions (Figures 1 to 3), which leads to greater NO_x titration than simulated in the coarse-resolution model, as evidenced by the most prominent reduction of ozone biases occurring during the cold months (Figures 5 and 6). These improvements have nothing to do with the effect of two-way coupling that the authors claimed in the abstract and throughout the manuscript. Models with emissions data at a higher horizontal resolution are expected to better resolve the urban-to-rural chemical regimes. In fact, resolving the urban-to-rural emission contrasts and associated chemical processes that the authors discussed in the manuscript can be easily achieved with a regional-scale air quality model at very high horizontal resolution (e.g. 12x12 or 36x36 km²). Why do you think we need to “couple” a global model with a regional model to better simulate these resolution-dependent processes in particular (Lines ~25, Page 25791 and Lines ~5, Page 25791)?

2. Summertime ozone biases in the eastern US and Europe (Lines 15-20, Page 25791 + discussions on Figure 6)

While increasing model resolutions can certainly solve some problems, it does not solve all. The positive ozone biases at northern mid-latitude regions found previously are most prominent during summer (e.g. Fiore et al., 2009 and HTAP 2010). I don't believe the referenced papers have claimed that the limitation in model resolution is a major contributor to the ozone biases. In fact, Figure 6 in the manuscript shows that increasing model resolution show little improvements for the simulation of summertime ozone, consistent with previous findings by Zhang L. et al. (2011, AE). There may be some fundamental, non-resolution-dependent, chemical processes we don't understand well, such as isoprene nitrate chemistry, as discussed in Fiore et al. (2015, AE).

A.M. Fiore, J.T. Oberman, M.Y. Lin, L. Zhang, O.E. Clifton, D.J. Jacob, V. Naik, L.W. Horowitz, J.P. Pinto: Estimating North American background ozone in U.S. surface air

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with two independent global models: Variability, uncertainties, and recommendations, *Atmos. Environ.*, 96, 284-300, 2014, doi:10.1016/j.atmosenv.2014.07.045

I believe many statements in the current manuscript overreach the benefits of increasing model resolution, with very vague discussions on the physical processes. I'd suggest that the authors rephrase the discussions; clearly stating what processes in what season can be better simulated by increasing model resolution and what processes in what season cannot, not only in the main text, but also in the abstract.

3. Simulations of ozone over China (Fig.3c)

The difference in ground-level ozone between the nested model and the global model alone is much larger over China than over US and Europe. But there are no discussions on how well the model simulation of surface ozone over Asia compares with observations. While ozone observations over China are sparse, there are some measurements published in the literature (e.g. Lin An, Mt. Tai, Mt. Hua, and Mt. Huang, Miyun etc.). These measurements have been previously used to evaluate global and regional models (see the references below). A major finding from these papers is that surface ozone over central eastern China peaks in May-June before the onset of the Asian summer monsoon. Can the models presented in the present manuscript realistically simulate observed ozone levels over China and its seasonality (incl. the May-June peak)?

ACP - Special issue The Mount Tai Experiment 2006 (MTX2006) http://www.atmos-chem-phys.net/special_issue147.html

Wang, Y., McElroy, M. B., Munger, J. W., Hao, J., Ma, H., Nielsen, C. P., and Chen, Y.: Variations of O₃ and CO in summertime at a rural site near Beijing, *Atmos. Chem. Phys.*, 8(21), 6355–6363, 2008

Lin, M., T. Holloway, T. Oki, D.G. Streets, and A. Richter: Multi-scale model analysis of boundary layer ozone over East Asia. *Atmos. Chem. and Phys.*, 9, 3277-3301, 2009

Wang, Y., Zhang, Y., Hao, J., and Luo, M.: Seasonal and spatial variability of sur-

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face ozone over China: contributions from background and domestic pollution, *Atmos. Chem. Phys.*, 11, 3511-3525, doi:10.5194/acp-11-3511-2011, 2011.

4. Section 5.2 and Figures 8 and 9: Tropospheric Ozone Profiles

It is not clear, based on the analysis shown, whether lower ozone in the free troposphere simulated in the nested model are due to reduced transport from the pollution source region or from stratosphere-to-troposphere transport (STT). Both models underestimate the ozone variability throughout the troposphere by a factor of 2-3, as inferred by comparison of observed versus simulated ozone standard deviation (horizontal bars in Figures 8 and 9), indicating that the underlying processes controlling tropospheric ozone variability are poorly represented in GEOS-Chem regardless of model resolution. The referee is surprised that this is not even discussed in the current manuscript. More in-depth analysis and discussions are required.

5. Section 5.3 and Figure 10: Comparison of tropospheric column ozone:

It is not clear whether the averaging kernels of the OMI/MLS or OMI/LIU ozone products have been applied to the model results. The averaging kernels of the satellite retrievals must be applied to the model results to enable an apples-to-apples comparison.

6. Simulations of Stratospheric ozone intrusions?

One of the key resolution-dependent processes that the authors do not explicitly discuss is stratospheric ozone intrusions, which are known to have filamentary structures in satellite water vapor imagery, ozone lidar, and ozonesonde measurements (e.g. Appenzeller et al., 1992). Models with higher resolution typically better simulate the vertical structure and intensity of deep stratospheric intrusions (e.g. Roelofs et al, 2003; Lin et al., 2012; Lin et al., 2015).

In the current manuscript, the authors mentioned at a couple of places about vertical transport, but it is not clear what vertical transport processes they are talking about. Increasing model resolution does not necessarily lead to reduced ozone in the free tropo-

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sphere. It can also increase the strength of ozone transported from the stratosphere or pollution transported from the boundary layer by storms, especially for episodic events.

Appenzeller, C., and H. C. Davies (1992), Structure of stratospheric intrusions into the troposphere, *Nature*, 358 (6387), 570–572, doi:10.1038/358570a0

Roelofs, G. J., et al. (2003), Intercomparison of tropospheric ozone models: Ozone transport in a complex tropopause folding event, *J. Geophys. Res.*, 108 (D12), 8529, doi:10.1029/2003JD003462.

Lin M., A. M. Fiore , O. R. Cooper , L. W. Horowitz , A. O. Langford , Hiram Levy II , B. J. Johnson , V. Naik , S. J. Oltmans , C. Senff (2012): Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions, *Journal of Geophysical Research*, 117, D00V22, doi:10.1029/2012JD018151

Lin, M., A.M. Fiore, L.W. Horowitz, A.O. Langford, S. J. Oltmans, D. Tarasick, H.E. Reider (2015): Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions, *Nature Communications*, 6, 7105, doi:10.1038/ncomms8105

Recommendations:

1. Introduction: discussions on previous modelling approaches to address the resolution-dependent processes are currently missing. There are also recent developments on global high-resolution models. For instance, the GFDL-AM3 global climate model has been run with full chemistry at 50x50 km² horizontal resolution, which has been demonstrated to better simulate long-range pollution transport and stratospheric intrusions (Lin M. et al., 2012a, 2012b). The MOZART-4 global chemical transport model has also been run at horizontal resolution of 0.7x0.7 degrees (Emmons et al., 2010). There are also a number of studies using regional-scale models driven by chemical boundary conditions from global-scale models (e.g. Huang et al., 2010; Lin M. et al., 2010). I'd suggest the authors conduct a thorough literature re-

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view on these previous modelling approaches aimed to address resolution-dependent processes, their strengths and limitations, and where the new model discussed in the present manuscript fits into the picture.

Lin, M., A. M. Fiore, L. W. Horowitz, O. R. Cooper, V. Naik, J. Holloway, B. J. Johnson, A. M. Middlebrook, S. J. Oltmans, I. B. Pollack, T. B. Ryerson, J. X. Warner, C. Wiedinmyer, J. Wilson, B. Wyman: Transport of Asian ozone pollution into surface air over the western United States in spring, *Journal of Geophysical Research*, 117, D00V07, 2012, doi:10.1029/2011JD016961.

Emmons, L. K., Apel, E. C., Lamarque, J.-F., Hess, P. G., Avery, M., Blake, D., Brune, W., Campos, T., Crawford, J., DeCarlo, P. F., Hall, S., Heikes, B., Holloway, J., Jimenez, J. L., Knapp, D. J., Kok, G., Mena-Carrasco, M., Olson, J., O'Sullivan, D., Sachse, G., Walega, J., Weibring, P., Weinheimer, A., and Wiedinmyer, C.: Impact of Mexico City emissions on regional air quality from MOZART-4 simulations, *Atmos. Chem. Phys.*, 10, 6195–6212, doi:10.5194/acp-10-6195-2010, 2010

Lin, M., T. Holloway, G. R. Carmichael and A. M. Fiore: Quantifying pollution inflow and outflow over East Asia in spring with regional and global models. *Atmos. Chem. Phys.*, 10, 4221–4239, 2010.

Huang, M., et al. (2010), Impacts of transported background ozone on California air quality during the ARCTAS-CARB period a multi-scale modeling study, *Atmos. Chem. Phys.*, 10, 6947–6968, doi:10.5194/acp-10-6947-2010.

2. Need more careful analysis to isolate the “coupling” effects

If examining the influence of the “coupling effects” is one of the major goals of this manuscript, the authors could conduct several additional analyses, including:

1) Look at background sites outside of the nested domain. For example, examining the simulated difference in day-to-day variability of ozone at Mauna Loa Observatory in the subtropical North Pacific.

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2) Time series analysis for ozone at remote baseline sites in Europe and the western USA.

3) Examining the difference during long-range transport and stratospheric intrusion events.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 25789, 2015.

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15, C9117–C9123, 2015

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