

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

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General Response:

We thank the reviewers for comments, which have been incorporated into the revised manuscript.

Our paper is focused on how small-scale nonlinear chemical and physical processes (especially those over the pollution source regions) affect the global ozone chemistry. We use a modeling approach to show this effect, by analyzing the influences of scales that are resolved by the regional nested models but not by a typical coarse-resolution global model. In particular, the two-way coupled system better captures the small-scale processes over the pollution source regions (within the nested model domains). More

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importantly, the two-way system also allows for the improved representation of these small-scale processes to affect the global simulation, by allowing for the nested model results to modify the global model results in such way that the improvement can be transported outside the nested domains. In turn, the improved global simulation affects the lateral boundary conditions of nested models and thus improves the nested model simulations. These points were shown in the original manuscript, and they have been further clarified in the revised manuscript (e.g., the abstract, the introduction, Sect. 5.1, and various other places). Therefore, by using the model approach, we show that such interactions of processes between different spatial scales (as represented by the two-way “coupling”) does affect the global and regional ozone chemistry. We believe that ACP is an appropriate outlet for our analysis.

We have substantially revised the introduction to better clarify that small-scale processes not resolved by the global model and the interactions between small and large scales are important not only for the regional domains but also for the global domain. Our discussion is relatively brief in the original manuscript, as we felt our previous paper on CO (Yan et al., 2014, ACP) have made this point clear. In the revised manuscript, we have taken the reviewers’ suggestions and added much more discussions on these aspects.

We have added a new Sect. 5.1.1 to specifically show that the two-way coupling affects the lateral boundary conditions of the nested models, and thus improve the ozone simulation upon a typical “one-way” nested/regional model. We have also updated the abstract and conclusion to reflect this new analysis.

We agree that the ozone differences between the global model alone and the two-way coupled system are a result of multiple factors, including the sub-coarse-grid chemical variability resolved by nested resolution (i.e., emission spatial variability and associated chemical contrast), the sub-coarse-grid variability of non-chemical factors (such as topography), a slight difference in the magnitude of natural emissions (mainly for biogenic NMVOC emissions, Sect. 2.1), and a slight difference in the magnitude of

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STE (Sect. 4.1). In the new Sect 4.3, we have added additional simulations to delineate the individual effects of these factors. The results confirm our previous conclusion that the ozone difference is dominantly driven by the small-scale (i.e., sub-coarse-grid) chemical variability associated with the emission spatial variability.

Based on the reviewers' comments, we have decided to remove the old Sect. 4.3 discussing the realization of the nonlinear ozone chemistry (i.e., NO_x and VOC dependence) in our particular chemical scheme, as we agree that this information is relatively well understood and is thus not necessary to be presented in detail here. Instead, we have made a new Sect 4.3 to delineate the individual effects of various factors relevant to the simulations here (see above paragraph). For similar reasons, we have also removed the old Sect 4.4 and incorporated the associated analysis in the new Sect. 4.3.

Reviewer 1

This paper examines the impact on global model simulations of tropospheric ozone of including three higher resolution nested regions which feed back to the global model. The benefits of the coupled system are assessed by comparison with atmospheric measurements and are shown to reduce biases in the stand-alone global model. The paper is well written, with the methods and results clearly presented and explained. The model evaluation against available tropospheric measurements is also thorough.

We thank the reviewer for comments.

My main concern is that the results obtained from this one study with one model, GEOS-chem, may not be sufficiently general or provide sufficient scientific insight for publication in ACP and that GMD might be a more appropriate journal for what is principally a model validation paper. It is not surprising that changing the resolution of a model changes the results, nor that a higher resolution, albeit not for the full model domain, leads to a better simulation. The authors make frequent references to 'non-linear chemistry' as an explanation for the differences, but I'm not convinced that the

C11461

actual chemical mechanisms are fully elucidated. Interpretation of the results is also hampered by having the emissions and STE also change within the nested regions so that ascribing changes in the simulated ozone to a particular cause becomes difficult. Nevertheless, I do consider the study merits publication in either ACP or GMD.

In this study, we demonstrate how the interaction between processes of various scales affects the global ozone chemistry. The nonlinear ozone chemistry and NO_x-VOC dependence is well known. Here we extend this point to demonstrate that the small-scale variability over the pollution source regions affects the global ozone chemistry (i.e., both within and outside the nested domains), a result much less discussed in the literature. We believe ACP is an appropriate outlet. Further, the sign of ozone change with increased resolution is consistent with previous studies (Wild and Prather, 2006; Lin et al., 2008; Krol et al., 2005). We find no evidence why our finding here could be substantially model-dependent, although we only use one model (GEOS-Chem) to demonstrate the point. We agree and hope that the importance of small-scale variability and scale interactions be analyzed in other global model studies in the future, especially to address the effects of scales below 100 km and even 50 km. We have added in the conclusion that "Similar simulations with other global models would further test the importance of small-scale chemical variability for the global ozone chemistry."

In the revised manuscript, the effects of emission magnitude, emission spatial variability and STE are quantified (see the General Response). In particular, the effects of the small differences in emission magnitude and STE are both very small for the simulated global tropospheric ozone. The emission spatial variability and associated chemical variability is the dominant factor.

Specific Comments

P.25792, L.10

It could be noted that 0.667x0.5 is still a relatively coarse resolution compared with many regional CTMs (which still struggle to match point measurements near emission

C11462

sources).

We agree and have added a sentence in the end of the paragraph: “Note that our nested model resolution is still relatively coarse compared to some other regional model studies (Kuhlmann et al., 2015; Terrenoire et al., 2015); our future studies will take advantage of the new generation GEOS-Chem nested models at 0.3125° long. x 0.25° lat. to capture smaller-scale processes not resolved on a 0.667° long. x 0.5° lat. grid.”

P.25792, L.10

It might be helpful to include a global map with the three nested regions indicated.

The map is shown in the new Fig.1.

P.25793, L.19

Has it been tested whether it is appropriate to change the model’s horizontal resolution while retaining the same vertical resolution?

The vertical and horizontal resolutions are both determined by the GEOS-5 met fields. The coarse-horizontal-resolution met fields are regridded from the native fine-horizontal-resolution met fields, during which process the vertical resolution is unchanged. Both the coarse-horizontal-resolution global GEOS-Chem model and its fine-horizontal-resolution (one-way) nested models are widely used and well documented in the literature.

P.25796, L.8

Why are the isoprene emissions higher at higher resolution? How does the land cover change?

Biogenic emissions of NMVOC follow the MEGAN model (Guenther et al., 2012) with a PCEEAA approach, driven by monthly mean MODIS leaf area index data specific to the horizontal resolution of individual models

C11463

(http://acmg.seas.harvard.edu/geos/wiki_docs/emissions/megan.pdf).

Isoprene emissions nonlinearly depend on temperature, solar radiation and other factors. Accounting for the spatial variability of these parameters within a global model gridcell tends to lead to increased emissions, as also shown in Yan et al. (2014).

We have added more information on biogenic emissions in the revised manuscript.

P.25796, L.22

Did you check that afternoon ozone is close to the max 8h average in both model and measurements? It would be reassuring to know that the simulated diurnal cycle is somewhat realistic.

We did. We have added in the revised Sect. 3.1 that “The afternoon mean ozone is close to the maximum 8-hour average ozone in both measurements (36.1 ppb versus 39.3 ppb averaged over the 1420 sites) and model simulations (46.8 ppb versus 48.4 ppb for the global model alone; 42.6 ppb versus 44.5 ppb for the two-way coupled system). Models also capture the diurnal cycle of measured ozone fairly well, although with positive biases in both daytime and nighttime (not shown), consistent with our previous work (Lin and McElroy, 2010).”

Section 3.3

I appreciate the authors are being careful, but I found the detailed discussion of the two TCO products a little distracting from the main modelling results and suggest it might be shortened a little.

We have elected to retain the discussions as we feel some readers (e.g., the second reviewer) may be interested in how we treat the products.

P.25799, L.8 ‘The two way coupling.. (Yan 2014)’ Does this statement apply only to CO? Otherwise it appears to preempt the results about to be presented and discussed.

We have removed the sentence and revised the paragraph as follows:

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“This section examines the effect of two-way coupling on the simulated tropospheric ozone budget in 2009 (Sect. 4.1), with additional discussions on NO_x, CO, NMVOCs, OH, and lifetimes of methane and methyl chloroform (MCF) (Sect 4.2). In Sect. 4.3, we delineate the chemical and non-chemical factors driving the differences between the two-way system and the global model alone.”

P.25800, L.12

‘... indicates a significant effect of model resolution’ Or of changed emissions?

We have revised the sentence:

“Although both lifetimes calculated here are broadly consistent with previous studies (19.9–25.5 days from ACCMIP (Young et al., 2013) and 17.3–25.9 days from ACCENT (Stevenson et al., 2006)), the reduction due to our model coupling indicates a significant effect of small-scale processes resolved by the finer resolution, especially the fine-scale spatial variability of emissions and associated chemistry.”

P.25800, L.13-21

All the differences discussed are quite small, in line with what might be expected from any minor change to the model configuration. How significant do the authors consider them?

Although the changes in chemical production and loss magnitudes are relatively small (i.e., within 3%), the changes in ozone burden and lifetime (about 10%) is significant. It indicates the importance of considering small-scale (sub-coarse-grid) variability for the global ozone chemistry.

P.25801, L.25

Add brief comment on why it is okay to do this analysis while the model is still spinning up.

We felt analyzing the spin-up period was sufficient to demonstrate the effect of small-

C11465

scale nonlinear ozone chemistry. Nonetheless, we have removed this discussion in the revised manuscript, as we have added additional full simulations to delineate the effects of individual factors.

P.25802, L.7 to P.25803, L.8

I didn’t find Figures 2 and 3 and the associated discussion particular enlightening. Is there more to say than that the higher resolution model shows more detail and more urban rural contrast?

We have replaced the old Sects. 4.3 and 4.4 with a new Sect. 4.3. See the General Response.

P.25802, L.25

‘...reflecting the resolution dependent non-linear chemistry’, Expand and give a more detailed explanation.

We have replaced the old Sects. 4.3 and 4.4 with a new Sect. 4.3. See the General Response.

P.25803, L.8

‘...the amount of ozone produced is affected by model resolution’. Again, expand

We have replaced the old Sects. 4.3 and 4.4 with a new Sect. 4.3. See the General Response.

Section 4.4

This section is somewhat perfunctory. Contrasting chemical and non-chemical impacts of changing resolution merits a little more discussion.

We have replaced the old Sects. 4.3 and 4.4 with a new Sect. 4.3 and detailed analysis of these aspects. See the General Response.

Section 5.1

C11466

My understanding is that the ozone within the nesting domain is being compared with the measurements after being re-gridded back onto the coarser global domain? So Figures 5 and 7 are comparing point measurements with the model ozone 'averaged' over a 2.5x2 degree grid cell. This could be mentioned. Is it worth looking at the bias of the model ozone on the original high resolution nested grid? The biases versus the EMEP measurements could be compared with the biases between those measurements and the EMEP/MSW model regional CTM reported in the annual EMEP Status Reports (www.emep.int/mscw/mscw_publications.html).

Indeed, Figures 5 and 7 are comparing point measurements with the model ozone 'averaged' over a 2.5x2 degree grid cell. We have clarified this point in the revised manuscript. We did not save results from the nested model component of the two-way system.

The biases here are larger than the EMEP/MSW model, as discussed in the revised Sect. 5.1:

"The two-way simulation biases against the EMEP measurements are larger than those for the EMEP/MSW regional CTM at a horizontal resolution of 50 km x 50 km driven by year-specific emissions (within 10%) (http://emep.int/publ/reports/2014/sup_Status_Report_1_2014.pdf). Our higher biases are partly because the 2005 EMEP NO_x emissions used here are higher than those in 2009 by 25.3% (http://webdab.umweltbundesamt.at/official_country_trend.html)."

P.25809, L.7

I'm not convinced that it has been fully demonstrated that the model improvements are driven by better representation of chemistry rather than by changes in emissions or STE. If the DSMACC simulations are a key element in this argument why relegate them to an appendix? The broad conclusion that small scale processes are important for global chemistry and higher resolution gives a better simulation is somewhat general, and adding more specific details to the conclusions would be beneficial.

C11467

In the new Sect 4.3, we have used multiple additional model simulations to show that the effects of differences in emission magnitude and STE are small, and that the emission-chemistry spatial variability is the dominant factor. With the new Sect. 4.3, we think it becomes unnecessary to show the discussion related to DSMACC. See the General Response.

Technical Comments

P.25791, L.12 destructed -> destroyed Modified

P.25791, L.23 its nonlinear -> their nonlinear Modified

P.25793, L.8 upon -> over Modified

P.25796, L.24 ozone are sampled -> ozone is sampled Modified

P.25797, L.5 of polluted environment -> of the polluted environment Modified

P.25797, L.6 aircrafts -> aircraft We think the word is correct.

P.25797, L.23 ozone are sampled -> ozone is sampled Modified

P.25798, L.21 TCO are calculated -> TCO is calculated Modified

P.25800, L.26 by global model -> by the global model Modified

Table 2 Is 'Fos' in row 5 a typo? Modified

Figure 9 caption Frankfort -> Frankfurt Modified

â€” Reviewer 2

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

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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:

Our original manuscript made it clear that the small-scale processes (resolved by the nested models but not by the global model) affect the ozone chemistry not just within the nested domains but also outside the nested domains via the two-way “coupling” (i.e., nested models feed back to the global model). In addition, changes in the global chemical background will further affect the lateral boundary conditions of the nested models, a feature not seen in the traditional “one-way” nested modeling. In the revised manuscript, we have taken some of the reviewer’s suggestions to further clarify this point – for example, see the new Sect. 5.1.1. We believe the effect of small-scale processes and scale interactions (or feedbacks through “coupling”) on the global and regional ozone chemistry found here is an important new contribution to the literature, and that ACP is an appropriate outlet. Please see the General Response for more discussions.

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

C11469

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)?

Again, the “coupling” allows for the nested model results to affect the global model simulation, which in turn affects the lateral boundary conditions of the nested models, i.e., it allows for “feedbacks” between models and interactions between scales resolved by models of different resolutions. We have revised the introduction, Sect. 5.1.1 and may other places to further clarify these points. See the response above and the General Response for more details.

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

We agree that model ozone bias is a multi-factor problem. In the revised introduction, we have clarified that model bias can be caused by many factors including emissions, meteorological fields, chemical mechanisms, and resolution. In this paper, we address

C11470

the effect of small-scale (sub-coarse-grid) processes by comparing models of different resolutions and setups.

Summertime afternoon surface ozone is not improved significantly by the two-way coupled system. We have explained in the revised manuscript that:

“It is partly because the enhanced ozone production from the increased natural precursor emissions (Table 2) compensate to some extent for a stronger chemical ozone loss; a sensitivity global model simulation adopting emissions in the two-way system produces more summertime ozone than the original global model by 1.7 ppb over the eastern U.S. (100-70°W, 30-50°N) and by 2.1 ppb over Europe (10°W-30°E, 35-70°N). Furthermore, although the nested models reduce the net chemical production of ground-level ozone (Sect. 4.3), the effect is partly offset by stronger vertical transport that brings more high-ozone air aloft down to the ground (Roelofs et al, 2003; Lin M. et al., 2012b). The persistent large summertime bias may also be due to some non-resolution-dependent factors such as isoprene nitrate chemistry and dry deposition (Lin et al., 2008; Fiore et al., 2014; Monks et al., 2015). Although the two-way coupling leads to a relatively small improvement in summertime ground-level ozone simulations over the U.S. and Europe (Fig. 4), the coupling results in large error reductions for tropospheric ozone (see Sects. 5.2 and 5.3 below).”

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.

We have substantially revised the manuscript to incorporate the reviewer's suggestions. Please see the General Response.

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

C11471

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

We have added more analyses on surface ozone simulations outside Europe and North America, including model evaluation at three sites over China from WDCGG (see the new Figs. 5 and 6 and related discussions in Sect. 5.1). We don't have other detailed (i.e., hourly or 3-hourly) ozone data over China for model evaluation in the present paper. The observation data shown in the papers suggested by the reviewer are all prior to 2009, and we don't have the corresponding detailed data in 2009. Although monthly mean ozone measurements can be inferred from the literature, our initial analysis suggested that detailed ozone data are necessary for quantitative model evaluation, in

C11472

order to minimize the sampling bias associated with the large diurnal and day-to-day ozone variability; see for example Fig. 5 for the large day-to-day variability. In the future, we hope to conduct a more specific analysis over China when detailed ozone observation data become available.

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.

The effect of STE difference is very small, as analyzed in the new Sect. 4.3. Please see the General Response.

The larger ozone variability in the observed profiles is simply because the observation is sampled at every 0.15 km vertically, at a much finer resolution than the vertical resolution of the model. The figure below shows that if the observations are mapped to the vertical resolution of the model, the observed variability is greatly reduced to a level comparable to the modeled variability. We have added this discussion in the revised manuscript.

Figure 1. Vertical distribution of observed (black) and two-way simulated (green) ozone at the 11 MOZAIC sites. Here the observations are mapped vertically to the resolution of the model.

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

C11473

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.

OMI/MLS does not provide averaging kernel data.

Modeled monthly mean TCO is calculated from all daily data at the OMI overpass time (13:00–15:00 LT, local time) applied with the monthly mean averaging kernel from the OMI/LIU product. Daily averaging kernel data are not available. The modeled global annual TCO with and without applying the averaging kernel differ by 0.6%. We have added this information in the revised manuscript.

We note that although it is useful to apply the averaging kernel when comparing to satellite data, many studies (e.g., Young et al., 2013) do not apply the averaging kernel.

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 troposphere. 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.

C11474

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

In our model setup, we adjusted the STE in the nested models so that the simulated STE is very close to the STE simulated by the global model alone. This is indeed to ensure the differences between the two-way coupled system and the global model alone are not determined by STE. This point was shown in Sect. 2 of the original manuscript, and has been further clarified in the revised Sect. 2.

The new Sect. 4.3 further shows that the slight residual difference in STE between the global model alone and the two-way coupled model has a marginal effect on the simulated ozone.

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 review on these previous modelling approaches aimed to address resolution-dependent

C11475

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.

We have revised the introduction substantially to incorporate the reviewer's suggestions. Please see the General Response for why we elected to only provide a brief introduction in the original manuscript.

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

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simulated difference in day-to-day variability of ozone at Mauna Loa Observatory in the subtropical North Pacific.

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.

Our original manuscript already shows that the two-way coupling affects ozone simulations both within and outside the nested domains, for example, the effect is clearly seen in various global maps and related analyses. In the revised manuscript, we have further strengthened this point by adding more analyses on the surface ozone in Sect. 5.1., including background sites within and outside the nested domains like Mauna Loa.

As shown in the new Sect. 4.3, the effect of STE difference is marginal for the ozone difference between the global model alone and the two-way coupled system.

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

C11477

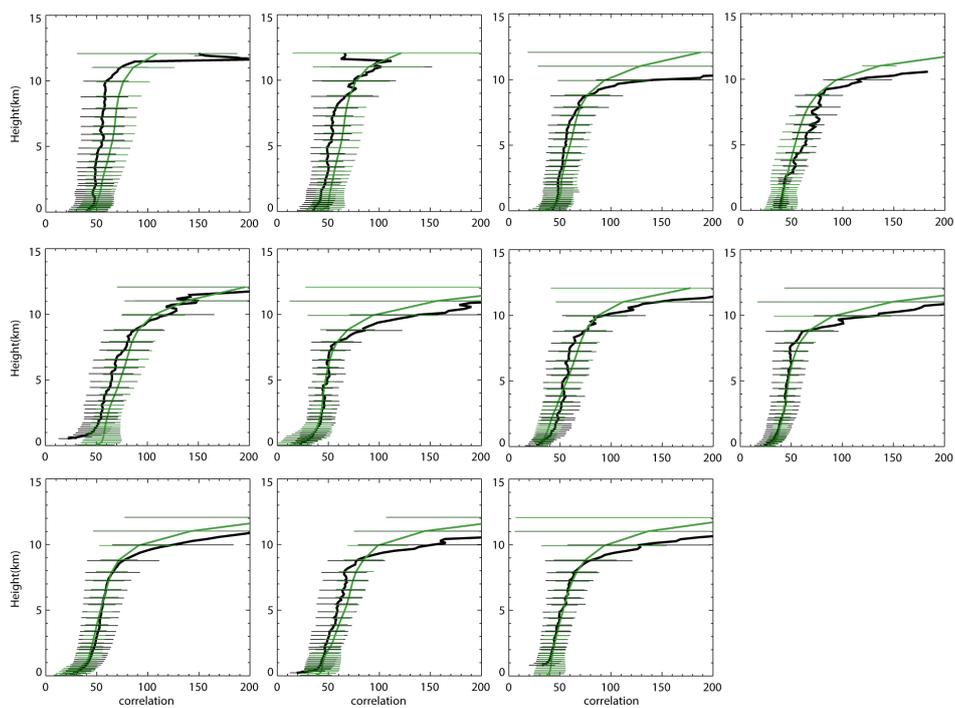


Fig. 1.

C11478