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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Application of WRF/Chem-MADRID and WRF/Polyphemus in Europe – Part 2: Evaluation of chemical concentrations, sensitivity simulations, and aerosol-meteorology interactions" *by* Y. Zhang et al.

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Reply to Interactive comment on "Application of WRF/Chem-MADRID and WRF/Polyphemus in Europe – Part 2: Evaluation of chemical concentrations, sensitivity simulations, and aerosol-meteorology interactions" by Y. Zhang et al. Anonymous Referee #1 Received and published: 31 March 2013

In the two manuscripts, Zhang et al. compare two air quality models, an offline-coupled model and an online-coupled model to assess their capabilities and differences in simulating air pollutants and aerosol-meteorology interactions over Europe. In the second part, the model performance is evaluated, model sensitivity is investigated and aerosol and meteorology interactions simulated by online model are quantified. Although the method of this research is solid, the results are not surprising and do not contribute significant scientific understanding. Differences between two models with different vertical layers and inputs are expected without doing simulation. Simulation of sensitivity to grid resolution is a meaningless effort to prove what already known to the scientific field. Also, more investigation is needed to explain the feedbacks of aerosols. Thus, I don't recommend the publication of this manuscript in ACP without major revisions according to following comments.

Reply:

We thank the reviewer for valuable comments. We've addressed all review comments into the revised manuscript. Please see our point-by-point replies below. Major comments:

1) Comparison of the two models is far-fetched. The differences are expected and you can attribute the differences to anything since there are too many differences as the authors do: "The differences in model predictions are caused by differences in the heights of the first model layers and thickness of each layer that affect vertical distributions of emissions, model treatments such as dry/wet deposition, heterogeneous chemistry, and aerosol and cloud, as well as model inputs such as emissions of soil dust and sea-salt and chemical boundary conditions of CO and O3 used in both models."

Reply:

We agree with the reviewer that the two models have many differences. However, Polyphemus and WRF/Chem are representative offline and online coupled models used in Europe. Given that both types of models are commonly used in the com-

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munities and have their own merits to be continuously developed and applied in parallel, we believe that this comparison is a valuable contribution to the community. As the reviewer pointed out, differences between model predictions may be caused by many factors. However, we identified through this comprehensive comparison some very specific factors/processes that are useful for further model development and improvement. 2) It is well known to the field that finer grid resolution gives better model performance. This part is of no scientific interests.

Reply:

Based on our experience doing multiple nested simulations using various meteorological models (MM5 and WRF) and air quality models (CMAQ and WRF/Chem) as well as publications in the literature, model predictions using finer grid resolution do not always give better model performance. The model performance depends on many factors including accuracies in model inputs and model treatments. However, there are large uncertainties in the model configurations (e.g., horizontal and vertical grid resolutions, nesting options, and data assimilation options). In fact, we've often seen worse performance with finer grid resolution in many applications. As we pointed out in the Introduction section:

"Several studies, on the other hand, showed that a coarser grid resolution provided similar or even better air quality predictions than a finer grid resolution (Mathur et al., 2005; Arunachalam et al., 2006; Cohan et al., 2006; Zhang et al., 2006; Queen and Zhang, 2008; Liu et al., 2010). Bailey et al. (2007) and Valari and Menut (2008) found that model results do not improve monotonously with resolution."

For this particular application, our results showed that "For both models, the use of a finer grid resolution generally leads to an overall better statistical performance for most variables, with greater spatial details and an overall better agreement in temporal variations and magnitudes at most sites."

Therefore, we believe our results illustrate cases where there is a the benefit of using

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finer grid resolutions and they are interesting to the community.

3) How does biogenic emission generating method affect SOA? How much is the effect to anthropogenic SOA and biogenic SOA? The change is due to the increase of oxidants or VOC precursors?

Reply:

As pointed out in our APCD paper, page 4093, lines 18-19, "Different BVOCs modules produce significantly different BVOCs emissions in terms of total emissions and their spatial distributions." Different BVOCs emissions will in turn affect SOA formation, because they are SOA precursors. Such different emissions will affect mostly biogenic SOA, which dominates total SOA formation. For example, comparing to the simulation with offline BVOCs, the simulation using MEGAN BVOC module gives lower SOA and BSOA concentrations, by 1.32 ug m-3 for the domainwide mean (-30%) and as low as -19.7ug m-3 (by -92.5%). The impact on the absolute concentrations of anthropogenic SOA is very small, e.g., using MEGAN 2 BVOC module gives lower anthropogenic SOA concentrations, by 0.009 ug m-3 domainwide mean and as low as 0.06 ug m-3. The change is due to the changes in oxidants, VOC precursors, and, to a lesser extent, to the organic aerosol mass available for SOA absorption. Note that biogenic emissions predicted by both modified Guenther and MEGAN2 modules are lower than those based on offline emissions, making more OH available for oxidation of other gaseous species.

To address the comments, we've added the plots for OH and ASOA from WRF/Chem offline BVOCs simulation, and the absolute differences in their concentrations between the three simulations in Figures 13 and 14. We also added the above points in the relevant discussion section.

4) Feedbacks of aerosols to meteorology and so as to the pollutants concentrations are interesting. It seems that with or without aerosol feedbacks makes large differences to pollutants concentrations especially sulfate and nitrate (Figure 14). In Figure 14, the

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maximum difference for nitrate is 15 ug/m3, for sulfate is 9 ug/m3. The difference due to aerosol feedbacks can be such larger? The changes are close to the predicted sulfate and nitrate (check the numbers in Figure 2). It is hard to believe.

Reply:

It seems that the reviewer may have misunderstood Figure 14 in the original manuscript, which shows the differences in chemical species concentrations from two simulations, one from WRF/Chem-MADRID that includes all model species and mechanisms and one from the same model but without PM primary emissions and secondary PM formation. This was indicated in our original Part I paper, page 4004, lines 24-28 and page 4005, lines 1-2:

"To estimate the effects of aerosols on model predictions through various feedback mechanisms, an additional simulation is performed using the online-coupled WRF/Chem-MADRID with the MEGAN2 BVOCs module by turning off primary aerosol emissions and secondary aerosol formation. The differences in the model predictions between this simulation and the simulation using WRF/Chem-MADRID with the MEGAN2 BVOCs module that include all primary aerosol emissions and secondary aerosol formation represent the effects of aerosols via various feedback mechanisms.

While the differences in the meteorological variables shown in Figure 13 in the original paper (now Figure 5 in the revised paper Part I) and gaseous phase species concentrations shown in Figure 14 in the original paper (which remains as Figure 14 in the revised paper Part II) represent the effects of aerosols via various aerosol formation and feedback mechanisms on meteorology and gas-phase chemistry, respectively, those plots for aerosol species do not reflect aerosol feedbacks. They reflect primary aerosol emissions and second aerosol formation processes such as aerosol thermodynamics and cloud chemistry that were included in the simulation using WRF/Chem-MADRID with the MEGAN2 BVOCs module that includes all primary aerosol emissions and secondary aerosol formation. This is why their values are similar to Figure 2 in the part

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II paper. Simulated concentrations of NH4+, NO3-, and SO42- in the original Figure 14 are slightly lower than those in Figure 2, because the values in Figure 2 include PM species concentrations from their initial and boundary conditions (ICs and BCs), in addition to those generated from all primary aerosol emissions and secondary aerosol formation. The contributions of ICs and BCs to aerosol concentrations are excluded in the original Figure 14 by taking differences between the two simulations, so the differences represent the net contributions from all primary aerosol emissions and secondary aerosol formation to the concentrations of secondary aerosol species.

To avoid any confusion, we've clarified this point in the relevant discussion section and indicated clearly that they are formed from emissions of primary SO42- and NO3- and gaseous precursors for secondary aerosols. We also changed the subtitle to be "Impact of Aerosols on Gaseous Pollutant Predictions"

5) What are the mechanisms for aerosol feedbacks, what cause the large difference? -34 to 18 m change of PBL height in Figure 13 is not likely to explain large change of nitrate. Does this change happen during day time or night time?

Reply:

As explained above, the reviewer may have misunderstood the changes in concentrations of aerosol species including nitrate in the original Figure 14. Nitrate is formed through gas-to-particle conversion process such as aerosol thermodynamics and cloud chemistry. Specific comments:

1) Figures 1-2, the cover area of D01 and D02 are different and the color bars are different. It is very difficult to see the differences. Please change to same color bar for same species. The numbers of color bar for column 3 of Figure 2 has some problem. Also the quality of each field plot is very low. It is hard to get a clear look no matter how much I zoom in.

Reply:

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All figures for the same species used the same color and Y scale except for one plot, i.e., the SO2 plots over D01 from WRF/Chem-MADRID. This has been fixed by using consistent Y scale values with all other SO2 plots. The low resolution probably resulted from the PDF conversion by the online paper submission system. We've included the eps version of those figures, which should greatly improve the resolution of all figures. 2) Figure 10, it is hard to tell the difference between each column, it is better to show the difference.

Reply:

The second and third columns in Figure 10 have been replaced by two sets of differences plots among the three simulations with WRF/Chem-MADRID: (1) Modified Guenther - Offline BVOCs (2) MEGAN 2 - Offline BVOCs The plots in the first column are kept to show the spatial distributions of predicted species concentrations from the simulation with offline BVOCs emissions to be compared with changes in the spatial distributions caused by the use of the modified Guenther or MEGAN 2 BVOC online modules in the second and third columns.

3) Figure 14, the color bar should be changed. For example, the nitrate panel, the range of nitrate is -0.1 to 15, why the color bar is -20 to 20, it is difficult to get the values from color bar. Same for other plots.

Reply:

We've adjusted the color bar for some plots including nitrate, CO, O3, NH3, NH4+, SO42-, NO3-, SO2, and HNO3 in the original Figure 14 in Part II (now Figure 13 in the revised paper Part II). We also made similar adjustments for COT, T, CCN, CDNC, and PBLH plots in the original Figure 13 in Part II (now Figure 5 in Part I).

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4059, 2013.