Responses to the reviewer's comments on Li Li's manuscript

Referee 1:

1. How many vertical layers used in MM5 and CMAQ? In the paper, line 19-20 on page 15054 "Both the MM5 and CMAQ employ 14 vertical layers of varying thickness with denser layers in the lower atmosphere to better resolve the mixing height". What are the detailed 14 vertical layers?

   Answer: The number of vertical tropospheric levels used in CMAQ is 14 from the surface up to 500hpa. The vertical resolution of the 14 layers corresponds to sigma levels of 1.000, 0.995, 0.988, 0.980, 0.970, 0.956, 0.938, 0.893, 0.839, 0.777, 0.702, 0.582, 0.400, 0.200, and 0.000 at the boundaries of the layers.

   Text regarding the details of the vertical layers has been added to the manuscript.

2. In Table 1, the unit of relative humidity, may change to water mixing ratio.

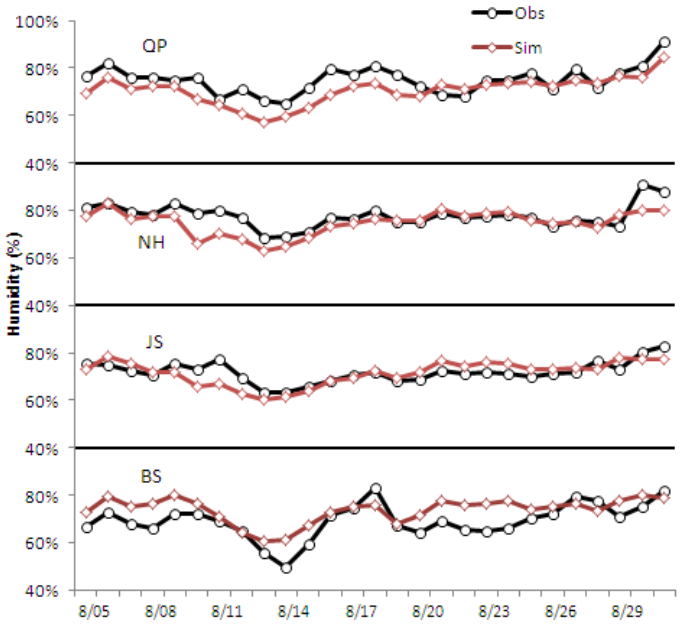
   Answer: The unit of relative humidity has been changed to water mixing ratio. Text has been revised.

Table 1 Statistical results between MM5 model and observation data at surface stations in Shanghai

		BS	JS	NH	QP	Average
Wind Speed	RMSE(m/s)	1.76	1.43	1.13	1.24	1.39
	Bias(m/s)	0.94	0.57	0.43	0.65	0.65
	IOA	0.4	0.44	0.57	0.51	0.48
Wind Direction	Gross Error(deg.)	38.02	30.88	29.43	39.79	34.53
	Bias(deg.)	3.21	4.55	-2.83	5.05	2.50
Temperature	Gross Error(K)	0.82	1.18	1.55	1.21	1.19
	Bias(K)	-0.91	-0.71	1.36	0.77	0.13
	IOA	0.76	0.81	0.83	0.90	0.83
Relative Humidity	Gross Error(%)	5.77%	3.35%	3.81%	5.14%	4.52%
	Bias(%)	4.41%	-0.33%	-2.77%	-4.41%	-0.78%
	IOA	0.96	0.97	0.96	0.96	0.96

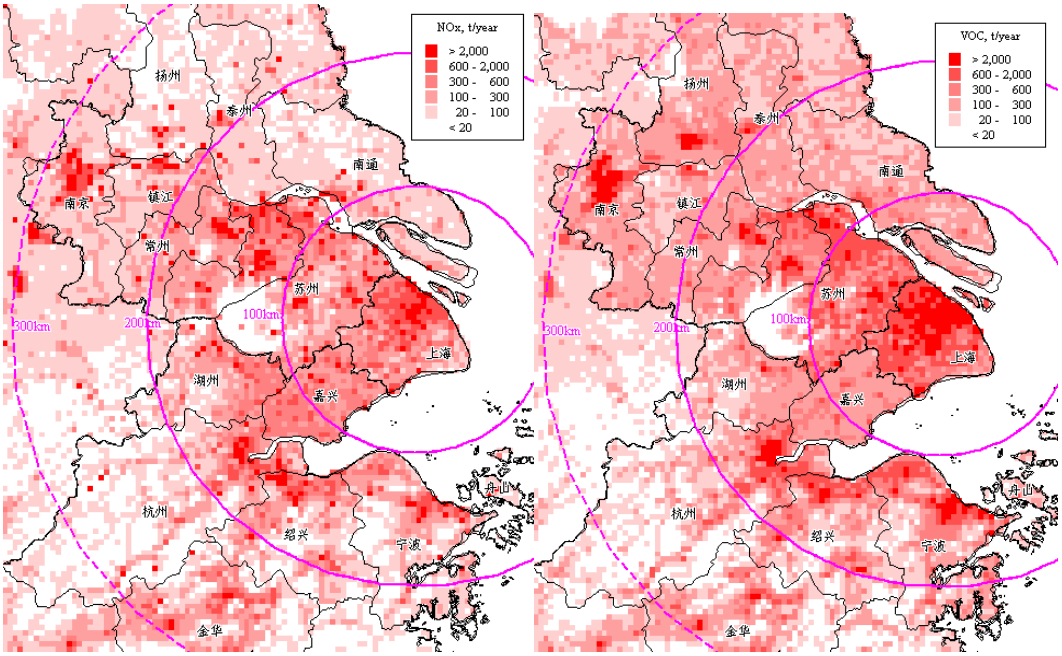
3. Figure 3 on page 15075, the humidity values are around 20g/kg, are they correct? Also, in the tile of this figure, the relative humidity is not suitable.

   Answer: We have changed the humidity from g/kg to water mixing ratio (%). The sub-figure has been revised.



4. How about the distribution of emissions of NO_x over YRD used in CMAQ model?

   Answer: The distribution of emissions of NO_x over YRD used in the CMAQ model is shown in the following figure. We have added the emission distribution of NOx and VOCs to the manuscript to help explain model results.



5. There are no vertical O_3 observations to compare with the simulations, if the vertical simulation results were compared with the measurements, it will be better to use process analysis method to understand the different layer results.

Answer: This is a very good suggestion. The authors would also like to see real measurements of vertical O_3 concentrations. Since there is a lack of observations of the vertical O_3 concentrations in the YRD, it is not possible to include them in this paper. We hope that such concentration data will become available in the future and we can include them in our future work. Text regarding the vertical O_3 observations has been added to the conclusions part in the paper.

Response to the Interactive comment on “Process analysis of regional ozone formation over the Yangtze River Delta, China using the Community Multi-scale Air Quality modeling system” by L. Li et al.
Anonymous Referee #2

The paper creates a conceptual model describing high ozone events over the Yangtze River Delta. The authors accomplish the task by using photochemical modeling by the Community Multi-scale Air Quality (CMAQ) model and observations from local monitors but their analysis and conclusions mostly rely on the CMAQ modeling and its options for Integrated Process Rates (IPR) analysis. The latter shows that regions switch between ozone production and destruction based on emissions and that ozone transport from production regions aloft play a critical role in high concentrations versus time. Their conceptual model seems consistent with the analysis and observations but does have faults.

(1) It does not employ quantitative measures when regions switch between production and destruction. The measure could have used the emission inputs or CMAQ predictions to determine when or where ozone chemistry switches between NOX and VOC limiting conditions. Alternatively, additional analysis could have used Integrated Reaction Rate (IRR) analysis to quantify what reactions and compounds dominate the photochemical sources and sinks of ozone. IRR is another analysis tool available in the CMAQ model.

Thanks to the reviewer for this very good suggestion. This is very important in evaluating the model results. It is correct that the IRR method deals with the details of the chemical transformations and it can help to explain how the ozone is formed in the chemical mechanism. We are currently working on this, and we will report the study results later. In this manuscript, we have added the emission inputs (including emissions rates of O_3 precursors and diurnal profile) of the four sites analyzed to help determine when or where ozone chemistry switches between NO_x and VOC limiting conditions. Text regarding the emission inputs have been added to the paper.

(2) IPR identifies horizontal transport as an important factor leading to high ozone over the delta. However, results do not discuss what amount of the transport comes from outside the smallest nested model domain. Figure 10 attempts to describe this amount but the figure is difficult to interpret based on the number and resolution of sub-figures. It may improve by reducing the number of sub-figures and increasing the sizes of the remainder.

The number of sub-figures in figure 10 (in the revised paper is figure 17) has been reduced and the sizes of the remainder have been increased to help explain the amount of the transport coming from outside the smallest nested model domain.

(3) The authors do not discuss uncertainties that may alter the conceptual model. For example, the modeling set-up used clear air conditions as the boundary conditions for the outer most nested domain. The condition introduces an uncertain because a strong

potential exists long range transport of ozone and its precursors from outside the nested modeling domains such as Korea, Indo-China and Japan. Another example is that the index of agreement decreases from NO2 to NOX. The decrease infers a problem modeling partitioning between NO2 and NO either from emissions or the photochemical mechanism. The paper's conclusions may weaken because this partitioning plays in switching between ozone production via NO2 photolysis and destruction via NO titration.

Three-dimensional air quality models like CMAQ are governed by a set of differential equations for atmospheric dynamics and thermodynamics in a generalized coordinate system. Therefore, the major inputs to the model may affect the uncertainty of the modeling results, including meteorological inputs, emissions, initial and boundary conditions, and the chemical mechanism.

Previous studies (e.g., Jiménez et al., 2007) show that the impact of IC for ground-level ozone is negligible after a 2-day spin-up period.

For the boundary conditions, there are usually three methods for providing BC to the numerical model: (1) fixed, time-independent concentration profiles; (2) concentrations extracted from the mother domain with application to the nesting approach; (3) concentrations from the GEOS-Chem or MOZART global model. According to Borge's research (Borge et al., 2010), the CMAQ model sensitivity to BC for NO_2 and SO_2 is small, and the CMAQ nesting approach performs better than the others in prediction of NO_2. However, significant domain-wide differences were found when modeling O_3, depending on the BC. Related studies show that model-derived, dynamic BC improved CMAQ predictions when compared to those based on static concentrations prescribed in the boundaries. Aggregated statistics suggest that the GEOS-Chem model produced the best results for O_3 and $PM_{2.5}$ while NO_2 and PM_{10} were slightly better predicted under the CMAQ nesting approach.

In this paper, the largest CMAQ domain covers the whole of China, Japan, Korea, parts of India and Southeast Asia, and we use a 4-day spin-up period, so we assume that the influence from long-range transport from outside the largest domain is not significant for O_3 compared to the influences from local emissions, although we understand that it does have some affect. Some of our on-going work involves incorporating the Global GEOS-Chem model in China to help decrease the BC influence when running the YRD regional CMAQ.

The emission inventory and the chemical mechanism may also affect the modeling results. In this paper, the index of agreement for O_3 and NO_2 shows that the model captures well the diurnal variations of the pollutants. However, as the reviewer observes, the index of agreement decreases from NO_2 to NO_x This decrease suggests that the partitioning between NO_2 and NO is not as accurate as one would like, either due to uncertainties in the emissions or the photochemical mechanism. Such model biases may affect the process analysis results to some extent, though we don't believe that the effect is very large. The under prediction of NO may result in some under prediction of the ozone titration contribution to the O_3 change. Nevertheless, the results from this study do provide valuable insights into the governing processes that control O_3 concentrations.

Text regarding the discussion of uncertainties affecting the conceptual model have been added to the manuscript.

Jiménez, P., Parra, R., Baldasano, M.J.: Influence of initial and boundary conditions for ozone modelling in very complex terrains: a case study in the northeastern Iberian Peninsula. Environmental Modelling and Software 22, 1294-1306, 2007.

Borge, R., López, J., Lumbreras, J., Narros, A., Rodríguez, E.: Influence of boundary conditions on CMAQ simulations over the Iberian Peninsula. Atmos. Environ., 44, 2681-2695, 2010.

(4) The analysis uses the August 16th thru 17th period based on the SAES site but the authors need to give more information on whether the period covers typical conditions during ozone events such as the wind patterns, synoptic or emissions conditions. Such evidence will make their conclusions more robust.

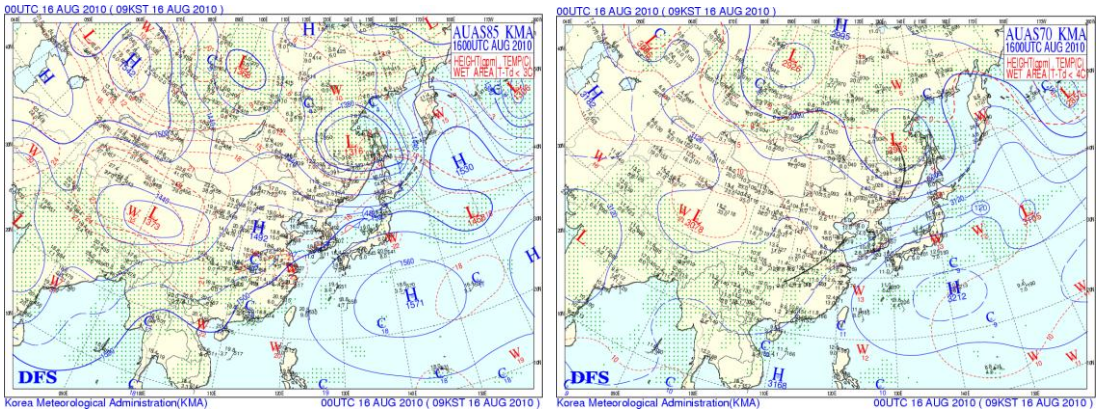
Shanghai is usually controlled by a subtropical high (NW Pacific high) in summer, but there are still several periods:

(1) In most summer season, the ridge of the subtropical high is over 30° N, Shanghai is in the strong descendind and divergence area, and the prevailing wind is southeast. Under this weather condition, the air pollutants are easy to disperse, so the quality is usually good. For example, the 4th thru 7th August, 2010 belongs to this weather condition.

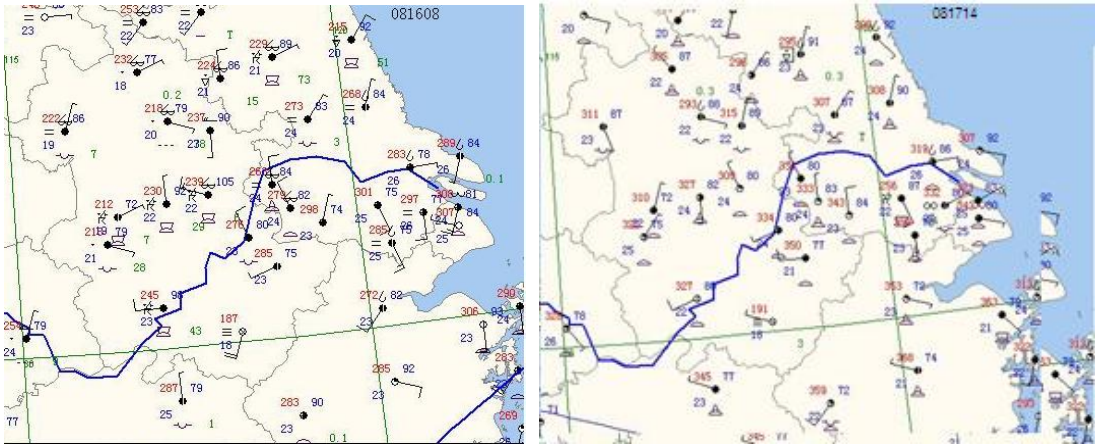
(2) Sometimes the subtropical high is rebuilding and strengthening, and move from south towards the north, Shanghai is at the edge of the north of subtropical high. During this period, the descending airflow becomes stronger. The surface wind direction changes to southwest and becomes bigger. Under this condition, the high temperature usually occurs. However, since the wind speed is relatively fast, the O_3 concentration is not easy to accumulate.

(3) When there is a weak trough in the westerly coming from the northwest, the subtropical high pressure starts to move toward the south and becomes weak. Under this special condition, a weak shear line at the convergence zone formed. Shanghai is at the edge of the northwest of the subtropical high pressure system. Both the pressure field and the southwest wind are weak. Thus it is easy for air pollutants to accumulate, and a high pollution episode occurs. The high ozone pollution episode occurred on 16 and 17 August, 2010 was under this meteorological condition.

The following figures show the weather patterns at 850hpa and 700hpa over the eastern Asia at 8:00 a.m., 16 August 2010.



The following figures show the convergence zone formed over Shanghai and the YRD area, and the surface wind direction became southwest. The air pollutants accumulate and a high pollution episode occurred.



We have added more information related to the weather conditions during August 16th thru 17th when the high ozone episode occurs.

This reviewer believes that the paper merits final acceptance if the authors revise it to remove a majority of the above faults.

Thanks to the kind reviewer, we have revised the manuscript to respond to the concerns expressed.

The below specific comments illustrate the above points or suggest additional ways to revise the paper.

(1) Page 15053, line 26: The paper should replace Dennis et al. (1996) and Byun et al. (1998) with references that give information more relevant to the CMAQ version 4.6.

The below examples give potential replacements.

Byun, D. and Schere, K.L.: Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality (CMAQ) modeling system, Applied Mechanics Reviews, 59, 51–77, 2006.

www.cmascenter.org

www.cmaq-model.org

Note that the below additional example refers to CMAQ version 4.7 but the reference gives information better describing the photochemistry and deposition processes in CMAQ version 4.6.

Foley, K.M., Roselle, S.J., Appel, K.W., Bhave, P.V., Pleim, J.E., Otte, T.L., Mathur, R., Sarwar, G., Young, J.O., Gilliam, R.C., Nolte, C.G., Kelly, J.T., Gilliland, A.B., Bash, J.O.: Incremental testing of the community multiscale air quality (CMAQ) modeling system version, 4.7., Geosci. Model Dev. Discuss. 2, 1245-1297, 2009.

We have downloaded the references and related text has been revised in the discussion paper.

(2) Page 15054, line 18: The methodology uses the clear air conditions for the outer most or largest modeling domain. This usage assumes that ozone and other pollutant have insignificant contributions transport from areas adjacent to the largest domain. Discuss whether and how the assumption can affect the paper's results.

See response above.

(3) Page 15057, lines 4-10: The paper does not make clear whether the selected episode characterizes typical condition during high ozone events. The beginning of the paper implies typical temperatures and relative humidity but states the measured ozone was extremely rare. Please clarify by discussing whether the August 16th thru 17th has other meteorological parameters typical in high ozone event such as wind patterns, weather fronts or dominant pressure systems.

See response above.

Text regarding the weather conditions during August 16th thru 17th when the high ozone episode occurs has been revised in the manuscript.

(4) Page 15058, line 19: The index of agreement noticeably decreases from NO2 to NOX. The change indicates problems simulate NO. Discuss how the error may affect accurately determining where ozone production and titration occurs over the domain.

See response above.
Text regarding the discussion of uncertainties affecting the conceptual model has been added to the manuscript.

(5) Page 15065, lines 9-22: The YRD appears to contain regions which are NOX or VOC

limiting based on local sources and times. Could the author discuss ozone concentration based on the two limits?

In the urban cities like Xuhui of Shanghai, and Hangzhou, the emissions of NOx from vehicles are more significant than the VOC, and thus the O_3 concentrations are more sensitive to VOC (Li et al., 2011), while in the oil and chemicals industrial region and the rural areas where the NO_x emissions are not so significant, the O_3 concentrations are likely to be NOx-sensitive. In the NOx-sensitive areas like Jinshan district, the ozone production rates from photochemical reaction are higher than the urban area; together with the significant horizontal transport, the O_3 concentration is higher than in the urban area which is a VOC limited region. Both the precursor emissions of NO_x and VOC at the Nanjing site are very high, causing the O_3 concentration to be the highest, while those at the Hangzhou site are lowest, and the O_3 concentrations are not so high as those at other sites during the simulation period.

Li, L., Chen, C. H., Huang, C., Huang, H. Y., Zhang, G. F., Wang, Y. J., Chen, M. H.,Wang, H. L., Chen, Y. R., Streets, D. G., Fu, J. M.: Ozone sensitivity analysis with the MM5-CMAQ modeling system for Shanghai, Journal of Environmental Sciences, 23, 1150--1158, 2011.

Text regarding the discussion of ozone concentration based on the two limits has been added to the manuscript.

(6) Page 15056, lines 23-27: The paragraph implies that paper is a novel use of IPR within the CMAQ model system. It then recommends IPR for future use. A simple search shows IPR has been used in many publish journal article so IPR's utility has already been establish. The author should revise or delete the paragraph.

This paragraph has been deleted.