

Interactive comment on “Process analysis and sensitivity study of regional ozone formation over the Pearl River Delta, China, during the PRIDE-PRD2004 campaign using the CMAQ model” by X. Wang et al.

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Received and published: 6 April 2010

Response to Reviewer #3

Thanks for your constructive comments. The followings are our responses.

General comments:

(Q1) In this paper the authors performed a modeling study using the CMAQ modeling system with attempts to characterize ozone formation, quantify the contribution of different physical and chemical processes the in situ O₃ formation through a process

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analysis, and investigate the O₃ response to emission reduction through the Brute Force method in the PRD region in China. The paper adds values in understanding the ozone chemistry in the specific region. The paper stands in a good structure, but revisions are needed as suggested below before it is accepted for publish in ACP. A major concern is on the emission estimates. No measurements of NO are presented, which is a better indicator of the NO_x emissions (since NO₂ is heavily affected by chemistry and the model-observation emission evaluation through the model-measurement comparison normally places emphasis on morning hours). It is hard to tell from Fig 5 whether VOCs are reasonably simulated or not. More comparisons on speciated VOCs (if measurements are available) would help to identify possible needs for further adjustments of the VOC emissions. In addition, a more detailed and clearer classification of the O₃ pollution pattern is needed.

(A1) Thanks. Since the concerns listed here are expanded later as specific comments, we respond to each individual specific comment in the following instead of here.

Specific comments: The comments are listed in the order of appearance in the paper. Several comments may converge to a same issue (such as emissions and PA analysis).

(Q2) P26839, regarding the emission inventory (EI). Since the emission data is a major input to and a major uncertainty in the CTM modeling, and modeling results and conclusions can be altered by the emissions, it is necessary to briefly describe how the inner-domain high-resolution EI is constructed, and discuss its uncertainty. Also specify the resolution of the TRACE-P EI.

(A2) The emission inventory (EI) used for inner domains was projected from a 2001 base year inventory (HKEI) prepared/compiled by the Hong Kong Environmental Protection Bureau (HKEPB). This 2001 base year inventory has been used in the Pollutants in the Atmosphere and their Transport over Hong Kong (PATH) model system for air quality modeling over PRD, though focusing on Hong Kong (Huang et al., 2005, 2006). Four major source categories are reported: point sources (mainly power gen-

eration and industrial sources), mobile sources (on-road and off-road vehicles, marine traffic, and aircraft), anthropogenic area sources (e.g., domestic and commercial fuel combustion, industrial processes, solvent evaporation loss, storage and transport of petroleum products, and agricultural activities) as well as biogenic sources.

The projection from 2001 to year 2004 was based on the growth of local economy and traffic population in Guangdong province (including PRD and non-PRD areas). In addition we added sources that are missing from 2001 EI or new for 2004, especially from Guangdong province.

Uncertainty in this 2004 EI mainly comes from the lack of local representative emission factors of VOCs from industrial and domestic solvent use and limitation on detailed information of open biomass burning emissions. However, the EI has been developed continuously for several years by the joint efforts of HKEPB and Guangdong Provincial Environmental Monitoring Center (GDEMC).

In addition, on page 26839 line 5-6, we have specified the resolution of the TRACE-P EI as the following:

“The TRACE-P anthropogenic emissions inventory (Streets et al., 2003) with a $1^{\circ} \times 1^{\circ}$ resolution is used for the outer 36-km domain.”

(Q3) P26839, L8: specify the “some results”. L18: specify what databases are input to BEIS (landuse, vegetation leafmass distribution. . .), and it's time and spatial resolution. What meteorological input is used to drive BEIS?

(A3) We have compared our emission inventory to the following emission estimation studies that covers the Guangdong province though in different years: Cao et al. (2005) developed an emission inventory of biomass burning over China for the year of 2000, including the emissions of SO₂, NO_x, NH₃, CH₄, CO, VOCs, EC and OC from crop residue, firewood, forest and grassland burning; Song and Xie (2006) developed a vehicular emission inventory in China for the year of 2002; Liu et al. (2008a) reported

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an anthropogenic VOCs emission inventory in China for the year of 2000. This process helped us to remove some suspicious emissions estimates.

For the biogenic emission, we replaced the 1993 land use data set from the U.S. Geological Survey (USGS) with the 2002 land use data sets with the 30m resolution over PRD area (derived from the Landsat TM Imagery). This dataset better represent the distribution of various land use (urban area, cropland, grassland, various forests, water, etc.) in the early 2000s in PRD. This updated land use data set and the default BEIS3 emissions factors (containing leaf area index, dry leaf biomass, normalized emissions flux for ISOP, OVOC, MONO and NO) are used as inputs to BEIS. The hourly meteorological fields (temperature and ground level radiation) are used to drive the BEIS.

(Q4) P26839, Ls19-25 and Table 1: The VOC/NO_x ratio in the anthropogenic emissions is very low (~ 1.5) compared to many urban areas around the world, which alone suggests the O₃ formation is VOC-limited according to the rule of thumb (even though it is dependent on the VOC composition). This ratio is lower than that in Los Angeles in 80s and in Mexico City in 90s. Any idea why the ratio is so low? How about it compared to other cities in China, say Beijing and Shanghai? It would be helpful to add the mean biogenic emissions of NO_x and VOCs during the model period in Table 1, which gives the reader an idea the relative contribution of biogenic emissions.

(A4) PRD is one of the most urbanized and industrialized regions in China. The power plants and mobile sources contribute a large amount of NO_x emissions, resulting in very low VOC/NO_x ratio in PRD. The VOC/NO_x ratio in PRD is within the same range as those reported in other areas in China. Based on the TRACE-P EI for the year 2000, the VOC/NO_x in the anthropogenic emissions is ~ 1.7 for Beijing, ~ 1.0 for Shanghai and ~ 1.9 for Guangdong province. By the INTEX-B anthropogenic emissions for the year 2006 (Zhang et al., 2009), the VOC/NO_x ratios were ~ 1.5 for Beijing, ~ 0.9 for Shanghai and ~ 1.2 for Guangdong province. Wang et al. (2010) reported the pollutant emissions in Beijing in the summer of 2008. The ratio values were ~ 2.1 in June 2008 and ~ 1.7 during the Olympic Games. According to the study by Zheng et al. (2009),

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the anthropogenic VOC/NO_x emission ratio is ~1.0 over PRD in 2006.

The mean biogenic emissions of NO_x and VOCs in October 2004 are added into Table 1 for comparison with anthropogenic emissions.

Table1. Summary of annual emissions of NO_x and VOCs by source category over the PRD in 2004 (kilotons/yr) (See the supplement).

(Q5) P26840 Ls3-20: There are not descriptions of the measurement techniques for O₃ and NO_x (and maybe CO too, see Comment #7). In addition, explain why C2-VOCs are not included when compared with the on-line observation.

(A5) The measurement techniques for O₃ and precursors are detailed in Zhang et al. (2008b), and more observed parameters at the super sites (GDEMC and Xinken) are reviewed in Zhang et al. (2008a). We added the following words and citations as well to describe the O₃ and NO_x measurements in section 2.2:

“The levels of O₃ and NO_x were measured by TECO commercial instruments TECO 49C and 42C, respectively, at the super sites and stations of PRD air quality monitoring network (Zhang et al., 2008b).”

Due to only NMHCs from C3 to C12 were measured by the online technique (Wang et al., 2008), the simulated C2 species are not included in the comparison between the simulation data and the on-line NMHCs observations.

(Q6) P26841, PA analysis and relevance in Section 3.3: The PA analysis is made on a single grid cell for each of the three sites. The three sites are selected to represent the areas, by my understanding, where O₃ formation is mainly affected by local emissions (GDEMC), mainly affected by transport (Xinken), and comparably affected by both local emissions and transport (Donghu). The problem is that the single cell results may not be representative to these three area's conditions, in particular if a grid cell is heavily influenced by point or uncharacteristic highly localized emission sources. It would be more representative by including more surrounding cells in each of the grids.

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(A6) Yes, we select these three sites to represent three different typical receptor areas. The GDEMC site, locating at urban core of Guangzhou city, is heavily influenced by local sources. The conditions would not be significantly changed even though more surrounding cells are included because intensive precursor emissions distribute over the urban Guangzhou and the urban Foshan nearby as illustrated in Fig. 2. The Xinken site sits at the rural coastal area with less local emissions distribution in the grid cell in which Xinken locates and in its surrounding cells. Although Donghu is an urban site, it is not as heavily influenced by local emissions as the GDEMC site. Another reason to select single grid cell for PA analysis is to well match the observations at these three sites. GDEMC and Xinken are super sites specially set up for the campaign, and Donghu site observed the maximum O₃ levels during this period, the analysis at the exact grid cells for the sites will help obtain more insight on the O₃ formation at the selected sites.

(Q7) P26843, Table 3, add performance statistics for NO₂ (and NO) and NMHCs. Fig. 3, replace the time series at an unmentioned site, such as Huijingcheng, with Foshun. The Foshun data may help to illustrate the Category #3 pollution pattern.

(A7) We added the similar statistics for NO₂ and NMHCs evaluation in Table 3. In addition, we removed the cut-off of 40 ppb for the O₃ performance statistics. For more detailed discussion on the removal, please refer to the response to the second question of reviewer #4. The Huijingcheng site is exactly the air quality station located in Foshan city (see Fig. 1 of the manuscript), which reflects the O₃ pollution in Foshan.

Table 3. CMAQ performance statistics for the simulated hourly concentrations of surface O₃, NO₂ and NMHC against observations over the PRD during 4–31 October 2004 (See the supplement).

(Q8) P26843 Ls9-17 and Figs 4-5. First, from the way presented as in Fig. 5, it is difficult to tell if NMHCs are reasonably predicted or not, particularly when the canister samples have coarser time resolutions (3-hour span or whatever) than that of the model

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output. It would be more appropriate to use scatter plots. Second, it would be desirable to provide or discuss the comparisons of speciated VOCs. These comparisons may direct to a need for the adjustment of the emissions. This is important because the conclusion of the sensitivity study could be changed. Third, as mentioned in the general comment, it would be necessary to include NO comparison if observations are available. If the NO comparison is included, since the NO_x emission evaluation would be focused on morning hours when vertical mixing also play a critical role in determining a species's concentrations, CO (or other chemically relatively inert species) data may be needed to examine the vertical diffusion.

(A8) We replaced the time series plots in Fig. 5 with the scatter plots for comparisons of the simulated and observed NMHCs (see the supplement). The statistics for NMHCs evaluation are presented in Table 3. Based on the comparisons of speciated VOCs, the under-predictions in alkanes and aromatics account for the majority of the deviation in simulated NMHC. The simulated NO show more negative bias against the observations than that of predicted NO₂. Therefore, we added the followings in Section 3.1 to extend our discussion on the deviations between model results and measurements of O₃ and precursors:

“The measurements of NO_x and VOCs are greatly influenced by local emissions. October is harvest season for agricultural crops in PRD and biomass burning in open fields was significant and was observed during the campaign. Although the emissions by burning crop residues were considered in the inventory, an exact estimation of precursor emissions was difficult due to the limited information on the details of burning events (i.e., place and duration of biomass burning, amount of burned crop residues), which resulted in the uncertainties in the simulated precursors concentrations and O₃ levels as well, especially at rural sites (e.g., Tianhu, Wangqingsha and Xinken). Another important uncertainty in estimating VOCs emissions comes from the lack of local representative emission factors of VOCs from industrial and domestic solvent use, which also accounts for the under-prediction of ambient VOCs concentrations, espe-

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cially for aromatic levels. Nighttime vertical diffusions are not easily simulated by current mesoscale meteorological models, especially over the complex topography and land use in PRD, which is an important reason for higher deviations of precursor levels between model results and measurements during night and early morning. In addition, observations reflect the on-site levels of ambient pollutants, whereas the model results represent the concentrations averaged in a grid cell with a horizontal resolution of 4 km, different spatial resolution is also a factor causing the deviation of simulations from observations.”

(Q9) P26845-26846 on classification of the simulated O₃ pollution patterns. Couple issues here. First, the description presented lacks clarity, particularly for Categories #2 and #3, both formed due to the interaction between a weak synoptic circulation and the sea-land breeze, but hard to tell how they differ, which may lead to confusions. An example of the resulting confusion is indicated in Fig 11, where both Day 16 and 22 seem to belong to Category #2, but at Table 4 Day 22 is put in Category #3. I understand that a comprehensive classification would need a separate paper, but the authors should present a bigger and clearer picture about the synoptic flows and local circulations (and not just the surface flows), how they interact and evolve that lead to the different patterns. Second, are the simulated patterns consistent with the real world? The latter is more important in the context of air quality and health effect. In another word, upgrading from the “simulated pollution pattern” to the “pollution pattern” would be more relevant. Third, name the three categories that characterize the pollution, such as O₃-South (O₃S), O₃-Southwest (O₃SW) and O₃-Northwest (O₃N), including at Table 4.

(A9) The related three questions are responded as follows.

(1) Classification of O₃ pollution patterns:

The classification of the simulated O₃ pollution patterns proposed in the manuscript aims to summarize the spatial distribution of elevated O₃ and to pick out the repre-

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sentative days or short periods for detail analysis of surface O₃ evolutions. Two major aspects are considered for this classification: the first is the regions where elevated surface O₃ occurred in the afternoon; the second is the feature of near ground flows. For Category #1, although high O₃ sometimes occurred in the southwestern inland and coastal areas, similar as the distributions of Category #2, no influence of sea breezes differentiates Category #1 from the other two categories.

The differences between Category #2 and #3: (1) Flow conditions: the daytime near ground flows over inland areas were dominated by northeasterly for Category #2, whereas easterly (including winds from east-southeast, east and east-northeast) for Category #3. (2) The O₃ pollution in Jiangmen and southeastern coastal areas: for Category #2, the high levels of O₃ in the afternoon mainly distributed over southeastern Foshan, Jiangmen and southeastern coastal areas, Jiangmen usually experienced a heavy O₃ pollution; for Category #3, the elevated O₃ mainly occur over Foshan and Zhaoqing, Jiangmen experienced a relatively light O₃ pollution in the afternoon, especially along the southwestern coastal areas were found less O₃ pollution.

Both Category #2 and #3 are formed due to the interaction between a weak synoptic circulation and the sea-land breeze, differ from each other driven by the daytime inland flows with dominant directions of about 45 degrees difference. It could be understood that elevated O₃ distributions on several days of Category #3 have not significant differences to those on days of Category #2, e.g. the mid-afternoon O₃ distribution on Day 22. If Day 29 is taken an example of Category #3, we would notice a remarkable difference in O₃ distribution against those on Day 16 (see Fig. 7). Day 22 is selected for process analysis in Section 3.3 due to the following considerations:

First, Day 22 is one of the consecutive seven days (16-22 October) selected for process analysis. During this selected period, the PRD experienced all three O₃ pollution patterns and the CMAQ simulation shows good performance at most sites.

Second, Over Jiangmen city, especially along the southwestern coastal areas, the O₃

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levels around mid-afternoon illustrated a more severe O₃ pollution on Day 16 than that on Day 22.

Last, Fig. 12 also presents some interesting differences in NO_x transport over the depth of layers 1-7 between Day 16 and 22. The accumulated transports of NO_x from 17:00 to 9:00 LST of the next day had a similar feature with high contributions in central Jiangmen (center figures of Fig 12a and 12c). However, after transports of six more hours (from 9:00-15:00 LST), the transported NO_x mainly distributed over eastern and southern Jiangmen on Day 16 (right figure of Fig. 12a), while high contributions of transport shifted to northern Jiangmen and southern Foshan on Day 22 (right figure of Fig. 12c), which reveals the different influences of flow conditions over layers 1-7 between Category #2 and Category #3.

(2) Consistency with the real world:

The authors agree on the reviewer's comment. To evaluate the consistency of the simulation with the real world, a comparison was made between the simulated O₃ and the observed ones collected at a monitoring network covering the majority regions of PRD. The comparison showed the simulation could reproduce the pollution pattern observed over PRD. Moreover, the high levels of O₃ simulated over Jiangmen and western Foshan would imply that more attentions should be paid on western PRD in the fall season (a typical period for the most severe O₃ pollution in a year in PRD). Also, it would be suggested that the existing monitoring network should be expanded to cover western PRD, so that more data can be collected for further study in this sub-region where O₃ pollution has not been adequately addressed so far.

(3) Name of the three categories:

The categories are named as O₃-South (O₃S), O₃-Southwest (O₃SW) and O₃-West (O₃W) for Category #1, #2 and #3, respectively, and included at Table 4.

(Q10) P26846-26847 on model layers and time used in the PBL process analysis. First,

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it would be more suitable to use simulated time-varying PBL height for the PA analysis of O₃ production in the boundary layer. The 7 layers used do not represent well the PBL height most time of the day. Second, in different pollution patterns that reflect the interaction of synoptic and local circulations, the contributions to O₃ formation of each process may vary significantly, particularly for the transport process. It would be more appropriate and insightful by conducting the PA analysis under different categories and summarizing the results. Averaging over the three patterns might smooth out and mask some important results.

(A10) (1) Model layers used in PBL process analysis:

The time range from 9:00 to 15:00 local standard time (LST), during which daytime maximum O₃ levels are built up in the PBL, is focused by our process analysis.

On P26846-26847, when discussing the O₃ formation at the three selected sites (GDEMC, Xinken and Donghu), we present the contributions of different processes from layers 1 to 8 (shown in Fig. 9), which illustrates the vertical variations of roles of atmospheric processes under the PBL. From Fig. 9, the net O₃ change and the contributions of chemical process in the 8th layer (corresponding to height from 1000m to 1500m) are significantly lower than those in other layers at all the three sites, which suggests that the 8th layer could be upper boundary for the PBL and the results in layers 1-7 have a reasonable representation of the O₃ evolution in PBL during 9:00-15:00 local time. Fig. 8 shows the process analysis results in the lowest three layers (0-80 m, approximately corresponding to the vertical height of monitoring sites), which is used to provide information of near ground O₃ evolution.

When applying process analysis on regional O₃ pollution (P26848-26849), we use the integrated values over the depth of layers 1 to 7 to discuss the influences of atmospheric processes on O₃ levels and analyze the transport of NO_x during nighttime over PRD. In nighttime, the transports of NO_x emitted by elevated point sources usually occur above the PBL, the analysis in the fixed 7 layers, including both NO_x within

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PBL and those above, could well describe the physical transport of NO_x from source regions to downwind areas, which plays critical roles on the NO_x supply for daytime O₃ photochemical production over the PRD region, especially in downwind rural areas. Although the fixed 7 layers do not represent well the PBL height most time of day, the analysis within the lowest 7 layers helps to explain the occurrence of the elevated O₃ around mid-afternoon, and reveal the close interactions among precursor emissions, transports and O₃ photochemical production over PRD. The analysis under the time-varying PBL height is an alternative way to investigate the high O₃ formation in afternoon, which provides an exact picture in the PBL (the fixed 7 layers are an approximation to the height of PBL from late morning to afternoon). However, by analyzing within the time-varying PBL, we might find vertical NO_x entrainments with the rise of PBL height in the morning, but might not find direct clues to identify whether the entrained NO_x is emitted by local sources or transported from other areas.

(2) Time used in PBL process analysis:

We have conducted the day-by-day PA analysis to investigate the roles of various processes on O₃ build-up from 9:00-15:00 LST from October 16 to 22 (same Figures as shown in Fig. 9), but the results on days under different categories share much common features, and the average results are given in Fig. 9 to illustrate such common features. In addition, Fig. 8 also shows the daily variations of O₃ concentrations and the contributions of various processes in near ground layers, although PA results illustrate some difference among these days, the common features in days under different categories are very clear and the major of the diurnal variations could be well represented by the mean one (also shown in Fig. 8).

(Q11) P26848 Ls13-14 “the chemical process dominated the O₃ enhancement from morning to mid-afternoon” is contradictive to P26846 Ls17-18 “the chemical process exhibited a significant consumption of O₃ during the whole day”; positive CHEM term at Table 5 and in Fig 9 is also contradictive to Fig 8 where CHEM is negative throughout the day at GDEMC.

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(A11) P26846 Ls17-18 and Fig 8(a) discuss the roles of chemical process on O₃ evolution in the lowest three layers (corresponding to the height of 0~80 m), which shows a chemical consumption in layers near ground. The accumulated contributions of chemical process (CHME_O₃) from 9:00 to 15:00 LST at GDEMC, shown in Fig. 9(a), also present consistent results (i.e., negative contributions) at layers 1-3.

In P26848 Ls13-14 and Table 5, the layers 1-7 are taken as a whole to represent an overall condition in the PBL during 9:00-15:00 LST, and the integrated values over the depth of layers 1 to 7 are used in discussion. Intensive local NO_x emissions at GDEMC result in the chemical consumption of O₃ in lower layers, but chemical O₃ production in upper layers (see the CHEM_O₃ shown in Fig. 9(a)), which suggests a quite different role of chemical processes at different heights of the GDEMC site. If we take the overall contribution of chemical processes over the depth of layers 1 to 7 for analysis, the chemical processes are dominant contributor for O₃ enhancement at GDEMC for the 6 hours.

(Q12) Fig 16. The data points are so intense that they mask each other. Consider diluting the data points. Describe the sites Kaiping and Duanfen in Page 26852 (not just in the figure caption).

(A12) We have re-drawn Fig.16 and added the descriptions of the sites Kaiping and Duanfen in Page 26852 as the following:

“Kaiping and Duanfen are located in the central and southern Jiangmen city, respectively (see Fig. 1), which are selected here to investigate O₃ photochemical sensitivity in the southwestern inland and coastal areas of the PRD.”

(Q13) P26852 Ls13-16. This sentence seems not right. According to Fig 16, delta-P(O_x) decreases with increasing NO_x/NO_y in the VOC-reduction case, but opposite for the NO_x-reduction case.

(A13) The former expression of P26852 Ls13-16 is not clear. The sentence has been

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revised as the following:

“In the case of 25% reduction of NO_x emissions, the percentage change in P(O_x) tends to increase from about -30% to 30% with increasing NO_x/NO_y ratio, whereas a decreasing tendency is obtained in the condition of VOCs emission reduction.”

Technical Comments:

(Q14) P26839 L8, personal communication, misses the contact person's name(s).

(A14) We added the contact person's name, Dr. Peter Louie, for the communication on PRD local emission inventory.

(Q15) P26839, L18: add a reference for the Chinese plantation survey dataset.

(A15) Reference for the Chinese plantation survey dataset:

Pearl River Delta environmental protection planning committee: Pearl River Delta environmental protection planning, China environmental science press, Beijing, 2006.

(Q16) P26851 L12, change fewer to less.

(A16) Corrected.

(Q17) Fig 14, explain the ellipses in the figure caption

(A17) The following is added in the figure caption to explain the ellipses:

“The blue and red ellipses mark the regions with the O₃ change characterized by NO_x-limited chemistry and by VOC-limited chemistry, respectively.”

References:

HKEPD: Study of air quality in the Pearl River Delta Region: Final report, Agreement No. CE 106/98), 2002,

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/9/C11897/2010/acpd-9-C11897-2010-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 26833, 2009.