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Atmospheric Chemistry and Physics Discussions

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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Response to Reviewer #1

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

General comments:

(Q1) The article investigates, with a 3-D air quality modeling system, ozone pollution events in the Pearl River Delta region. Data of a one-month field campaign are used either for initializing the model for the meteorological fields or for evaluation of the

modeling system's performance. Three evolution patterns of elevated ozone are distinguished depending on differing near-ground flow conditions. An integrated process rate analysis is applied to determine the main contributors to the elevated ozone levels. The article investigates the interactions between precursor emissions, transport and ozone photochemical production. In addition, sensitivity studies reveal areas within the Pearl River Delta, which are either VOC- or NOx-limited.

This paper fits into the scope of ACP. It deals with the modeling of tropospheric ozone pollution, including precursor emissions, transport, gas-phase chemistry, and the meteorological conditions over the greater region of the Pearl River Delta, one of the regions in south-east Asia with very bad air quality. Investigating in detail the underlying mechanisms is not only of regional, but also of general interest. The integrated process analysis and the sensitivity study with either reduced VOC or NOx are the strong points of the paper, together with the use of observational data of a comprehensive one-month field campaign. The results are presented in a clear and well structured way. I recommend publication in Atmospheric Chemistry and Physics after consideration of the following comments.

(A1) Thanks for your general comments.

Specific comments:

(Q2) In the introduction I am missing some references to studies for comparable Megacities or regions. Are there comparable studies, what was investigated and found?

(A2) We added the following to the end of the first paragraph in Section 1 to cite several comparable studies that investigate O3 problem in Mega-cities or regional scale:

"The O3 pollution in megacities and regional areas is investigated recently by many studies (Evtyugina et al., 2006; Goncalves et al., 2009; Lei et al., 2007; Kimura et al., 2008; Yu et al., 2009; Chang, 2008; Ran et al., 2009; Wang et al., 2009), in which the topics addressed include O3 photochemical production, chemical sensitivity to pre-

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cursors, roles of meteorological conditions, O3 source attributions and development of control strategy."

(Q3) In section 2.1, model setup and inputs, the authors mention that observations in the ABL during PRIDE-PRD2004 were used to prepare the initial and boundary conditions for the MM5 meteorological simulations. Later on, in section 3.1, evaluation of model performance, it is stated that simulated surface meteorological fields are examined against surface hourly observations made during the field campaign. Could the authors clarify which observations were used as model input, which ones for evaluation?

(A3) (1) Boundary conditions (BCs): All the available meteorological observations in the ABL during PRIDE-PRD2004 (including pressure, wind speed and direction, temperature and humidity) are used to prepare the BCs for the 36-km coarse domain; whereas the BCs for the 12-km domain are derived from the output of the 36-km domain, and the BCs for the 4-km domain are based on the results of 12-km domain. (2) Initial conditions (ICs): The entire month of October 2004 is divided into six separate periods for MM5 simulation, each simulation with 5- or 6-day running, and separate ICs are prepared based on all the available meteorological observations during the campaign for each period and for each nested domain.

The MM5 performance in the 4-km domain is evaluated by using all the surface hourly observations over PRD in the whole October (including wind speed and direction, temperature and humidity). Although the same observations are used as modeling inputs and for model evaluation, only an indirect influence (via BCs) and a time-limited influence at the beginning of simulations (via ICs) occur when evaluating the consistency of the simulated meteorological fields and the observed ones.

Generally, the observation nudging technique has influence on the MM5 performance. In our simulation, the observational data collected during the campaign have been weaved into 3-dimensional (3-D) reanalysis data, then used in the 3-D analysis nudging

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for winds, temperature and humidity at six hours intervals, and in the surface analysis nudging for winds only at three hours intervals. However, the applied three/six hours intervals would limit the influence of analysis nudging on performance evaluation which is based on all the hourly observational data.

(Q4) In section 2.2, model evaluation protocol, the monitoring network is described, and in particular the measurement techniques for NMHC. However, I am missing some description or reference for the NO2 observations, which are discussed in section 3.1 and in Figure 4.

(A4) We added the following and a reference as well to describe the NO2 measurements in section 2.2:

"The levels of NOx were measured by TECO commercial instruments TECO 42C at the super sites and stations of PRD air quality monitoring network (Zhang et al., 2008)."

(Q5) In section 2.5, ozone sensitivity testing, it is described that the emissions of VOCs and NOx, respectively, were reduced by 25%. Are these reductions linear over all emission sources, or have some been more reduced, some less?

(A5) The 25% reduction in the NOx and VOC emissions are only applied on the anthropogenic emission sources, no changes to biogenic emissions. And the reductions are linear over various kinds of anthropogenic sources, all reduced with the same percentage, 25%.

(Q6) In section 3.1, evaluation of model performance, the authors describe the evaluation statistics for simulated ozone concentrations. Is it possible to give similar statistics for NOx and NMHCs or to include them in Table 3?

(A6) 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.

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

(Q7) In section 3.3, process analysis of ozone formation, the authors explain that their process analysis focuses mainly on the ABL. However, it would be interesting if there are also informations about possible impacts of entrainment of precursors (or even ozone) from the free troposphere, or long-range transport of precursor emissions during the measurement campaign in October 2004.

(A7) The ozone formation under ABL is of the most interest in this manuscript. When we discuss the contributions of transport processes to O3, the results used here are the net influence, including the impacts of physical transports in ABL and those from the free troposphere. The current integrated process rate (IPR) technique implemented in CMAQ are not ready to quantify the individual impacts of pollutant entrainments from the free troposphere. The northern areas to PRD are Nangling mountains, which separate PRD from the northern mainland areas, resulting less influence of long-range transport from northern areas, especially during the campaign period with less strong cold air intrusions from the north. The transport of precursors identified by IPR is also an overall effect, including the contributions from the source emissions inside the PRD as well as those from outside PRD by long-range transport. Updating the IPR technique to include more flexible analysis or applying other techniques to quantify the influence of entrainments from free troposphere and of long-range transport from outside PRD are beyond the scope of this work.

(Q8) At the end of section 3.4, ozone production efficiency, the values found for OPE over the PRD are also compared to values found for Beijing, US cities and the Mexico City Metropolitan Area. However, for the latter two no values are given, only for Beijing. Numbers for the other two cases would be interesting.

(A8) We added the values of ozone production efficiency found in US cities and Mexico

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City Metropolitan Area as the following:

"The simulated OPEs at the super sites are comparable to those observed in urban Beijing, ranging from 3.9 to 9.7 (Chou et al., 2009), and those observed or modeled in U.S. cities (Nashville, 2.5–4.0, Nummermacker et al., 1998; New York City, 2.2–4.2, Kleinman et al., 2000) and in the Mexico City Metropolitan Area (4-10, Lei et al., 2007)."

Technical corrections:

(Q9) Page 26837, line 13: '... modeling studies have rarely been performed ...' Are there or not? If yes, then please cite at least one.

(A9) The studies reported by Wang et al. (2005) and Wei et al. (2007) are 3-D CTM studies over both the inland PRD region and Hong Kong. We have briefly introduced their works in the manuscript. For a clear expression, we changed the sentence in lines 13-15 as the following:

"In spite of the works by Wang et al. (2005) and Wei et al. (2007), there are still very limited studies to address these issues for O3 episodes over the inland PRD region."

(Q10) Page 26839, line 18: Please give a reference for BEIS 3.09

(A10) We have added the following reference for BEIS3.09:

Vukovich, J. M. and Pierce, T.: The implementation of BEIS3 within the SMOKE modeling framework, available at

http://www.epa.gov/ttn/chief/conference/ei11/modeling/vukovich.pdf, 2002.

(Q11) Page 26842, line 5: 'chemical process'. This term appears in the following several times (also in Figure captions), however, I find it a bit confusing as it sounds like a specific chemical process (however, not nearer specified). I suppose the authors refer to the gas-phase chemistry (CHEM) of their integrated process analysis. It would read better to replace 'chemical process' by 'gas-phase chemistry'.

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(A11) Thanks for your suggestion. The 'chemical process' is replaced by 'gas phase chemistry' for a clear expression in the context of process analysis of O3 formation.

(Q12) Page 26847, lines 1-2: Would read better written as: At the rural Xinken site, vertical transport contributed mainly to the ozone levels and was then decreased on a similar magnitude by horizontal transport during the daytime (Fig. 8b).

(A12) Accepted and revised.

(Q13) Page 26848, line 24: '... busy harbors are located ...'

(A13) Accepted and corrected.

(Q14) Page 26849, line 26: '... on the other two days.' From the text alone, it is not clear which days (21/22, or 15/16 October)?

(A14) We revised it as: '. . .what happened on 15–16 and 21–22 October.'

(Q15) Page 26852, lines14/15: The description of Figure 16 is a bit confusing when reading the graph from the perspective 'as a function of the NOx/NOy ratio'. It would read better like: '... the percentage change in P(Ox) tends to decrease from about 30% to –30% with increasing NOx/NOy ratio, whereas an increasing tendency ...'

(A15) The former expression of P26852 Ls13-16 is not clear. The sentence has been revised as the following:

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

(Q16) Page 26853, line 14: new sentence after '... horizontal outflow.

(A16) Accepted and corrected.

(Q17) Page 26862, caption Table 3: Please include a brief explanation what NMB and NME stand for.

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(A17) We added the brief explanations for NMB and NME below the table.

(Q18) Page 26863, caption Table 4: Could the three evolution pattern categories named? Or include anything like 'refer to text for category explanation' in the caption.

(A18) The categories are now named as O3-South (O3S), O3-Southwest (O3SW) and O3-West (O3W) for Category #1, #2 and #3, respectively, and included at Table 4.

(Q19) Page 26867, Figure 3: Is it possible to enlarge the figure for better readability?

(A19) The 12 time series in Fig. 3 are enlarged for a clearer presenting in two pages.

(Q20) Page 26875, Figure 11: Is it possible to enlarge the legend text for better readability?

(A20) We revised the legend text of the figures for better readability.

(Q21) Page 26876, Figure 12: Is it possible to enlarge the legend text and titles of the figures for better readability?

(A21) We revised the legend text and titles of the figures for better readability.

(Q22) Page 26878, caption Figure 14: There should be a reference to the corresponding text to explain what the ellipses are marking.

(A22) 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 NOx-limited chemistry and by VOC-limited chemistry, respectively."

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/9/C11878/2010/acpd-9-C11878-2010-supplement.pdf

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