Response to the Interactive comment on "Air quality and emissions in the Yangtze River Delta, China" by L. Li et al. K.-s. Lam (Referee) cekslam@polyu.edu.hk Reference No.: acp-2010-615

The paper is scientifically sound and worth publication. It presents a useful update of emission inventory for YRD and reveals the air quality of YRD using chemical transport model. In the past decade, modeling of air quality of China using 3-D mesoscale chemical transport models has been developing very fast. It is quite well known that the emission inventory (EI) of China is considerably under-estimated. There is a pressing need to compile a reasonably accurate EI especially in the China northeast industrial region, the YRD and the Pearl River Delta region. This paper reported the latest improvement of the emission inventory of the YRD region and this should be the main contribution of this manuscript. However, there are a few omissions and ambiguities. The English standard of this manuscript could be further improved.

Dear prof. Lam, we quite agree that the emission inventory is very important, and our team has kept on improving it. This paper reports the study on the air quality and emissions in the YRD in 2004, and we will report the updated results in 2007. The study in this paper outlines a general idea for the region in 2004, and will be compared to the data in 2007 in other papers.

Major comments:

(1) The English standard should be improved to remove ambiguities in a few places. A few improvements have been listed in the specific comments below.

The ambiguities that have been pointed out by the kind reviewer have been removed or clarified.

(2) There are two models used in this study – MM5 and CMAQ, the authors should clarify which model they are referring when they are reporting their inputs, boundary conditions and initial conditions in section 2. Another point is all 3 domains of MM5 are clearly defined but the domain and vertical levels of CMAQ were not mentioned.

The models are described in more detail in section 2, and the CMAQ domain and vertical levels have been added in the text. The revised text is as follows:

The CMAQ model domain is based on a Lambert Conformal map projection, using a one-way nested mode with grid resolutions of 36 km (covering the whole of China), 12 km (covering eastern China), and 4 km (covering the YRD region), as shown in Figure 1. The MM5 domain is larger than the CMAQ domain, with three grids more than the CMAQ domain on each boundary. The mother domains for both MM5 and CMAQ are centered at

(110°E, 34°N). The YRD domain for CMAQ has 118 \times 136 horizontal grid cells and includes 16 major cities, which are also shown in Figure 1. Each "city" in the domain is a large administrative area that includes smaller cities, towns, and villages. Thus, the 16 cities together cover the majority of the YRD area. The pollution episodes chosen are January 1-31, 2004, and July 1-31, 2004, which represent winter and summer seasons, respectively. The initial conditions of CMAQ for each seasonal run are prepared by running the model five days ahead of each start date with clean initial conditions (IC). Sensitivity tests show that the influence of IC generally dissipates after about three days. The boundary condition (BC) used for the mother domain of CMAQ is clean air, while the BCs for the nested domains are extracted from the CCTM concentration files of the larger domain. Both of the two models employ 14 vertical layers of varying thickness with denser layers in the lower atmosphere to better resolve the mixing height.

(3) Sections 2.3, line 3 stated that the new emission inventory consists of biomass burning and biogenic sources but no more details were given afterwards. This is a major omission.

The emissions from biomass burning are not included in this paper. For biogenic VOCs Emissions, this paper used the natural VOCs emission inventory of GEIA Global Emissions Inventory Activity 1990 (http://geiacenter.org). In July, total biogenic VOCs emissions in Shanghai were 3286 tons, taking a share of 23% of the total biogenic emissions in the YRD. It is no doubt that the data is out-of-dated, we have update these part of emissions for the year 2007 and will report the results in another paper soon. Text has been added to Section 2.3.

(4) Some discussions about the level of pollutants are stated without support; some are too subjective which makes the arguments speculative.

We will try to revise the discussions based on scientific data.

(5) The labeling of the 16 cities in the figures is quite confusing. There are two Taizhou but they are not properly differentiated in the figures.

In Chinese, the two Taizhou are "泰州" and "台州", the previous one is located in Jiangsu, while the second one is located in Zhejiang. We have differentiated them by using "Taizhou" and "Taizhou", We changed the Taizhou in Zhejiang Province to "Taizhou", and revised it in the Table 1 and Fig.1.

(6) The term "average" is used very loosely throughout the manuscript.

We will try to revise it.

Specific comments:

(1) In the abstract, only the monthly averaged SO2 is reported, it is recommended that the

modeled NO2, PM10, and Ozone should be given too.

We will add the monthly averaged NO_2 , PM_{10} and O_3 soon.

(2) Pg 2, last paragraph: the line "driven by a new, comprehensive emission inventory." should be rewritten. In this manuscript, only the YRD inventory is updated, all the other regions are not "new". Neither is the term "comprehensive" appropriate.

Text has been revised in the manuscript, and the "comprehensive" has been removed.

(3) It is mentioned in pg 3, section 2.1 that "The driving meteorological inputs are provided by MM5", presumably, the authors are referring to CMAQ input? Also what inputs are used to drive MM5?

The meteorological inputs of CMAQ are provided by MM5. The inputs for MM5 are NCEP FNL (Final) Operational Global Analysis data, which are on 1.0x1.0 degree grids continuously at every six hours. Text has been revised in the manuscript.

(4) Section 2.2, 1st line: "The model domain" should be changed to "The MM5 domain"?

It should be "The CMAQ domain", and the MM5 domain is usually with 3 grids larger at each boundary of the CMAQ domain.

(5) Figure 1: There is one city between Huzhou and Changzhou that is not labeled. Including this city, there should be a total of 17 cities. Are the emissions of this city not included?

The region between Huzhou and Changzhou belongs to Wuxi, the color of this region is the same as that in Wuxi. Thus, the emissions are included in Wuxi.

(6) Section 2.2, 2nd line: I don't understand the term three-way nested, I have heard of one-way and two-way interactions but not 3-way.

Sorry for using the ambiguities word. It's true that nesting can be accomplished by either a "one-way" or a "two-way" method. In one-way nesting, the coarse domain simulation is run independently for the nest; while two-way nesting involves feedback between the coarse and the nest domains. This paper adopts a one-way nesting method, with one coarse-resolution domain and two nest domains. (Because the CMAQ version used in the paper allows only static grid nesting). The ambiguities text has been revised in the manuscript.

(7) Section 2.2, line 10: How boundary condition is considered was not shown, and if the study only use the default BC given in the CMAQ model, it is not helpful to give an ideal and reliable initial condition only by running the model five days ahead of start date with

clean initial conditions because the default BC is also clean and without day-to-day variation.

The boundary condition (BC) used for the largest domain of CMAQ is clean air, while the BCs for the nested domains are extracted from the CCTM concentration files of the larger domain. Text has been added to Section 2.2.

(8) Section 3.1.2, 3rd paragraph: PM2.5 and NH3 emissions were stated for stationary sources but they are not stated for mobile sources. In line 6, the words "larger share" is wrong choice of words.

The particulate matters emitted form vehicles are mostly fine particles, thus the PM10 stated in the 3rd paragraph in Section 3.1.2 should be PM2.5. NH3 are mostly emitted from livestock, poultry breeding and chemical fertilizer application, which is not included in this study, and have been improved in the 2007 emission inventory. We will report this study in another paper.

(9) Section 3.2.1: Hourly concentrations for SO2, NO2, PM10 were selected during January 11-20, 2004, and July 11-20, 2004. Since the comparison of observed and model results focus on this time period, why Table 2 only assess the MM5 performance of Jan 01,02,20,25,28, and Jul 01, 02? Why these days are selected and why not shows the results of January 11-20, 2004, and July 11-20, 2004.

The specific time for MM5 evaluation are selected randomly, we will give the comparison results for January 11-20, 2004, and July 11-20, 2004.

(10) Section 3.2.2: The term "coefficients" should be defined more properly.

The term "coefficients" in section 3.2.2 has been revised to "correlation coefficients".

(11) Section 3.2.2, figure 6: Is the NCEP data FNL data? Is it reanalysis data? The source of NCEP data should be given. Also, if the NCEP data is used to drive the MM5 model (my speculation), then it is not appropriate to compare the model results with its input data. It is more appropriate to compare model outputs with observational data.

The inputs for MM5 are NCEP FNL (Final) Operational Global Analysis data, which is added to manuscript. The NCEP data is not the real observational data. Since we don't have real observational meteorological data for the simulation period, we have to use NCEP data instead. We realized that the observational data should be applied to do the MM5 model evaluation and FDDA, and we will try our best to improve this point in the future study.

(12) Figure 7: the labels of the x-axis basically have no meaning.

We will modify the label of x-axis to the exact date.

(13) Section 3.2.3, pg 8, line 7: The phrase "true SO2" is a strong phrase. Suggest to replace the phrase by "reflect the general SO2 concentration in the YRD region."

The "true SO2" has been changed to "reflect the general SO2 concentration in the YRD region."

(14) Section 3.2.3, figure 8: The legend of figure 8 stated monitoring average and model average. There are a few monitoring stations in Shanghai, how the monitoring results are averaged? Also, how the model results are averaged? Which grid (all grids in Shanghai) or which vertical layers (only surface layers or all layers in BL) are included?

The finest grid resolution is $4\text{km} \times 4\text{km}$, with the grid area of 16 km^2 . Some observational sites are quite close and are within one grid, as is shown in fig.5. Thus, we give the monitoring max, min and average values of the monitoring data at several sites within one grid, to be compared with the model result. The average model result means the data of that specific grid which contains those observational sites. And the vertical layer is only the surface layer. Text has been clarified regarding to discussion on fig.8 in Section 3.2.3.

(15) Section 3.2.3, NO2: "the NO2 simulation results are not as good as for SO2". After looking at figure 8 and figure 9 and Table 3 as well, I quite disagree with this statement. Table 3 shows that the hourly average of NO2 in both January and July are quite similar. More information should be given to indicate why NO2 simulation is not as good as SO2.

Pollutants	$SO_2 (mg/m^3)$		$NO_2 (mg/m^3)$	
Item	Obs	Model	Obs	Model
Hourly Average	0.04	0.05	0.04	0.04
Max.	0.19	0.32	0.12	0.12
Min.	0.00	0.00	0.01	0.00
R ² correlation coefficient	0.6		0.65	
Index of agreement	0.67		0.77	
Bias	0.19		0.26	
Factor of two	0.67		0.81	

We averaged the indicators of Jan and July, only to get the following table. Therefore, we agree with your view and have revised the text in Section 3.2.3.

(16) Section 3.2.3, PM: "Results show that CMAQ can reflect the trends of PM10 in January, while in July the model tends to underestimate the PM10 concentrations." My reading of Figure 10 indicates the opposite. The model underestimated PM in January in both Nantong and Ningbo.

Sorry, the description is opposite. As is shown in Table 3, the Bias of PM10 in Jan is -24%, and 26%. The text in Section 3.2.3 has been revised.

(17) Section 3.2.3, Factor 2 analysis: Since the MRF scheme is not satisfactory, then why not use another scheme?

Although MRF scheme is not perfect, it is the best we can choose. More description and related references will be added to the manuscript.

(18) Section 3.2.4, Equation 7: I think there is an error in equation 7. The symbol oi is not defined. The i should be a subscript. Also bar o is defined as average observed ozone concentration, the word "ozone" should be deleted?

Right. It should be "oi" instead of "oi", and word "ozone" should be deleted.

(19) Section 3.2.4, pg 11, line 9: I disagree that "generally wind speed is low in winter" in YRD. Wind speed can be quite high during winter monsoon season in East China.

Text regarding to the wind speed descript has been revised.

(20) Section 4.1, line 4: "The NO2 subsequently reacts with O3 and reduces the O3 concentration." This sentence is over simplified and needs to be rewritten.

This sentence is changed to: "In the clean atmosphere away from its sources, NO represents generally less than one third of NOx during the day, and is rapidly titrated after sunset by ozone. In contrast, in the polluted atmosphere, such as in urban areas, where NO is constantly emitted from the surface, ozone is slowly titrated by NO throughout the night, forming high NO2 mixing ratios." Text has been revised in Section 4.1.

(21) Figure 14: The title needs to be changed. A scatter diagram cannot reveal the relationship between O3 and NO2, it can only show its correlation. The corresponding discussions in section 4.1 should be corrected as well.

The Fig.14 title "Relationship between monthly average concentrations of O3 and NO2 in 16 cities in the YRD" has been changed to "Correlation between monthly average concentrations of O3 and NO2 in 16 cities in the YRD". Text in Section 4.1 regarding to the discussions of this fig has been revised accordingly.

(22) Figure 15: Specific selection of July 5 and not other days should be justified. It would help the interpretation if wind arrows are added to this figure.

On July 5, the maximum hourly ozone concentration is the highest during the modeling period. Thus, we chose this day to illustrate the formation and dissipation of ozone. In

section 4.1, we have added the following sentence to justify selection of the specific day. To illustrate the formation and dissipation of ozone, Figure 15 presents the variation of hourly O3 concentration on July 5, 2004, when the highest hourly ozone concentration occurs during the modeling period.

We will add wind arrows to fig 15.

(23) Section 4.1, 2nd paragraph: The sentence "high O3 gradually diffuses from Zhoushan" should be re-written. The description is over simplified, apart from advection, there could be dispersion and photochemical production. Furthermore, the high O3 first appear over Zhousan is very interesting but there is little discussion about this ozone evolution. Figure 15 indicates that Zhoushan is a coastal area. Under southeasterly winds, there is no land based emission sources upwind of Zhoushan, why there is considerable O3 over Zhoushan first?

This is a very good question. We suppose that maybe the boundary conditions also influence the occurrence of this interesting phenomenon, however, we need to check the simulation result for the 12km grid resolution area for the same day. More discussions will be added after we look more deeply into the model results.

(24) Section 4.2, equation 8: definition of extinction coefficient should be given.

The sum of the scattering and absorption coefficients is called the extinction coefficient. Text has been added in the manuscript.

(25) Figure 19: One city label is missing in Figure 19. Figure 19 basically duplicate Figure 13. Also, number of cities in Figure 19 is different from that in Figure 18.

Revised.



Fig. 19. Monthly average concentrations of NO2 in the 16 cities of the YRD.

(26) Why monthly average of dcv is presented and discussed instead of PM10?

We suppose that dcv can better reflect the composite pollution instead of PM10. However,

we agree that the PM10 results should also be given together with SO2 and NO2.

(27) In the conclusion, line 1, "first time" is a sensitive phrase. Recommend to delete "for the first time".

The "first time" has been deleted.

On behalf of all the co-authors, I would like to thank Prof.Lam for your time on this manuscript and the valuable suggestions made, which not only help greatly to improve this paper but also help us to get more improvement in our future study.

Thanks and with best regards,

Li Li On behalf of all the co-authors.