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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The study of emission inventory on anthropogenic air pollutants and VOC species in the Yangtze River Delta region, China" *by* C. Huang et al.

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RC C1388: 'reviewer's comments', Anonymous Referee #3, 31 Mar 2011

1. P955, L5: any reference for the satellite picture?

Re: The satellite data comes from GOME. We will insert the reference in the revised manuscript. In Section 2.1, Page 955, line 7: ". . .. . . based on the measurements of the Global Ozone Monitoring Experiment (GOME) on the ESA European research satellite ERS-2 (http://www.temis.nl/airpollution/no2.html)."

2. Sections 2.3 and 3.5 The emission factors used in the inventory were generally those already compiled by Zheng et al. (2009). In order to add new information to this compilation, it is recommended to develop the discussion on the uncertainty associated with these emission factors; especially for the emission factors which originate from US or European studies and for EF which are associated with high activity numbers (because they will have a significant weight in the total emission). For the EF which have been determined in China, and which have equivalents in US and/or Europe, would it be possible to give the range of differences? (i.e. they differ by a factor of 2? 3?...more?, it is of course source dependent but this could be given for the main sources).

Re: We agree with the reviewer's recommendation. The emission factors we use to compile emission inventory are still limited. To better illustrate the selection of the emission factor, we would like to supplement some discussions and reviews on the relative literatures by re-writing the following sections: (1) Re-write the section 2.3.1, page 956, line 21 to page 957, line 7: "For SO2 emissions, the annual inventory was compiled using the mass balance method by Equation (2). The emission factors of NOx, CO, PM10, PM2.5, and VOCs were taken directly from the latest literature. Domestic measurements or relative studies in recent years are prior to be cited in this study. Foreign studies in line with current technology level in China will be considered when there is no domestic study available. The literature reviews of previous studies obtain a range of 2.38-10.0 kgâĂćt-1 for NOx emission factors (Kato and Akimoto, 1992; Hao et al., 2002; Streets et al., 2003). To better understand the NOx emission level of coal-fired boilers in China, Tian (2003) studied more than 100 power plant boilers and obtained a mix of NOx emission factors of various boiler types with or without LNB. Based on Tian's study, Zhang et al. (2007) predicted the NOx emission factors of coal-fired boilers for 1995-2004 in China, which we believe are more representative to current NOx emission level of the facilities in the YRD region. Previous studies illustrate the big differences of CO emission factors between different types of combustion facilities. Ge et al. (2001) obtained the average CO emission factor (15 kgâĂćt-1) of automatic stoker

furnaces based on the measurements. While the CO emission factor of hand-feed stoker furnaces could be 7 times higher according to USEPA's study (2002). Zhang et al. (2000) tested 19 household stoves and obtained a range of 19-170 kgâĂćt-1 for CO emission factor. Based on these studies, Wang et al. (2005) summarized the CO emission factors of each kind of fuel combustion source in China, so we cited the CO emission factors from the study of Wang et al. (2005). PM emission factors usually depend on ash content in coal, boiler technology, and the efficiency of exhaust control. There is little PM emission measurement study in China. Zhang (2005) summarized the domestic and foreign studies from USEPA (2002), Klimont et al. (2002), and Zhang et al. (2002) and reported a group of PM10 and PM2.5 emission factors which we listed in Table 1 and 2 according to the technology level of the facilities in China. The emission factors of other pollutants were mainly based on the European or American results due to the lack of corresponding measurement data in China. VOCs emission factors came from the study of Bo et al. (2008). NH3 emission factors were negligible in the fuel combustion sources."

{Kato, N., and Akimoto, H.: Anthropogenic emissions of SO2 and NOx in Asia: Emission inventories, Atmospheric Environment, 26, 2997-3017, 1992. Klimont, Z., Cofala, J.,Bertok, I., Amann, M., Heyes, C., Gyarfas, F.: Modelling particulate emissions in Europe: A framework to estimate reduction potential and control costs. Interim report, IR-02-076, International Institute for Applied Systems Analysis, Laxenburg, Austria, 2002a.}

(2) Re-write the section 2.3.2, page 957, lines 17-18: "USEPA (2002) reports the emission factors of each industrial process source category in AP-42 emission factor compilation. The emission factors of major PM emission sources in this study, such as coke production, iron & steel manufacturing, steel foundries, ferroalloy, and aluminum products, etc., are mainly cited from AP-42 with some adjustments to reflect poorer performance and lower particulate collection efficiencies of the technology level in China. VOCs emission from the industrial process is an important source which contributes

about 31.5% of the total in China according to the previous study (Wei et al., 2008). For the industrial processes of beer & spirits manufacturing, paint & ink, plastic, fibre and tyre production, we collect the emission factors from the study of Klimont et al. (2002b) and Bo et al. (2008) which originate from European and American studies, respectively. The VOCs emission factor of oil refining is hard to determine because of its complicated processes and miscellaneous fugitive segments. USEPA (2002) reported an overall THC emission factor of 3.54 kgâĂćt-1 for the refinery industry without control. European studies obtained lower emission factors, which are about 1.63 kgâĂćt-1 (Passant et al., 1998) and 1.05 kgâĂćt-1 (EEA, 2006). We cite a median value (2.65 kgâĂćt-1) reported by Klimont et al. (2002b) to represent current level of oil refining in the YRD region. VOCs emission factors of industrial uses of paint and solvent have large differences in the different literatures (USEPA, 2002; EEA, 2006; Wei et al., 2008; Bo et al., 2008). Based on the fact that the paint used in industry is mainly conventional solvent-borne paint with high solvent content, we determine the emission rates of paint application, printing, degreasing operations, and solvent use by Klimont et al. (2002b) and USEPA (2002). The emission factors of cement and bricks manufacturing are based on the domestic studies (Lei et al., 2008; Zhang et al., 2009). Table 3 lists the emission factors and their references of non-combustion industrial process sources."

{Passant, N.R., Vincent, K.: Review of the efficiency and cost of control measures for sulphur dioxide and volatile organic compounds. Draft final report AEAT-3851. AEA Technology, Culham, UK, 1998.}

(3) Re-write the section 2.3.4, page 958, lines 13-17: "The fugitive VOCs emission sources considered in this study mainly consist of domestic paint use for building and furniture, domestic solvent use, and oil/gas dissipation. For the architectural and domestic use of paint, this study uses paint consumption as the activity unit for these activities and for vehicle treatment. The emission factors of domestic paint and solvent uses and waste landfills are cited from Klimont et al. (2002). The volatilization and

leakage of VOCs emissions from the distribution and storage process of petroleum products mainly involve with the aspects including liquid loading losses, tank breath losses, and vehicle refueling operation losses. Bo et al. (2008) indicates that most gas stations adopted underground tanks and had no control in vehicle refueling operation by 2007. Therefore, we select the emission factors from that study."

(4) Re-write the section 2.3.5, page 958, lines 21-23: "Anthropogenic ammonia sources include livestock feeding, N-fertilizer application, sewage treatment, waste landfills, and human discharge in this study. The emission rates of major ammonia sources are subject to be influenced by local geography and climate conditions, so the emission factors mainly come from the domestic studies (Dong et al., 2009; Zhang et al., 2010; Yin et al., 2010). Only human discharge emission factor is referenced by European study (EEA, 2006) since there is no local study available. The activity data of these sources are collected from the annual statistical data of each administrative region."

(5) Re-write the section 2.3.6, page 959, lines 2-5: "Biomass burning emissions in this study come from household use and crop residue burning. The burning mass is estimated based on the statistical data of crop production, residue/crop ratio, and the percentage of burning. Most of emission factors in the studies on biomass burning emissions originate from Andreae and Merlet (2001) and Zhang et al. (2000). We use the summarized emission factors reported by Cao et al. (2005) in this study."

3. P959: is the emission height taken into account in the source allocation?

Re: The emission height is not considered in the source allocation in this study.

4. P961: SO2, NOx, PM10 annual concentrations are presented: where do these results come from? Why have these three specific pollutants been selected?

Re: The annual concentrations of SO2, NOx, PM10 come from the "2007 Ambient Air Quality Bulletin" of each city in the study. These specific pollutants were the three indicators which we called "API (Air Pollution Index)" to evaluate the air quality level

and to judge if the city exceeds the national standards or not. Other pollutant data like O3, PM2.5 is not available in the public.

5. Section 3.2 (and FiG. 3 and 4): Why CO has not been included in the figures and in the discussion? This section is named "emission contribution by source categories" but also includes a discussion on the comparison with the regional emission inventory from the Pearl River Delta region. Comparison with existing inventories is an important issue and would deserve an independent section. In the introduction the authors state that "low-resolution inventories were thought to cause under-estimation of air pollution simulation in recent modeling studies". Therefore it would be very valuable to compare the emissions derived from this study for the YRD region with the emissions of the corresponding grid cell of the national inventory. The comparison with the regional PRD emission inventory would also deserve more discussion.

Re: We would like to add CO in Fig 3 and 4. Please check it in the supplements. In the Section 3.2, Page 961, line 20, we supplement some discussions: "CO emission mainly comes from Iron & steel manufacturing and on-road vehicle, which contribute 34% and 30% of the total, respectively."

Based on the reviewer's recommendation, we think it is necessary to supplement an independent section on the comparisons of emission inventory with other studies. More discussions of the comparison between our study with the existed emission inventory studies are supplemented in the new section. To discuss the difference of the regional EI and the national one, we take the paragraph in Section 3.5, Page 965, Lines 1-8 to the new section. New section is inserted behind Section 3.2, Page 962, line 5 as follows: "3.3 Comparisons of emission inventory with other studies To better understand the emission level in the YRD region, we select another developed city cluster in southern China named the Pearl River Delta (PRD) region (including Guangzhou, Hong Kong, etc.) to compare their emission intensities of SO2, NOx, PM10, PM2.5, VOCs, NH3, and source contributions as shown in Fig. 4. The emission data come from the 2006 PRD emission inventories (Zheng et al., 2009b; Yin et al., 2010). The emission

intensities of SO2, NOx and VOCs in the YRD and PRD regions are all relatively high, reached (22-25) tâĂćkm-2 and (18-22) tâĂćkm-2, respectively. However, the PM10 and PM2.5 emission intensities in the YRD region are more than 2.5 times the values in the PRD region. Primary particulate matters from power plant and other industry sectors could be the major contributor which needs to be focused on the air pollution control strategies in the YRD region. Relatively, the industry sector plays a more important role in the emissions of the YRD region. The statistical data indicate that the products of oil refining, ethylene, cement, iron, and steel in the YRD region are about 2.0, 1.8, 2.6, 7.8, and 6.3 times of the products in the PRD region respectively (National Bureau of Statistics of China, 2008b). In contrast, road transport in the PRD region contributes more (about 36% and 53% of NOx and VOCs emissions) than that of YRD region (about 12.3% and 12.4% of NOx and VOCs emissions). To reveal the emission contributions of the YRD region to the whole China, we compare our work to the emission inventories for China as a whole (in Table 7) as published by ACCMIP datasets (Lamarque et al., 2010), EDGAR v4.1 (EC-JRC/PBL, 2011), GAINS-Asia (Amann et al., 2008), and INTEX-B (Zhang et al., 2009). Power generation is an important source of SO2 and NOx emission. The power sector of the YRD generates nearly 20% of the electricity in the whole China and its NOx and PM10 emissions contribute about 11%-40% of the total compared with the studies mentioned above. While the proportion of SO2 emission in the YRD region is only 5%-10% due to the installations of FGD units in some power plants by 2007. The industry sector of the YRD totally consumes 18% of the energy in the industry sector of the whole China and its SO2, NOx, PM10, and PM2.5 emissions contribute about 8%-28% of the total. The proportion of VOCs emission in industry of the YRD is relatively higher, about 23%-120% of the whole industry in China, which is mainly because that some industry sectors with high VOCs emissions, such as oil refinery, petrochemical industry, etc. are largely gathering in the YRD region. The road transport sector in the YRD region contributes 4%-23% of the emissions and 18% of the automobile ownerships correspondingly. In general, the comparisons reveal that the YRD emission inventory in this study is in line with most

of the national inventories. However, there are still some contradictions between this inventory and some studies which require further investigations in the future. The 2006 INTEX-B Asian emission inventory (Zhang et al., 2009) is compared to the emission inventory after compilation. The result shows that the difference of SO2, NOx, PM10 and VOCs emissions between the relative grids of both studies are about -3.8%, 7.0%, 58.1% and 23.8%, respectively. SO2 and NOx emission have relatively high agreement, while the uncertainties of PM10 and VOCs emission in both inventories are all higher than the others. The correlation coefficients of the grids are about 0.81, 0.80, 0.78 and 0.79, implying the similar spatial distribution patterns of emissions for the two studies. The emission intensities of PM10 and VOCs emission in this study are generally higher than INTEX-B. It might be because the activity data collected based on bottom-up approach in this study are usually more detailed and concentrated in some grids compared with the national or continental scale inventory."

{Amann, M., Jiang, K.J., Hao, J.M., Wang, S.X., Zhuang, X., Wei, W., Deng, Y.X., Liu, H., Xing, J., Zhang, C.Y., Bertok, I., Borken. J., Cofala, J., Heyes, C., Höglund, L., Klimont, Z., Purohit, P., Rafaj, P., Schöpp, W., Toth, G., Wagner, F., Winiwarter, W.: Scenarios for cost-effective control of air pollution and greenhouse gases in China. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria, 2008. European Commission: Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), Emission Database for Global Atmospheric Research (EDGAR), release version 4.1. http://edgar.jrc.ec.europa.eu/index.php (last access: 02 April 2010), 2011. Lamarque, J.F., Bond, T.C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M.G., Shindell, D., Smith, S.J., Stehfest, E., Van Aardenne, J., Cooper, O.R., Kainuma, M., Mahowald, N., McConnell, J.R., Naik, V., Riahi, K., van Vuuren, D.P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. Atmospheric Chemistry and Physics, 10, 7017-7039, 2010.}

6. Section 3.3 P962: It is difficult to compare VOC distribution from an emission inventory with measurements performed on-board an aircraft (which are representative of a given time and a given region). If there is no additional information on this comparison, it could be deleted as not relevant for the discussion.

Re: We would like to delete the following sentences: Section 3.3, Page 962, lines 19-25: "The percentages of alkanes, alkenes, and aromatics in the total VOC emission are 20 44%, 11%, and 36%, respectively. However, an aircraft measurement shows the proportion of ambient VOC concentration is 52%, 16%, and 17% in the YRD region (Geng et al., 2009). Our results seem to overestimate the content of aromatic. The main reason for the difference is believed to be that the aircraft measurement was mainly conducted in the rural region and most of the aromatics were consumed for the ozone for25 mation during transportation in the downwind."

7. Section 3.4 The ozone formation potential discussion is not very conclusive, in particular because biogenic VOCs are not included in the inventory. If kept, then the discussion should be improved (reference to BVOC from GEIA 1990 should be updated: see the MEGAN emission inventory, Guenther et al., 2006).

Re: We mainly discuss the anthropogenic emissions in the YRD region. We think it is acceptable to delete section 3.4 since it is hard to support the ozone formation potential discussion when there are only anthropogenic emissions. However, we would like to keep Fig. 7 and combine the description of the figure in Section 3.3. Please see the last paragraph of modified section 3.3, page 963 as follows: "In the terms of OFP, chemical industry, domestic paint & printing, gasoline vehicle, industrial coating, and oil refinery are the five major sources to the ozone formation, accounting for 38.2%, 23.9%, 11.6%, 9.6%, and 4.2%, respectively. The VOC emission contributions of these sources are about 34.5%, 21.7%, 9.7%, 18.8%, and 6.0%, respectively. The results shows the ozone formation contributions of the sources are not always correlated with their emission values in the YRD region. It is worthwhile to mention that gasoline vehicles are responsible for 9.7% of VOC emissions, as well as 11.6%

for ozone formation. Fig. 7 shows the spatial allocation of the OFPs in the YRD region. It is shown from the figure that OFPs are mainly concentrated in the well developed urban and industrial areas along the Yangtze River and around the Hangzhou Bay."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C1703/2011/acpd-11-C1703-2011-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 951, 2011.