

Interactive comment on “Spatial and temporal variation of anthropogenic black carbon emissions in China for the period 1980–2009” by Y. Qin and S. D. Xie

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Received and published: 23 March 2012

(Please refer to Supplement for the PDF version of Response to Reviewer#1 Comments) The manuscript written by Qin and Xie presents the inventories of anthropogenic black carbon emissions in China for the period 1980–2009. Such data are very scarce and therefore the paper is helpful to understand its impacts on both climate change and air pollution. The method is basically correct and the interpretation of the data is sound. However, there are still some important issues that the authors should consider. A revision is needed for publishing in ACP. Detail comments are listed as below:

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Specific Comment 1: The authors have published another paper on black carbon emissions, that is, Qin and Xie 2011a. What is the major difference between the two papers?

Response to Specific Comment No. 1: Basically, this paper and Qin and Xie (2011a) used the same method to calculate black carbon emissions, with some modifications regarding emission factors usage and mileage calculation. Besides, this paper and Qin and Xie (2011a) focused on different time scales and specific contents. (1) Regarding method improvement: First, emission factor for indigenous coking was modified. In this paper, EFBC was adjusted to 3.8 g/kg according to the work by Bond et al. (2007); while in Qin and Xie (2011a), 4.8 g/kg was used according to the work by Bond et al. (2004). Second, mileage calculation was modified. In Qin and Xie (2011a), mileage of heavy duty vehicles and other-duty vehicles were obtained from (Cai and Xie, 2007). But in this paper, annual mileage of these vans in each year from 1980 to 2009 was calculated by the following equations 1 and 2 (See attachments).

where: Q_n represents the national cargo turnover in year n ; $Q_{p,n}$ and $Q_{p,n+1}$ represent the cargo turnover accomplished by vehicle type p (heavy duty vehicles or other-duty vehicles) in year n and year $n+1$, respectively; $V_{p,n}$ represents the population of vehicle type p in year n ; T_p represents the average tonnage carried by vehicle type p ; $\beta_{p,n}$ represents the actual loading rate of vehicle p in year n ; $VMT_{p,n}$ and $VMT_{p,n+1}$ represent the annual average mileage of vehicle p in year n and year $n+1$, respectively. (2) Regarding the differences of these two papers' emphasis: Qin and Xie (2011a) dealt with BC emissions in year 2000. Domestic EFs database in this year was built, detailed subsector contributions in each of the major sources were studied, spatial distributions of total BC emissions and BC from each major source in 2000 was presented, and possible underestimation of transportation emissions by previous studies were found and discussed. While in this paper, we mainly focused on the temporal variation and spatial evolution of Chinese BC emissions in the past 30 years. Dynamic EFs database in China from 1980 to 2009 was constructed, historical changing trends of Chinese BC emissions were studied, the evolution of BC spatial distribution was

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discussed, and the historical contributions of Chinese BC emissions to East Asia, Asia and the whole world were illustrated. To sum up, the method was modified in this paper compared with Qin and Xie (2011a), and the results of these two papers have their particular focuses.

Specific Comment 2: Please provide the sectoral emission data for each province from 1980 to 2009.

Response to Specific Comment No. 2: Accept. We would provide the sectoral emission data for each province from 1980 to 2009 as supplements in the revised manuscript.

Specific Comment 3: 2.2 Emission allocation: As a high resolution emission inventory, the geological information such as the locations of lakes and mountains shall be considered during emission allocation. This is very helpful for the improvement of air quality modeling performance. I would like to see that the authors improve their emission allocation results in the revised manuscript.

Response to Specific Comment No. 3: Great thanks for this suggestion, which is both logical and scientific. However, there are some problems for us to satisfy the reviewer's requirement. First, it is possible to identify the geological information such as the locations of lakes and mountains based on the GIS system. However, it is basically impossible to identify whether there are people living and BC emissions or not. Because of China's large population, it is quite common for people living in mountains or by the river. Besides, residues open burning and forest fire are likely to happen in the mountain, while mobile sources on the river are likely to emit BC. Therefore, we cannot conclude that there are no BC emissions in the river or on the mountain. Second, this resolution is generally as high as the current BC inventories, such as Streets et al. (2003), Cao et al. (2006), and Zhang et al. (2009). Third, as to air quality modeling use, we think the resolution of $0.25^{\circ} \times 0.25^{\circ}$ in our paper is enough. As Streets et al. (2003) stated that regional emissions were gridded at a variety of spatial resolutions for input to the atmospheric simulation models, ranging from $1^{\circ} \times$

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1° for most regional model applications down to $30 \text{ sec} \times 30 \text{ sec}$ resolution for urban-scale studies. Therefore, we believe the inventory in our work can provide information concerning the historical evolution of Chinese BC emissions' spatial distribution, and can be applied for atmospheric modeling use.

Specific Comment 4: Line 12-13 on page 32884: "Emission factors for open burning of agricultural wastes were obtained from local experiments (Cao et al., 2007)" There are several studies on the emission factors of open burning of agricultural wastes in China, such as Li et al. (2007). I suggest the authors to thoroughly search the literature and include more local experimental data.

Response to Specific Comment No. 4: Thanks Reviewer for this suggestion. But there are the following problems: First, there are few published papers dealing with domestic EFBC of residues' open burning. Second, these scarce studies did not cover most of the crop types and the crops used in experiments are from a certain province instead of being collected from representative places in all parts of China. For example, Li et al. (2007) only studied the EFBC of maize and wheat based on residues collected from a rural area in Shandong province. In our manuscript, we used the experimental results by Cao et al. (2007), who designed a combustion tower to simulate open burning of the main crop residues, which were collected from representative parts all over China. As this study included most of the straw types we identify for BC emissions calculation, and the experimental materials were collected across the country, we believe their results can be used concerning the representativeness as well as the consistency and the comparability of different straw types. Still, we can cite Li et al. (2007) in the revised manuscript for comparison use if reviewer thinks it is a good idea.

Specific Comment 5: Line 16-17 on page 32885: "Emission factors for coal, residue and wood burning were from local measurements (Chen et al., 2005, 2006, 2009; Zhi et al., 2008, 2009; Shen et al., 2010)" Same as comments above, more local studies shall be included. The references shall also include Environ. Sci. Technol. 2009, 43, 6076–6081.

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Response to Specific Comment No. 5: Thanks for this suggestion. Actually we did cite the reference Environ. Sci. Technol. 2009, 43, 6076–6081 in our manuscript, as you can see on Page 29, Table 4 (Black carbon emission factors for fossil fuels and biofuels burning in residential sector). Most papers regarding domestic EFs for residential coal, residue and wood burning were studied and compared, but we omitted this reference in the text. We are sorry for this omission and it would be added in the revised manuscript.

Specific Comment 6: Line 7-9 on page 32886: “Assuming the use ratio and removal efficiency changed linearly from 1995 to 2020, annual EF in these sectors can be inferred by Eq. (6) (Qin and Xie, 2011a).” The use ratio and removal efficiency shall change according to the change of control policies and regulations such as emission standards. Therefore, I do not think the assumptions given by the authors here is appropriate. Besides, the emission estimates are for 1980-2009, why do the authors assume the use ratio changed linearly from 1995 to 2020?

Response to Specific Comment No. 6: Great thanks for this suggestion which is sound and scientific. The use ratio and removal efficiency shall change according to the change of control policies and regulations, technologies advancement and new technologies introduction. Because China has such a large territory and huge regional development gap, the application ratio and removal efficiency vary significantly across the country. Besides, there are no statistical data regarding the annual national use ratio and removal efficiency, and it is impossible to survey these parameters in each year from 1980 to 2009. Therefore, we have to make a simple assumption which can relatively reflect the changing trends of the national use ratio and removal efficiency in each year, based on the available survey results by Streets et al. (2001). Because Streets et al. (2001) provided the use ratio and removal efficiency in 1995 by survey and in 2020 by projection. Therefore, we assumed the use ratio changed linearly from 1995 to 2020, and made an extrapolation to 1980. In this way, we could get the annual national use ratio and removal efficiency for the period 1980 to 2009. This assumption is not an ideal one, but it at least provides the relative changing trends of the national

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use ratio and removal efficiency, thus it is both feasible and acceptable. Moreover, the uncertainty resulted from the assumed use ratio and removal efficiency was included in the inventory uncertainty analysis.

Specific Comment 7: Table 2 on page 32903: please give the references of the data in this table.

Response to Specific Comment No. 7: Thanks for this suggestion. Vehicle mileage travelled (VMT) of Buses and Coaches are calculated by equation 3. National cargo turnover is accomplished by both heavy duty vehicles and other-duty vehicles, whose respective contributions were assumed to be positively correlated with their population and the average tonnage carried, as shown in equation 4. The mileage of heavy duty vehicles (other-duty vehicles) was calculated by equation 5. Lacking of appropriate statistical data to infer the mileage of passenger cars and motorcycles, they were obtained from summarized literatures survey (Cai and Xie, 2007; Bo et al., 2008). References in this table will be added in the revised manuscript.

where: $VMT_{b,n}$ is the mileage of buses and coaches in year n , $Q_{b,n}$ is the passenger turnover accomplished in year n , $\alpha_{b,n}$ is the actual loading rate of buses and coaches in year n , $VP_{b,n}$ is the population of buses and coaches in year n , and T_b is the average number of seats on buses and coaches.

where: Q_n represents the national cargo turnover in year n ; $Q_{p,n}$ and $Q_{p,n+1}$ represent the cargo turnover accomplished by vehicle type p (heavy duty vehicles or other-duty vehicles) in year n and year $n+1$, respectively; $VP_{p,n}$ represents the population of vehicle type p in year n ; T_p represents the average tonnage carried by vehicle type p ; $\beta_{p,n}$ represents the actual loading rate of vehicle p in year n ; $VMT_{p,n}$ and $VMT_{p,n+1}$ represent the annual average mileage of vehicle p in year n and year $n+1$, respectively

Specific Comment 8: Table 5. Chinese application ratio and removal efficiency for various control devices in industry sector in 1995 and 2020. (1) What is “Powder-

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ESP"? I guess it shall be "Power-ESP". (2) Why do the authors use data in 1995 and 2020 to estimate emissions 1980-2009?

Response to Specific Comment No. 8: Great thanks for this suggestion. I am quite sorry for our carelessness. "Powder-ESP" will be changed to "Power-ESP" in the revised manuscript. Because Streets et al. (2001) provided the use ratio and removal efficiency in 1995 by survey and in 2020 by projection. Therefore, we assumed the use ratio changed linearly from 1995 to 2020, and made an extrapolation to 1980. In this way, we could get the annual national use ratio and removal efficiency for the period 1980 to 2009.

Specific Comment 9: Table 6. Raw emission factors for black carbon in industry sector in China. The authors only use data from Streets et al. (2001), Bond et al. (2004), and Bond et al. (2007). The Chinese data shall be included, such as Wang et al. (2009).

Response to Specific Comment No. 9: Thanks for this suggestion. Wang et al. (2009) is a very helpful paper which deals with the emission characteristics of fine particles from grate firing boilers. However, there are following problems which render its application in our paper. First, we applied the use ratio and removal efficiency of different control devices from Streets et al. (2001), thus our estimation of industrial emissions is based on the corresponding classification. Second, actually, Wang et al. (2009) provided emission factors of PM_{2.5}, NO_x and SO₂, without providing black carbon emission factors in their paper. Therefore, we are waiting for further more complete local EFs data from their group to improve our emission inventory.

Specific Comment 10: Table 7. Chinese application ratio and removal efficiency for various control devices in power generation sub-sector in 1995 and 2020. (1) Same as comment 8, "Powder" shall be "power". (2) The use ratio of ESP was already 0.95 in 2008. Therefore, the use ratio of scrubber and cyclone shall be much lower than that given in Table 7. (3) Why do the authors use data in 1995 and 2020 to estimate emissions 1980-2009?

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Response to Specific Comment No. 10: Thanks for this suggestion. Please refer to Response to Comment No.6 and No.8 for details.

Specific Comment 11: Fig. 8: Please add the comparison with Lei (2008).

Response to Specific Comment No. 11: Accept. Comparisons with Lei (2008) would be added in the revised manuscript. "The changing trends of our emission inventory also matched well with the study by Lei (2008), who found that BC emissions peaked in 1995 from 1990 to 2000, and have then been continually increasing after 2000." "Bio-fuel contribution decreased from 44.25% in 1980 to 31.07% in 2009, despite emissions from this source continued to increase, similar to the finding by Lei (2008) that biofuel accounted for about 35% of national BC emissions from 1990 to 2005. Vehicle contributions increased from 2.93% in 1980 to 6.02% in 1990, and then to 12.82% in 2009, resembling the study by Lei (2008) that vehicle contribution increased from 5% to 11% during the period 1990-2005."

Specific Comment 12: References on page 32889: the two references, Qin and Xie 2011a and Qin and Xie 2011, are same.

Response to Specific Comment No. 12: Thanks for pointing out our carelessness. The repeated reference will be deleted in the revised manuscript.

Specific Comment 13: There are some other typos in the manuscript. Please correct them before submitting the revision.

Response to Specific Comment No. 13: Great thanks for this suggestion. We are quite sorry for the typos through the manuscript. Corrections would be made in the revised manuscript!

Reference

Bo, Y., Cai, H., and Xie, S. D.: Spatial and temporal variation of historical anthropogenic NMVOCs emission inventories in China, *Atmos Chem Phys*, 8, 7297-7316, 2008. Qin, Y. and Xie, S. D.: Estimation of county-level black carbon emissions and its spatial

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distribution in China in 2000, *Atmos Environ*, 45, 6995-7004, 2011a. Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J. H., and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.-Atmos.*, 109, doi:10.1029/2003JD003697, 2004. Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S. K., Roden, C., Streets, D. G., and Trautmann, N. M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, *Global Biogeochem. Cy.*, 21, doi:10.1029/2006GB002840, 2007. Chen, Y. J., Sheng, G. Y., Bi, X. H., Feng, Y. L., Mai, B. X., and Fu, J. M.: Emission factors for carbonaceous particles and polycyclic aromatic hydrocarbons from residential coal combustion in China, *Environ Sci Technol*, 39, 1861-1867, 2005. Cai, H., and Xie, S. D.: Estimation of vehicular emission inventories in China from 1980 to 2005, *Atmos Environ*, 41, 8963-8979, 2007. Chen, Y. J., Zhi, G. R., Feng, Y. L., Fu, J. M., Feng, J. L., Sheng, G. Y., and Simoneit, B. R. T.: Measurements of emission factors for primary carbonaceous particles from residential raw-coal combustion in China, *Geophys Res Lett*, 33, -, 2006. Chen, Y. J., Zhi, G. R., Feng, Y. L., Liu, D. Y., Zhang, G., Li, J., Sheng, G. Y., and Fu, J. M.: Measurements of Black and Organic Carbon Emission Factors for Household Coal Combustion in China: Implication for Emission Reduction, *Environ Sci Technol*, 43, 9495-9500, 2009. Cao, G. L., Zhang, X. Y., Wang, Y. Q., and Zheng, F. C.: Estimation of China's regional agricultural waste open burning emissions, *Chin. Sci. Bull.*, 52, 1826-1831, 2007. Cao, G. L., Zhang, X. Y., and Zheng, F. C.: Inventory of black carbon and organic carbon emissions from China, *Atmos Environ*, 40, 6516-6527, DOI 10.1016/j.atmosenv.2006.05.070, 2006. Lei, Y.: Research on Anthropogenic Emissions and Control of Primary Particles and Its Key Chemical Components, Tsinghua University Ph.d Dissertation, 2008. Li, X. H., Wang, S. X., Duan, L., Hao, J. M., Li, C., Chen, Y. S., Yang, A. L.: Particulate and Trace Gas Emissions from Open Burning of Wheat Straw and Corn Stover in China, *Environ. Sci. Technol*, 41, 6052-6058, 2007. Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions

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in Asia in the year 2000, *J Geophys Res-Atmos*, 108, -, 2003. Streets, D. G., Gupta, S., Waldhoff, S. T., Wang, M. Q., Bond, T. C., and Bo, Y. Y.: Black carbon emissions in China, *Atmos Environ*, 35, 4281-4296, 2001. Shen, G. F., Yang, Y. F., Wang, W., Tao, S., Zhu, C., Min, Y. J., Xue, M. A., Ding, J. N., Wang, B., Wang, R., Shen, H. Z., Li, W., Wang, X. L., and Russell, A. G.: Emission Factors of Particulate Matter and Elemental Carbon for Crop Residues and Coals Burned in Typical Household Stoves in China, *Environ Sci Technol*, 44, 7157-7162, 2010. Wang, S. X., Zhao, X. J., Li, X. H., Wei, W., Hao, J. M.: Emission Characteristics of Fine Particles from Grate Firing Boilers, *Environ Sci*, 30, 963-968, 2009. Zhi, G. R., Chen, Y. J., Feng, Y. L., Xiong, S. C., Li, J., Zhang, G., Sheng, G. Y., and Fu, J.: Emission characteristics of carbonaceous particles from various residential coal-stoves in China, *Environ Sci Technol*, 42, 3310-3315, 2008. Zhi, G. R., Peng, C. H., Chen, Y. J., Liu, D. Y., Sheng, G. Y., and Fu, J. M.: Deployment of Coal Briquettes and Improved Stoves: Possibly an Option for both Environment and Climate, *Environ Sci Technol*, 43, 5586-5591, 2009. Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, *Atmos Chem Phys*, 9, 5131-5153, 2009.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/11/C16182/2012/acpd-11-C16182-2012-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 32877, 2011.

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$$Q_{p,n} = Q_n \times \frac{VP_{p,n} \times I_p}{\sum_i VP_{p,n} \times I_p} \quad (1)$$

Fig. 1.

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$$VMT_{D,n+1} = \frac{Q_{D,n+1} \times VP_{D,n} \times \beta_{D,n}}{VP_{D,n+1} \times \beta_{D,n+1} \times Q_{D,n}} \times VMT_{D,n} \quad (2)$$

Fig. 2.

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$$VMT_{z,n} = Q_{z,n} / (\alpha_{z,n} \times VP_{z,n} \times T_z) \quad (3)$$

Fig. 3.

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$$Q_{p,n} = Q_n \times \frac{VP_{p,n} \times T_p}{\sum_i VP_{p,n} \times T_p} \quad (4)$$

Fig. 4.

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$$VMT_{D,N+1} = \frac{Q_{D,N+1} \times VP_{D,N} \times \beta_{D,N}}{VP_{D,N+1} \times \beta_{D,N+1} \times Q_{D,N}} \times VMT_{D,N} \quad (5)$$

Fig. 5.