

The authors developed an anthropogenic BC emission inventory in China for the period 1980-2009. The BC emission inventory is essential and important data for atmospheric science community and policymaker in the field of PM pollution as well as climate change due to the SLCF. The topic certainly is suitable for ACP. The manuscript presents the spatial and temporal variations for BC emissions in China, the comparison with other inventories, and uncertainty analysis. The author's inventory has some advantages in the targeted period covering the 30 years (1980-2009) and in the input data (time-varying emission factors, local emission factors for domestic sector, and others). However, the originality and new findings of the manuscript is much less compared to recent publication of ACP (Lu et al., 2011). From this point, the manuscript needs to be improved in the following aspects at least to be qualified science paper in ACP: (1) demonstrating clearly the scientific advance of the improvement of emission inventory due to the methodology and input data used in the work; (2) adding the more discussion of the comparison with bottom-up inventories (especially, Lu et al., 2011) and the top-down inventories (especially, Fu et al., 2011) the implication of their differences. In conclusion, I am recommending the major revision of this manuscript in the following points.

<References> Lu et al.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996-2010, ACP, 11, 9839-9864, 2011. Fu et al.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, ACPD, 11, 28219-28272, 2011.

### **Major Comments:**

(1) One of the advantages of the author's work is the using of timevarying emission factors for vehicles, industry, and power generation. However, the emission factors for industry and power generation are assumed based on the percentage of control devices and the removal efficiency in 1995 and 2020 from Streets et al. (2001). This is just a rough assumption; hence the authors should evaluate and demonstrate the validity and/or the limitation of the assumption. Additionally, the authors should demonstrate the temporal variations of vehicle emission factors used in this work (page 32882,

lines 20-24).

(2) Lu et al. (2011) presented the historical BC emissions in China for the period 1996-2010. Also, Fu et al. (2011) estimated BC emissions in China for 2006 by topdown constraints. It is recommended that the authors should compare to the emissions estimated by these works and discuss about the implication of their differences.

### **Response to major Comments:**

(1) Thanks for this comment. 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 national use ratio and removal efficiency in each year, 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 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.

The temporal variations of vehicle emission factors used in this work are based on the new emission standards introduced in different years as illustrated in Table 1 (page 26) in the previous manuscript. Temporal EFs for vehicle under different emission standards would be provided in the revised manuscript.

(2) Our manuscript used a consistent methodology to develop the Chinese historical

BC emission inventory for the period 1980-2009 based on time-varying emission factors and official activity data. Chinese dynamic emission factor database for the past thirty years were built up, and BC emissions were estimated correspondingly. Comparing with Lu et al. (2011), our manuscript has the following improvements: First, there have been some attempts to build up Chinese BC emission inventories. However, due to the substantial differences among these results, none of these studies have been accepted and used by the Chinese government. In order to identify Chinese historical BC emissions and the changing trends of Chinese contributions to other districts, a National Program on Key Basic Research Project (973 Program) addressing BC was established by the Chinese government, and this study is funded by this project aiming at providing Chinese recognized BC emission inventory based on our domestic EFs and public activity data. Parameters employed in this manuscript have been discussed and accepted by the program members, and the results are very likely to be used by the Chinese government concerning policy formulation. Second, BC emissions in the past thirty years were built up using a consistent methodology, which can clearly show their historical changing trends. Third, we provided the dynamic  $EF_{BC}$  database for the past thirty years which shows the basis of our historical inventory, and which can be used to extrapolate the present inventories to other years. Though Lu et al. (2011) established their inventories also based on temporal EFs, they did not provide the time-varying EFs database used in their study. Fourth, the focus of our manuscript and Lu et al. (2011) are different. Lu et al. (2011) focused on developing a comprehensive inventory of SO<sub>2</sub>, BC, and OC emissions from China and India for the period 1996–2010, uncertainties and seasonality of emissions were studied, and spatial distribution of national emissions were presented. While in our paper, we focused on historical BC emissions from 1980 to 2009. Historical fuel type and provincial contributions were studied, to provide respective suggestions for emissions control by fuel type and by province. Spatial distribution of national BC emissions and emissions from each source were presented to show the historical evolution of their spatial distribution. Besides, historical contributions of Chinese BC emissions to other districts were studied, which are deeply concerned by

the scientific community.

Actually, there have been relatively few emission inventories dealing with Chinese historical BC emissions based on time-varying domestic EFs and published activity data. The results and changing trends of our inventory for the period 1996-2009 were comparable to the study by Lu et al. (2011), which indirectly indicated that our inventory from 1980-2009 are reliable. Because these two papers have their respect time range and different focuses, and our paper has its practical meaning and further useful application, our manuscript can also provide valuable information to both the scientific community and the government reference.

Fu et al. (2011) estimated BC emissions in China for 2006 by top-down constraints. Both the method and the focus of this manuscript and our paper are different, thus we do not think Fu et al. (2011) reduce the originality of our paper. We would add comparison of our manuscript with Lu et al. (2011) and Fu et al. (2011) in the revised manuscript.

## **Minor comments**

### **Specific Comment 1:**

Eq. (1): Is the subscript “m” of EF correct? If so, the authors should explain how to estimate the EF by province.

### **Response to Specific Comment No. 1:**

Thanks for this suggestion. Equation 1 is correct. In this equation, only the EFs for vehicles may be different among provinces due to the implementation of various emission standards in different provinces in the same year. As we mentioned in Section 3.1, new vehicles with advanced emission-control technologies have been quickly introduced in the past ten years (Table 1). Because the same emission standard did not introduce at the same speed in different provinces, thus the corresponding EFs are different by province.

**Table 1.** Time tables for vehicle-emission-regulations implementation in China.

	Euro I	Euro II	Euro III	Euro IV
Beijing	1999	2002	2005	2008
Shanghai	1999	2005 <sup>a</sup>	2008 <sup>b</sup>	2009
National	2001	2005 <sup>a</sup>	2008 <sup>b</sup>	2010

<sup>a</sup> the actual implementation time is June, 2004;

<sup>b</sup> the actual implementation time is June, 2007.

**Specific Comment 2:**

Line 11, page 32882: Why is the navigation excluded?

**Response to Specific Comment No. 2:**

Accepted. We did not include navigation emissions as they are quite small, but we can include it in the revised manuscript if the reviewer thinks it is necessary.

**Specific Comment 3:**

Line 17, age 32885: The references are inconsistent with those in the footnote of Table 14.

**Response to Specific Comment No. 3:**

Thanks for this suggestion. In the text, we stated that “Emission factors for coal, residue and wood burning were from local measurements (Chen et al., 2005; Chen et al., 2006; Chen et al., 2009; Zhi et al., 2008; Zhi et al., 2009; Shen et al., 2010), while EFs for oil and gas burning from Bond et al. (2004), as listed in Table 4.” Here we listed most of the papers we used for comparison to determine the EFs used in our emission inventory. For instance, Chen et al. did and published a series of experiments regarding the EFs for residential coal consumption (Chen et al., 2005; Chen et al., 2006; Chen et al., 2009). But in their latest publication (Chen et al., 2009), they

provided a detailed data set of  $EF_{BC}$  for household coal burning, which was presented on the basis of 38 coal/stove combination experiments and which included 13 coals with a wide coverage of geological maturity tested in honeycomb-coal-briquette and raw-coal-chunk forms in three typical coal stoves. Therefore, we actually used  $EF_{BC}$  in their latest work in our inventory calculation. Besides, we carelessly omitted the reference Li et al. (2009) in the previous text which we cited in Table 4, which we would added in the text in the revised manuscript.

**Table 4.** Black carbon emission factors for fossil fuels and biofuels burning in residential sector (Units: g/kg)

Fuel type	Emission factor
bituminous briquette <sup>a</sup>	0.09
anthracite briquette <sup>a</sup>	0.004
bituminous raw coal <sup>a</sup>	3.05
anthracite raw coal <sup>a</sup>	0.007
gasoline <sup>b</sup>	0.07
Kerosene <sup>c</sup>	0.12
diesel <sup>b</sup>	0.25
LPG <sup>c</sup>	0.068
Gases <sup>c</sup>	0.0001
agriculture waste <sup>d</sup>	0.77
firewood <sup>e</sup>	1.49

<sup>a</sup> Chen et al., 2009

<sup>b</sup> Streets et al., 2001

<sup>c</sup> Bond et al., 2004

<sup>d</sup> The geometric mean of EFs in Shen et al. (2010) and Li et al. (2009)

<sup>e</sup> Li et al., 2009

**Specific Comment 4:**

Lines 7-8 and 25-26, page 32886: The authors need to explain how to extrapolate to the period 1980-1994.

**Response to Specific Comment No. 4:**

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 national use ratio and removal efficiency in each year. 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 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 5:**

Table 3: In the title of table, “for different biomass types for open burning in China” is better for clarification of “open burning” or “biofuel”.

**Response to Specific Comment No. 5:**

Accepted. Table 3 is changed as “Black carbon emission factors and burning efficiency for different biomass types for open burning in China”.

**Specific Comment 6:**

Figures: All figures are not clear. They need to be improved.

**Response to Specific Comment No. 6:**

Accepted.

**Specific Comment 7:**

Figs. 2 and 3: The figures showing the time evolution of emission amount as well as the relative contribution will give the useful information in the manuscript.

**Response to Specific Comment No. 7:**

Accepted. Figures showing the time evolution of emission amount as well as the relative contribution will be presented in the revised manuscript.

**Specific Comment 8:**

Figs. 4 and 7: The size of these figures is too small to be visible. These should be improved.

**Response to Specific Comment No. 8:**

Response: Accepted.

**Specific Comment 9:**

Figs. 5 and 6: A unit of emissions should be specified.

**Response to Specific Comment No. 9:**

Accepted.

**Specific Comment 10:**

Fig. 8: The continuous data of Ohara et al. (2007) are provided on the web site of <http://www.jamstec.go.jp/frcgc/research/p3/emission.htm>.

**Response to Specific Comment No. 10:**

Accepted. Thanks for this information. Continuous country data from 1980-2003 will be used. As for year 2004-2009, they only projected the gridded data.

**Specific Comment 11:**

Fig. 9: It is difficult to distinguish the blue lines showing 2.5%, Median, Mean, and 97.5% from the blue bar of emission uncertainty. Additionally, the horizontal axis label should be added.

**Response to Specific Comment No. 11:**

Accepted.

**References**

- 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.
- 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.
- Fu, T.-M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., Qu, W. J., Han, Z., Zhang, R., Wang, Y. X., Chen, D., and Henze, D. K.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, *Atmos Chem Phys Discuss*, 11, 28219-28272, 2011.
- Li, X. H., Wang, S. X., Duan, L., Hao, J. M., Nie, Y. F.: Carbonaceous aerosol emissions from household biofuel combustion in China, *Environ. Sci. Technol.*, 43, 6076–6081, 2009.

- Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996-2010, *Atmos Chem Phys*, 11, 9839-9864, 2011.
- Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, *Atmos.Chem. Phys.*, 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.
- 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.
- 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.