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Interactive comment on “Spatial and temporal variation of emission inventories for historical anthropogenic NMVOCs in China” *by* Y. Bo et al.

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We sincerely appreciate the constructive suggestions and comments of referee 2 on the necessity and the methods of evaluating the accuracy of our compiled inventory, which remind us of the importance of conducting an uncertainty assessment of the inventory to make a substantial improvement of it. Upon carefully reading the comments of referee 2, we think that referee 2 mainly put forward two issues of the manuscript, which are highly valued and answered as follows:

Comment (1):

"I find that year-to-year emission variation comes from activity data or economic movement."; "In most cases, emission factors are simply averages of all available data of acceptable quality, and are generally assumed to be representative of long-term aver-

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ages."

Response:

Most emission factors in 2005 in the manuscript were simply averages of all available data of acceptable quality obtained by referring to native research, published literatures, and the AP-42 Report (EPA, 1995), due to the lack of substantially historical emission factors and enough site-specific emission data. Nevertheless, the appropriate emission factors were chosen from the AP-42 Report according to the comparison of energy consumption levels between China in 2005 and the U.S. in 1995. Moreover, annual emission factors of specific sources for the years before 2005 were modified according to corresponding assumptions:

Regarding the sources of industrial and fossil fuel combustion, annual emission factors for the years before 2005 were modified based on the assumption that the emissions of NMVOCs per unit of fuel consumed were stable, and the temporal variance of the emission factors was correlative with the annual change of energy consumption per production, as shown by Equation (2) in the manuscript.

Regarding the source of solvent utilization, it was assumed that the emission factors were correlative with annual income per capital and the emission factors were modified according to Equation (3) in the manuscript.

Emission factors of biomass open burning, and storage and transport were assumed to remain constant over the years, in that the burning method of biomass open burning and the way of storage and transport in China changed little over the years.

As for the emission factors for vehicles calculated by the COPERT III methodology, average driving speeds, ambient temperature, and the vapor pressure of gasoline, three main factors which influence the emission factors of various categories of vehicles were fully considered to obtain convincing emission factors of vehicles over the period of 1980-2005.

Therefore, emission variation during the period of 1980-2005 comes from both activity data and modified historical emission factors, while the emission variation of the sources of biomass open burning and storage and transport was only ascribed to the changing activity data.

Comment (2):

"I highly suggest the authors should conduct uncertainty analysis of inventory to explain the quality of their work and the range of inventory. In particular, the work should focus on assessing the distribution of inter-unit variability as an estimate of uncertainty."

Response:

We appreciate very much the suggestion of referee 2 that we conduct an uncertainty analysis of the inventory, which is absolutely necessary and improves the quality of our current work. As referee 2 has pointed out, the uncertainty of our inventory derives from the use of different emission factors and activity data, most of which were selected from AP-42 report and other literatures, due to the lack of local measured firsthand data. As for our work which includes six major sources of NMVOC emission, the uncertainty analysis for each of the six sources is therefore conducted before the results are integrated into the final result of uncertainty analysis of the total emission inventory. Particularly, we analyze the uncertainty of the emission factors of each of the six major sources, and ignore the uncertainty analysis of the activity data of the four sources of storage and transport, industrial processes, solvent utilization and fossil fuel combustion, which were firsthand obtained and are therefore considered dependable. Besides, we analyze the uncertainty of the activity data of the two sources of vehicles and biomass open burning, which were estimated based on specific assumptions instead of being directly obtained from firsthand data sources.

Based on the US EPA's procedures recommended by referee 2 for conducting uncertainty analysis (EPA, 2007), it is difficult to calculate the uncertainty of those emission factors or activity data with few sample data, since the uncertainty analysis relies on

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adequate sample data to represent good-fitting theoretical probability density functions. To come over this difficulty, we prepare the data required by the uncertainty analysis according to the two principles enlightened by referee 2, which consider different sample sizes of specific units within a source category during the uncertainty analysis process.

With respect of those sources with large sample size of emission factors, theoretical probability density functions (PDFs) were applied to model the emission factor data. Candidate models were the Weibull, log-normal, and gamma PDFs. Maximum likelihood estimation was used to estimate the parameters of the theoretical distributions. Then, the propagation of uncertainty of each emission factor was calculated through the Monte Carlo Simulation, which was run for 10,000 times for each uncertainty assessment to obtain the target statistics of the median, mean, and 95th percentile values. There are three sources with large sample sizes of emission factors in our work: vehicles, fossil fuel combustion, and industrial processes. Taking the motorcycle from vehicle source for example, emission factor of motorcycle has a large sample size, and we summarize the data of emission factor from both model calculation of the thirty-one provinces in China including 4.74, 4.89, 4.80, 4.60, 4.50, 4.49, 4.46, 4.44, 4.85, 4.67, 4.71, 4.69, 4.79, 4.67, 4.80, 4.83, 4.68, 4.72, 4.74, 4.78, 4.80, 4.94, 4.59, 4.54, 4.46, 4.38, 4.79, 4.58, 4.45, 4.57, 4.56 g/km, and those from literatures including 7.30 (Fu et al., 2000), 5.25 (Hao et al., 2002), and 5.98 (Li et al., 2003) g/km, for uncertainty assessment. Data visualization (histograms and empirical cumulative density function plots) were performed to observe the range, skewness and other possible characteristics of the data. Then, Weibull, log-normal, and gamma PDFs were chosen to fit the datasets, and the log-normal distribution that best fits the data was selected, with a standard deviation of 0.44. Finally, the Monte Carlo Simulation was repeatedly implemented with new input values randomly selected from within their respective probability distribution. Simulations were run 10,000 times to get the propagation of uncertainty of motorcycle emission factors and the result revealed that the 95% confidence interval for emission factor of motorcycle was about [-17%,19%]. Similar way of uncertainty assessment was applied to the sources of fossil fuel combustion and industrial processes,

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and the results at the 95% confidence interval were generated for each of them.

There are three sources with a small sample size of emission factors in one unit and with no site-specific emission data: solvent utilization, biomass burning, and storage and transport. For these sources, an assumption is made that the uncertainty of their emission factors had log-normal distributions. The standard deviations for these distributions were estimated by the "expert evaluation method", which is based on the reliability of the data sources and the differences in investigators' results, and has been used in recent research of the uncertainty assessment of VOC emissions (Van der Sluijs et al., 2005;Wei et al., 2008). Uncertainty of each emission factor was calculated through the Monte Carlo Simulation (10,000 times), which output the target statistics of the median, mean, and 95th percentile values. Taking the emission factor of cornstalk from biomass open burning for example, we selected local measured data and emission factors from other estimated emission inventories for China, which include 10 g/kg (Li et al., 2007), 15.7 g/kg (Streets et al., 2003), 8.7 g/kg (Klimont et al., 2002), and 7.0 g/kg (Andreae and Merlet, 2001). We assumed that the uncertainty of emission factors had a log-normal distribution. The standard deviation for the distribution was estimated at 2.5 by the "expert evaluation method". Then, Monte Carlo Simulation was repeatedly implemented with new input values randomly selected from within their respective probability distribution. Simulations were run 10,000 times to get the propagation of uncertainty of emission factors. The result showed that the median was 9.7 g/kg, the 2.5th percentile was 5.99 g/kg, and the 97.5th percentile was 15.72 g/kg at the 95% confidence interval. Similar way of uncertainty assessment was applied to the sources of solvent utilization and storage and transport, and the results at the 95% confidence interval were generated for each of them.

The above procedures for small-sample-size objects were also applied to analyze the uncertainty of vehicle travel mileage (VMT) and biomass amount, the activity data of the sources of vehicles and biomass open burning, respectively, whose sample sizes are small. For example, the biomass volume in 2005 was estimated 107.70 Tg in our

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study. Another estimation was 131.78 Tg (Cao et al., 2006). Therefore, we made an assumption that the uncertainty of activity data had log-normal distributions, and the standard deviation for the distribution was estimated at 19.0 by the "expert evaluation method". Then, Monte Carlo Simulation was repeatedly implemented with new input values randomly selected from within their respective probability distribution. Simulations were run 10,000 times to get the propagation of uncertainty of activity data. The result shows that the median was 107.55 Tg, the 2.5th percentile was 75.31 Tg, and the 97.5th percentile was 149.04 Tg at the 95% confidence interval. Similar way of uncertainty assessment was applied to VMT, and the result at the 95% confidence interval was generated.

Upon the completeness of uncertainty analysis of emission factors and activity data for each of the six sources, the propagation of uncertainty of emission factors and activity data at the 95% confidence interval were identified. Subsequently, new values of emission factors and activity data of each source were randomly selected from the calculated propagation of uncertainty, using the Monte Carlo Simulation, and the emission of each source was calculated according to the equation E=EF*A. Then, emissions of the six sources were summed up as one total emission. This process was repeatedly run for 10,000 times, resulting in a sample set of 10,000 of the total emission, whose statistics of the median, mean, and 95th percentile values were calculated and therefore the propagation of uncertainty of the total emission at the 95% confidence interval was obtained. For the year 2005, the propagation of uncertainty of total emission at the 95% confidence interval was about [-63%, 165%]. The modeling results showed that the most sensitive elements were emission factors of motor vehicles, vehicle miles traveled and biomass amount of biomass open burning. As for the uncertainty of emissions of specific sources, the sources with the highest uncertainty were industrial processes and vehicle emission, with an uncertainty of [-88%, 227%] and [-77%, 189%], respectively.

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