

1 **Uncertainties in estimating mercury emissions from coal-fired** 2 **power plants in China**

3
4 Y. Wu¹, D. G. Streets², S. X. Wang¹, and J. M. Hao¹

5
6 ¹Department of Environmental Science and Engineering, and State Key Joint Laboratory
7 of Environment Simulation and Pollution Control, Tsinghua University, Beijing, China

8 ²Decision and Information Sciences Division, Argonne National Laboratory, Argonne,
9 Illinois 60439, U.S.A.

10 *Correspondence to:* Y. Wu (ywu@tsinghua.edu.cn)

11 12 ***Abstract:***

13 A detailed multiple-year inventory of mercury emissions from anthropogenic activities in
14 China has been developed. Coal combustion and nonferrous metals production continue
15 to be the two leading mercury sources in China, together contributing ~80% of total
16 mercury emissions. **However, many uncertainties still remain in our knowledge of**
17 **primary anthropogenic releases of mercury to the atmosphere in China. In situations**
18 **involving large uncertainties, our previous mercury emission inventory that used a**
19 **deterministic approach could produce results that might not be a true reflection of reality;**
20 **and in such cases stochastic simulations incorporating uncertainties need to be performed.**
21 Within our inventory, a new comprehensive sub-module for estimation of mercury
22 emissions from coal-fired power plants in China is constructed as an uncertainty case
23 study. The new sub-module integrates up-to-date information regarding mercury content
24 in coal by province, coal washing and cleaning, coal consumption by province, mercury
25 removal efficiencies by control technology or technology combinations, etc. Based on
26 these detailed data, probability-based distribution functions are built into the sub-module
27 to address the uncertainties of these key parameters. The sub-module incorporates Monte
28 Carlo simulations to take into account the probability distributions of key input
29 parameters and produce the mercury emission results in the form of a statistical
30 distribution. For example, the best estimate for total mercury emissions from coal-fired
31 power plants in China in 2003 is 90.5 Mg, with the uncertainty range from 57.1 Mg (P10)

1 to 154.6 Mg (P90); and the best estimate for elemental mercury emissions is 43.0 Mg,
2 with the uncertainty range from 25.6 Mg (P10) to 75.7 Mg (P90). The results further
3 indicate that the majority of the uncertainty in mercury emission estimation comes from
4 two factors: mercury content of coal and mercury removal efficiency.

6 **1. Introduction**

7 Concern about mercury (Hg) in the environment has grown as its dangerous effects
8 are well established. The confirmation of the ability of elemental mercury (Hg⁰) to
9 undergo long-range transport at hemispheric scale (Banic et al., 2003; Dastoor and
10 Larocque, 2004; Seigneur et al., 2001; Travnikov and Ryaboshapko, 2002) intensifies the
11 anxiety in some countries/regions that the quantities of imported atmospheric Hg may be
12 substantial and may interfere with the ability of domestic sources to comply with future
13 emission limitations (Jaffe et al., 2005; Seigneur et al., 2004; Selin et al., 2007; Steding
14 and Flegal, 2002; Weiss-Penzias et al., 2007). For example, Seigneur et al. (2004)
15 estimated that anthropogenic emissions of mercury in Asia contributed 21% to total
16 mercury deposition in the contiguous United States in 1998.

17 During the past two decades considerable progress has been made in better
18 estimating anthropogenic Hg sources at global-scale as well as at national-scale. Pacyna
19 and his co-workers continue to update global Hg emission inventories, and they have
20 generated estimates of 2140 Mg for 1990, 1910 Mg for 1995, and 2190 Mg for 2000
21 (Pacyna and Pacyna, 1996, 2002; Pacyna et al., 2006). According to the most recent
22 global inventory, about 65% of emissions came from stationary fuel combustion in 2000;
23 geographically, about 54% of the emissions came from Asia, and China was the largest
24 Hg emitting country (Pacyna et al., 2006).

25 Mercury contamination is a serious problem in China. Feng (2005) has summarized
26 a number of specific instances associated with industrial releases of Hg in past years.
27 High concentrations of Hg in the air of China's cities have also been reported in several
28 studies (Fang et al., 2001; Feng et al., 2003, 2004a, 2004b; Liu et al., 2002). **Furthermore,**
29 **Hg concentrations measured in the air of remote areas in China are also significantly**
30 **higher than other remote areas in the Northern Hemisphere (Fu et al., 2009a, 2009b;**
31 **Lindberg et al., 2007; Wan et al., 2009), which suggests contamination of remote areas of**

1 **China by anthropogenic Hg emissions.** Recently, a better understanding of China's Hg
2 emissions has been made. Since 2003, Tsinghua University and Argonne National
3 Laboratory have been developing a comprehensive multiple-year inventory of Hg
4 emissions from anthropogenic sources in China, following the precedent of the Asian
5 TRACE-P emission inventory (Streets et al., 2003a, 2003b). We have developed a
6 detailed assessment of emissions from coal combustion with a new technology-based
7 treatment for each province, supplemented with estimates of emissions from all other
8 significant man-made sources (no natural sources or re-emission). Hg emissions are
9 speciated using technology-specific factors and gridded for use in atmospheric models. A
10 detailed estimation of China's mercury emissions by province for the year 1999 is
11 presented in Streets et al. (2005), and the trends in anthropogenic Hg emissions in China
12 from 1995 to 2003 are presented in Wu et al. (2006). Hg emissions were stable at around
13 540 (± 20) Mg during the period 1995-2000, but increased quickly to nearly 700 Mg in
14 2003. Coal combustion and nonferrous metals production continue to be the two leading
15 mercury sources in China, together contributing ~80% of total mercury emissions over
16 the past decade (Wu et al., 2006).

17 However, many uncertainties still remain in our knowledge of primary
18 anthropogenic releases of mercury to the atmosphere in China. Specifically, we are
19 lacking actual measurements of Hg emission rates and Hg species profiles from Chinese
20 combustors and the capture of Hg in Chinese emission control devices. There are even
21 large discrepancies in estimates of the typical Hg content of coal in many provinces.
22 Based on a preliminary uncertainty analysis with coefficients of variation of various
23 contributing factors and combining total uncertainties with quadrature average,
24 approximately $\pm 40\%$ for power plants, $\pm 60\%$ for industrial coal use, and even larger
25 uncertainty ranges for other sources were estimated for Hg emissions in China in 1999
26 (Streets et al., 2003a, 2005). Further, the gap between Hg emission inventories and
27 atmospheric observations (Friedli et al., 2004; Jaffe et al., 2005; Pan et al., 2006; Weiss-
28 Penzias et al., 2007; Wu et al., 2006), has been driving an urgent need to better
29 understand the uncertainties embedded in the Hg emission estimate.

30 In this paper, we present a new comprehensive sub-module within our previous Hg
31 emission inventory (Streets et al., 2005; Wu et al., 2006) for estimation of Hg emissions

1 from coal-fired power plants in China as an uncertainty case study. With this effort,
2 stochastic simulation capability is incorporated into the model to address uncertainties.
3 Distribution functions are built for the key parameters, such as the Hg content of coal and
4 the Hg removal efficiencies of major control technologies. We take into account
5 probability distributions of those key input parameters, and produce the Hg emission
6 results in the form of statistical distributions. For this paper, the uncertainty results in Hg
7 emissions for the year 2003 are presented and discussed.

9 **2. Methodology, data sources, and key assumptions**

10 A new sub-module has been developed to conduct uncertainty analysis of Hg
11 emissions from coal-fired power plants in China. Mercury emissions are calculated using
12 coal consumption data and detailed Hg emission factors. The basic concept of the Hg
13 emission calculation is described by the equation:

$$14 \quad E = \sum_i \sum_j [ef_{i,j} \cdot A_{i,j} \cdot F_{RELj} \cdot (1 - F_{REMj})] \quad (1)$$

15 where E is the Hg emission; $ef_{i,j}$ is the Hg content of coal as burned; $A_{i,j}$ is the amount of
16 coal consumption; F_{RELj} is the fraction of Hg released to the atmosphere; F_{REMj} is the
17 fraction of Hg removed by emission control devices; j is the combustor type with/without
18 emission control devices; and i is the province.

19 The new module has up-to-date information regarding mercury content in coal by
20 province, coal washing and cleaning, coal consumption by province, mercury removal
21 efficiencies by control technology or technology combinations, share of each control
22 technology to coal power capacity in China, etc. As these parameters used in our new
23 sub-module involve uncertainties, we establish probability distribution functions for them
24 on the basis of the available data. Many of these were already collected and published in
25 our previous papers (Streets et al., 2005, 2008; Wu et al., 2006), supplemented with other
26 newly available test data from various researchers. To accomplish this, the data from
27 each source type are read into Crystal BallTM, a statistical software package, which, based
28 on the number of data points and scatter of the data, attempts to fit a distribution about
29 the data for that source type. In Crystal BallTM, a mathematical fit is performed to
30 determine the set of parameters for each set of standard distribution functions that best

1 describes the characteristics of the data. In this study, the goodness-of-fit is determined
2 using the Chi-square test and Anderson-Darling test. The Chi-square test is the oldest and
3 most common goodness-of-fit test. This test gauges the general accuracy by breaking
4 down the distribution into areas of equal probability and compares the data points with
5 each area to the number of expected data points. Generally, a p-value greater than 0.5
6 indicates a close fit. However, for those parameters with a long tail of the distribution, we
7 apply the more appropriate Anderson-Darling method instead. This goodness-of-fit test
8 method closely resembles the Kolmogorov-Smirnov test, except that it weights the
9 differences between the two distributions at their tails greater than at their mid-ranges.
10 We use this test when we need a better fit at the extreme tails of the distributions, such as
11 the lognormal distribution for Hg content in raw coal (see Fig. 1 as an example). Ideally,
12 statistics based on real-world measurements would be employed for this purpose.
13 However, limited data availability sometimes prevents us from taking this approach. In
14 these cases, judgments are made to develop subjective distribution functions
15 (Subramanyan et al., 2008). All distributions are visually examined for reasonableness.

16 By using Crystal BallTM, the sub-module incorporates a Monte Carlo stochastic
17 simulation approach to take into account the probability distributions of key input
18 parameters and produce the mercury emission results in the form of a statistical
19 distribution. The Monte Carlo sampling technique is one of the most widely used
20 techniques for sampling from a probability distribution, which is based on a pseudo-
21 random generator used to approximate a uniform distribution (i.e., having equal
22 probability in the range from 0 to 1). The specific values for each input variable are
23 selected by inverse transformation over the cumulative probability distribution. The
24 Monte Carlo sampling technique also has the important property that the successive
25 points in the sample are independent. To obtain reliable distribution results, the stochastic
26 simulations were run up to 4,000 samples for each forecast variable, e.g., the total Hg
27 emission for Guizhou Province. At the same time, a precision control confidence level
28 (95.0% in this study) was set up to ensure the quality of output results.

30 **2.1 Mercury content of coal**

1 A variety of measurement data, including the new USGS database and different
2 Chinese databases (Huang and Yang, 2002; Feng et al., 2002; Streets et al., 2005; USGS,
3 2004; Wang et al., 2000; Wu et al., 2006; Zhang et al., 1999), were gathered to build the
4 distribution functions for the Hg content of raw coal by province. As bituminous coal is
5 the dominant coal type for coal-fired power plants in China, we exclude other coal
6 samples (e.g., anthracite and lignite) in our databases. Fig. 1 shows example distribution
7 curves for the Hg content of raw bituminous coal in two provinces (Guizhou and Shanxi).
8 Using the Chi-squared test and the Anderson-Darling test, a lognormal distribution
9 function is found to best fit the data for the two provinces. The key characteristics (such
10 as P10, P50 and P90 values) of the distribution functions for mercury content of raw coal
11 by major provinces in China are summarized in Table 1. For other provinces that lack
12 sufficient coal samples, we use two ways to solve the problem. First, for those provinces
13 we believe are in similar coal geological regions, we apply the calculated distribution
14 curve from a related province to the province that lacks data. For example, we apply the
15 distribution curve of Anhui for Zhejiang. For provinces that do not clearly have
16 comparable coal geology, we simply apply the national-average distribution curve.

17 It should be noted that the P50 values are significantly lower than the mean values
18 used in our previous papers (Streets et al., 2005; Wu et al., 2006). For example, the P50
19 value of the Hg content of coal in Guizhou Province is 0.36 ppm, whereas its mean value
20 is 0.51 ppm, which is much higher. This is because of the nature of the lognormal
21 distribution curve, which has a long tail (see Fig. 1, e.g., the P90 value of Hg content for
22 Guizhou's coal is as high as 1.05 ppm). Although there are quite a few coal samples that
23 have high Hg content, we believe the dominant Chinese coal mines have lower Hg
24 content (see Table 1).

25

26 **2.2 Coal consumption by province**

27 The data on coal consumption for power plants are primarily from two data sources:
28 China Energy Statistics Yearbook (2005) and China Power Industry Yearbook (2004).
29 The two datasets match reasonably well, within $\pm 5\%$ for the majority of provinces.
30 Because there is uncertainty in these estimates, but not as a result of measurement error
31 that can be statistically sampled, a triangular distribution function is built for each

1 province. It should be noted that the selection of a triangular distribution is a subjective
2 judgment. Due to the limitation of data (here we only have two data samples), neither the
3 Chi-square test nor the Anderson-Darling test could be used to build the distribution
4 curve. Usually, the triangular or normal distribution function is applied for limited data
5 samples (Brinkman et al., 2005). For this parameter, we set the two statistical data points
6 of coal consumption by each province as the minimum and the maximum values, and the
7 average of the two as the most likely value to build the triangular distribution function.
8 Two examples for raw coal consumption in Guizhou Province and Shanxi Province are
9 shown in Table 2.

11 **2.3 Mercury removal efficiency by control technology**

12 In the model, the Hg removal efficiencies of three post-combustion control
13 technologies or technology combinations are built with distribution functions. They are
14 PM scrubbers, electrostatic precipitators (ESPs), and ESPs plus flue-gas desulfurization
15 (FGD). In 2003, the share of ESP installation in the total coal-fired power capacity was
16 ~95% nationwide, and the majority of the remaining 5% was installed with PM scrubbers.
17 Since the mid 1990's, FGD began to be installed in power plants to reduce SO₂ emissions
18 in China. By the end of 2003, the FGD capacity had reached 6.9 GW, ~2.5% of total
19 coal-fired generating capacity. It should be noted that the shares of control technologies
20 vary significantly from one province to another. For example, all the coal power in
21 Beijing was installed with ESP in 2003, among which ~24% was supplemented with
22 FGD. In this study, we apply the provincial-level technology data for our emission
23 inventory calculations.

24 At the present time there are 25 test values for ESPs, of which 18 are from the US
25 EPA database (US EPA, 1997, 2002; Srivastava et al., 2006) and seven are Chinese test
26 data from various sources (Chen et al, 2007; Wang et al., 2000; Wang et al., 2010a,
27 2010b; Zhang et al., 2008; Zhou et al., 2008; Zhu et al., 2002). **These Chinese tests**
28 **applied standardized test protocols, such as the Ontario Hydro Method, which provides a**
29 **good basis for comparison with U.S. test data.** It should be noted that all the test results
30 are for bituminous coal. The removal efficiency of the seven Chinese tests ranges from
31 20.4% to 41.0%, with an average of 30.4%, which matches well with the average of the

1 U.S. test data, 29.4%. A Weibull distribution is found to fit the best for the dataset with
2 both the Chi-squared test and the Anderson-Darling test, as shown in Fig. 2. The best
3 estimate (P50 value) is 29.4% for Hg removal efficiency by ESP, ranging from 8.8%
4 (P10) to 50.0% (P90). It should be noted that the distribution curve is truncated at the left
5 side (see Fig. 2), because the Hg removal efficiency cannot be less than 0.

6 The data for PM scrubbers and for ESPs plus FGD are scarce, so we have used the
7 limited data from U.S. tests to build the function curve. **It should be noted that even the**
8 **U.S. data samples for scrubbers and ESPs plus FGD are not enough to build such a**
9 **distribution curve, so we assume that the Weibull distribution curve (which best fits for**
10 **ESPs) fits for these two technologies. For FGD plus ESP, we have two data samples**
11 **available. We set the lower number as the P10 value, the higher as the P90 value, and the**
12 **average of the two as the P50 value. For PM scrubbers, we follow the same procedure.**
13 These curves need to be updated as soon as more test data become available. For pre-
14 combustion control technology, we apply a Weibull distribution function for coal
15 washing, which is based on limited test data (Streets et al., 2005; Wu et al., 2006). The
16 best estimate values (P50) are 6.5%, 69.0%, and 25.0% for Hg removal efficiency by
17 scrubber, ESP+FGD, and coal washing, respectively. The key characteristics for each of
18 the above distribution curves are summarized in Table 2.

20 **2.4 The ratio of clean coal output to raw coal input**

21 In 2003, clean coal contributed 2.2% of total coal consumption for the power sector
22 in China. The ratios of cleaned coal output to raw coal input are derived from the Energy
23 Statistics Yearbook (2005). A logistic distribution function is found to fit the best for the
24 dataset. The P10, the best estimate (P50), and P90 values are 0.67, 0.80, and 0.92,
25 respectively, for the ratio. Table 2 presents the key characteristics for this parameter.

27 **2.5 Mercury speciation split**

28 The limited Chinese test data on coal-fired power plant boilers show significant
29 differences in Hg speciation. The key finding is that the share of Hg⁰ to total Hg in
30 Chinese boilers is much higher than that found in U.S. boilers. **For example, the share of**
31 **Hg⁰ is 26% (±15%) on average for the outlet of ESPs tested in the 18 U.S. boilers (US**

1 EPA, 1997, 2002; Srivastava et al., 2006), while this same ratio increases to 48% ($\pm 11\%$)
2 on average for six Chinese boilers (Chen et al, 2007; Wang et al., 2010a; Zhu et al., 2002).
3 The chlorine content of coal could be a major factor causing this difference. Zhang et al.
4 (2008) indicate that the chlorine content of Chinese coals is generally lower than U.S.
5 coals. Chlorine can enhance the transformation from Hg^0 to divalent Hg (Hg^{2+}) (Chen et
6 al., 2007; Srivastava et al., 2006). The other finding is that the share of particulate Hg
7 (Hg^p) to total Hg for those measurements taken from the inlets of ESPs is significantly
8 lower from Chinese tests compared with U.S. tests (19% on average for Chinese data vs.
9 45% on average for U.S. data). We do not yet know the reason for this difference, but we
10 suspect that the high share of Hg^0 could be a factor. More test data are necessary to
11 support the two findings. The built-in distribution curve is based on the limited Chinese
12 test data only (Chen et al, 2007; Wang et al., 2010a; Zhu et al., 2002), and we assume that
13 the triangular distribution function best fits the dataset. This may be subject to change
14 when more test data become available. Table 2 summarizes the key characteristics of the
15 distribution curves for Hg^{2+} and Hg^p . For example, the most likely estimates for the share
16 of Hg^{2+} and the share of Hg^p to total Hg are 51% and 2%, respectively, for the outlet of
17 ESPs.

18

19 **3. Results and discussion**

20 With the Crystal BallTM software, we apply the Monte Carlo method to perform the
21 stochastic simulations. To get reliable outputs, we set the sampling number as 4,000. All
22 the results of total Hg, Hg^0 , Hg^{2+} , and Hg^p by each province are now represented by
23 distribution curves instead of single points. Figure 3a-d presents the output distribution
24 curves for emissions of total Hg, Hg^0 , Hg^{2+} , and Hg^p , respectively, from coal-fired power
25 plants in China in 2003. We also illustrate all the results for a specific province, Guizhou,
26 as an example, which are shown in Fig. 4a-d.

27 The curves show a wide range in uncertainties. For example, the total Hg emissions
28 in 2003 for the whole of China vary from a minimum of ~20 Mg to a maximum of ~280
29 Mg (see Fig. 3a), an order of magnitude different. The difference is even larger at the
30 province level. Total Hg emissions in 2003 for Guizhou range from a minimum of ~0.2
31 Mg to a maximum of ~30 Mg (see Fig. 4a), two orders of magnitude different. The

1 largest source of uncertainty is this factor: *Hg content in coal*. From Fig. 3 and Fig. 4., we
2 can see that the output distribution curves are close to “lognormal” shape; and this shape
3 is especially clear for some specific provinces, such as Guizhou. From our distribution
4 function database for the key input parameters, only the parameter *Hg content in coal*
5 shows a lognormal distribution. The long tails in the output curves for emissions of total
6 Hg and its three species for Guizhou are no doubt caused by the distribution of the Hg
7 content of Guizhou’s coal, which is highly variable (the difference between the maximum
8 and minimum is as high as ~200). **Furthermore, we designed several scenarios to evaluate**
9 **the contributions of various parameters to the uncertainty in Hg emissions (see Table 3).**
10 **As shown for Scenario 1 in Table 3, the parameter, *Hg content in coal*, plays the**
11 **dominant role in determining the best estimate of total Hg emissions in China and**
12 **Guizhou Province, as well as the uncertainty range.**

13 In previous studies (Streets et al., 2005; Wu et al., 2006), we thought that the
14 activity level contributed a somewhat similar uncertainty as the emission factor. However,
15 from this study, at least for the power-plant sector, this is not the case. First, the
16 differences in coal consumption for most provinces are quite small (within $\pm 5\%$). Second,
17 the clean coal consumption in the power sector in China is small, only 2.2%, although
18 this key parameter, *the ratio of cleaned coal output to raw coal input*, involved in
19 calculating Hg emissions from clean coal, shows a moderate uncertainty range (-
20 16%/+15%). **As a result, the activity level in the power sector in China plays only a small**
21 **role in the uncertainty estimate, as confirmed by the results of Scenarios 4 and 5 in Table**
22 **3.**

23 Hg removal efficiency is also a major factor affecting the uncertainty. This is
24 especially true for Hg removal efficiency by ESP, as ESP is the dominant control device
25 in China’s coal-fired plants. The uncertainty range for this parameter is wide, at $\pm 70\%$.
26 **As shown by Scenario 2 in Table 3, the parameter, *Hg removal efficiency by ESP*, ranks**
27 **as the second most important parameter affecting the uncertainty range of total Hg**
28 **emissions in China and Guizhou Province.** In the future, Hg removal efficiency by ESP
29 plus FGD could also play an important role, because the share of FGD to total power
30 capacity will reach over 80% within the next decade (Wang et al., 2010b). The current
31 uncertainty range for Hg removal efficiency by ESP plus FGD is not large, at $\pm 9\%$;

1 however, it should be noted this range is based on a very limited dataset. The uncertainty
2 level could become larger as more test data come available. The two parameters, *Hg*
3 *removal efficiency by FGD* and *Hg removal efficiency by coal washing*, contribute a
4 small share of the uncertainty level in the output distribution curve, as these two control
5 technologies are not popular in the power sector in China.

6 With the output distribution curves, we can summarize the statistical results for
7 each province and for the whole of China in four separate charts: total Hg, Hg⁰, Hg²⁺ and
8 Hg^P, as shown in Fig. 5a-d. The bar represents the P50 value of emissions, and the line
9 superimposed on each bar represents the range between the P10 and P90 values. Thus, for
10 the whole of China in 2003, (a) the best estimate for total Hg emissions from coal-fired
11 power plants is 90.5 Mg, with the uncertainty range from 57.1 Mg (P10) to 154.6 Mg
12 (P90); (b) the best estimate for Hg⁰ emissions is 43.0 Mg, with the uncertainty range from
13 25.6 Mg (P10) to 75.7 Mg (P90); (c) the best estimate for Hg²⁺ emissions is 45.4 Mg,
14 with the uncertainty range from 27.3 Mg (P10) to 80.2 Mg (P90); and (d) the best
15 estimate for Hg^P emissions is 1.8 Mg, with the uncertainty range from 1.0 Mg (P10) to
16 3.2 Mg (P90). The previous point estimate for total Hg emissions from the power sector
17 in China in 2003 was 100.1 Mg (Wu et al., 2006), 10.6% higher than our new best
18 estimate. **The lower P50 value as compared with the previous mean value is primarily**
19 **attributed to the factor, *Hg content in coal*. The use of lognormal distribution curves for**
20 **this parameter shifts the P50 value to a lower number.** It should be noted that the
21 uncertainty range is large, for example, -37%/+71% for the total Hg emission estimate.
22 The larger uncertainty at the right high-value bound (i.e., +71%) is primarily due to the
23 long tail of the distribution of the Hg content of coal. Hg⁰ emissions, 43.0 Mg, are much
24 higher than our previous estimate (Wu et al., 2006), which was 20.0 Mg, because of
25 incorporation of the new measured speciation data. This may help to close at least a
26 portion of the gap between the Hg⁰ emission inventory estimate and Hg⁰ atmospheric
27 observations in previous field studies (Friedli et al., 2004; Jaffe et al., 2005; Pan et al.,
28 2006; Weiss-Penzias et al., 2007). Conversely, Hg²⁺ emissions in this study, 45.4 Mg, are
29 considerably lower than our previous estimate (78.1 Mg). Hg^P emissions are quite close,
30 1.8 Mg vs. 2.0 Mg.

1 The top five provinces in total Hg emissions from the power sector in 2003 are as
2 follows: Shandong (6.9 Mg, -53%/+116%), Henan (6.2 Mg, -64%/+169%), Jiangsu (5.5
3 Mg, -53%/+130%), Guizhou (5.4 Mg, -68%/+200%), and Liaoning (5.0 Mg, -
4 50%/+121%). These five provinces contribute about one-third of the total national
5 emissions. It should be noted that the uncertainty range (especially at the right high-value
6 bound) at the provincial level is significantly higher than for the national estimate. For
7 example, the P90 value for Guizhou's total Hg emission estimate is as high as 16.2 Mg,
8 two times higher than the best estimate (P50 value). Further, the uncertainty level varies
9 from one province to another. The larger uncertainty range for provinces such as Guizhou
10 and Henan is primarily attributed to high uncertainty in the Hg content of coal there.

11

12 **4. Conclusions**

13 The results of stochastic simulations from this study show that the majority of the
14 uncertainty in Hg emission estimation results from one key factor, the mercury content of
15 coal. In addition, the mercury removal efficiency of ESP also plays an important role in
16 Hg uncertainty. As China is accelerating the installation of FGD systems to control SO₂
17 emissions, the Hg removal efficiency by ESP plus FGD could also be another major
18 factor in the near future.

19 The best estimate for total Hg emissions from coal-fired power plants in China in
20 2003 is 90.5 Mg, with the uncertainty range from 57.1 Mg (P10) to 154.6 Mg (P90). The
21 best estimate is about 10% lower than our previous point estimate for China (100.1 Mg),
22 and the uncertainty range is large (-37%/+71%). The best estimate for Hg⁰ emissions,
23 43.0 Mg (-40%/+76%), is 115% higher than our previous point estimate (20.0 Mg).
24 Conversely, the best estimate for Hg²⁺ emissions in this study, 45.4 Mg (-40%/+77%), is
25 43% lower than our previous estimate (78.1 Mg). Hg^P emissions are small and quite
26 similar in both studies.

27 The top five provinces in total Hg emissions from power sector in 2003 are
28 Shandong (6.9 Mg, -53%/+116%), Henan (6.2 Mg, -64%/+169%), Jiangsu (5.5 Mg, -
29 53%/+130%), Guizhou (5.4 Mg, -68%/+200%), and Liaoning (5.0 Mg, -50%/+121%).
30 The uncertainty range at the provincial level is significantly higher than that for the
31 national estimate. The uncertainty level varies significantly from one province to another.

1 The larger uncertainty range for provinces such as Guizhou and Henan is primarily
2 attributed to high variability in the Hg content of coal in those provinces.

3 Uncertainties exist in all of the variables involved in the calculation of Hg
4 emissions, and some of them are quite large. Thus a stochastic simulation, such as we
5 have adopted in this work, is better than a deterministic approach and comes closer to a
6 true reflection of reality. Currently, the database of Hg content in bituminous coal for
7 some major provinces (such as Shanxi) has been well established due to extensive
8 measurements. Similarly, the Hg profiles of ESP for bituminous coal in pulverized coal
9 boilers have recently been investigated in detail. However, many other parameters are
10 poorly known due to lack of measurements. More effort is needed to gather information
11 on important parameters, if the emission inventory is to be improved. For example, we
12 are still lacking field test profiles to build distribution curves for control technologies
13 such as FGD plus ESP, which is becoming the leading control technology in China's
14 power plants. Also, the new technology for NO_x emission reduction, selective catalytic
15 reduction (SCR), is beginning to penetrate the power sector in China, and its Hg removal
16 mechanisms need to be explored. As soon as these data become available, an update of
17 our emission inventory will be performed.

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Tables

Table 1. Key characteristics for distribution functions of mercury content of raw coal by major provinces in China (mercury content in ppm)

Parameters	Number of samples	Distribution function type	P10 ^a	P50 ^a	P90 ^a	Mean
Anhui	12	Lognormal	0.090	0.210	0.490	0.261
Guizhou	46	Lognormal	0.121	0.357	1.051	0.509
Hebei	13	Lognormal	0.036	0.111	0.343	0.164
Heilongjiang	12	Lognormal	0.040	0.077	0.150	0.088
Henan	18	Lognormal	0.058	0.171	0.505	0.245
Liaoning	8	Lognormal	0.043	0.134	0.418	0.189
Nei Mongol	7	Lognormal	0.098	0.192	0.379	0.221
Shaanxi	9	Lognormal	0.008	0.051	0.317	0.141
Shandong	18	Lognormal	0.060	0.141	0.330	0.176
Shanxi	69	Lognormal	0.024	0.091	0.347	0.157
Sichuan	17	Lognormal	0.050	0.114	0.260	0.140
China	218^b	Lognormal	0.029	0.105	0.376	0.172

^a P10 values mean that there is a probability of 10% that the actual result would be equal to or below the P10 values; P50 mean that there is a probability of 50% that the actual result would be equal to or below the P50 values; and P90 mean that there is a probability of 90% that the actual result would be equal to or below the P90 values.

^b All the 218 samples are from the USGS database (USGS, 2004).

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Table 2. Key characteristics for distribution functions of coal consumption, Hg removal efficiency, and other key parameters

Parameters	Number of samples	Distribution function type	P10 ^a	P50 ^a	P90 ^a	Mean
Coal consumption, 10 ³ Mg						
1) Guizhou	2	Triangular ^b	21,647	21,669	21,691	21,669
2) Shanxi	2	Triangular ^b	45,285	46,575	47,866	46,575
Hg removal efficiency by control technology, %						
1) PM Scrubber	2	Weibull	4.3	6.5	8.7	6.5
2) ESP	25	Weibull	8.8	29.4	50.0	30.4
3) FGD+ESP	2	Weibull	63.0	69.0	75.0	69.0
4) coal washing	5	Weibull	5.0	25.0	64.0	30.0
The ratio of clean coal output to raw coal input, %	20	Logistic	67	80	92	79
Hg speciation split, %						
1) no control, Hg ²⁺	6	Triangular ^b	26	36	46	36
Hg ^p	6	Triangular ^b	5	25	45	25
2) ESP, Hg ²⁺	5	Triangular ^b	32	51	70	51
Hg ^p	5	Triangular ^b	1	2	3	2
3) FGD+ESP, Hg ²⁺	4	Triangular ^b	0	12	24	12
Hg ^p	4	Triangular ^b	0	0.5	1	0.5

^a P10 values mean that there is a probability of 10% that the actual result would be equal to or below the P10 values; P50 mean that there is a probability of 50% that the actual result would be equal to or below the P50 values; and P90 mean that there is a probability of 90% that the actual result would be equal to or below the P90 values.

^b These values are for the minimum, the most likely, and the maximum values for the triangular distribution function instead of P10, P50, and P90 values.

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Table 3. The contributions of various parameters to the uncertainty in Hg emissions in China and Guizhou Province in 2003

	China		Guizhou Province	
	P50, Mg	Uncertainty range	P50, Mg	Uncertainty range
Baseline ^a	90.5	-36.8%/+70.9%	5.4	-68.0%/+199.8%
Scenario 1 ^b	90.0	-29.5%/+60.4%	5.4	-65.3%/+194.5%
Scenario 2 ^c	102.2	-27.7%/+25.3%	7.9	-27.9%/+25.5%
Scenario 3 ^d	101.5	-0.3%/+0.2%	7.9	-0.1%/+0.1%
Scenario 4 ^e	101.6	-1.0%/+0.7%	7.9	-0.8%/+0.5%
Scenario 5 ^f	101.4	-0.4%/+0.4%	7.9	-0.1%/+0.0%

^a Baseline is a complete stochastic simulation with all parameters in this study built with distribution functions.

^b Scenario 1 only sets the parameter, *Hg content in coal*, with distribution functions. The distribution functions for other parameters are removed. The results show the contribution of *Hg content in coal* to uncertainties of total Hg emissions.

^c Scenario 2 only sets the parameter, *Hg removal efficiency by ESP*, with distribution functions.

^d Scenario 3 only sets the parameter, *Hg removal efficiency by other controls, such as PM scrubber*, with distribution functions.

^e Scenario 4 only sets the parameter, *coal washing and cleaning*, with distribution functions.

^f Scenario 5 only sets the parameter, *coal consumption*, with distribution functions.

1 **Figures Captions**

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4 **Figure 1.** Distribution function curves for Hg Content of Raw Coal, (a) Guizhou
5 Province; and (b) Shanxi Province.

6
7 **Figure 2.** Distribution function curve for Hg removal efficiency by ESP.

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9 **Figure 3.** The output distribution function curves for emissions of a) total Hg, b) Hg^0 , c)
10 Hg^{2+} , and d) Hg^p , from coal-fired power plants in China in 2003.

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12 **Figure 4.** The output distribution function curves for emissions of a) total Hg, b) Hg^0 , c)
13 Hg^{2+} , and d) Hg^p , from coal-fired power plants in Guizhou in 2003.

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15 **Figure 5.** The best estimate and its uncertainty range of emissions of a) total Hg, b) Hg^0 ,
16 c) Hg^{2+} , and d) Hg^p , from coal-fired power plants for the whole of China and by each
17 province in 2003. The bar represents the P50 value, i.e., the best estimate of emissions,
18 and the line superimposed on each bar represents the range between the P10 and P90
19 values.

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