

Emission of Hg from Coal Consumption in China and Its Summertime Deposition Calculated by CMAQ-Hg

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ABSTRACTS

Mercury (Hg) emission from coal combustion in China is estimated in this study and its deposition calculated using the Models-3 Community Multi-scale Air Quality (CMAQ-Hg) modeling system with meteorological fields calculated by MM5. Three kinds of Hg speciation were included in the models: particle-bound mercury (PHg), gaseous elemental mercury (GEM) and gas oxidized mercury (RGM). Much of the Hg was released as RGM from coal combustion in China, accounting for 58.4 - 66.8% of total emissions. The large proportion of RGM in overall Hg emission also led to large Hg deposition. Based on simulations, Hg deposition originating from coal combustion reaches as high as 2 - 6 g m⁻² month⁻¹ in southwestern China (particularly Guizhou Province), the industrialized and populated regions of east central and coastal China, and Liaoning Province in the northeast.

Key words: Hg emission, Deposition, CMAQ-Hg, Coal combustion

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1. INTRODUCTION

Human activity, including: the combustion of fossil fuels, incineration of waste, gold mining and other applications of Hg have significantly increased the emission of Hg into the atmosphere. Determination of Hg in peat cores in Norway has proved that the Hg accumulation over the last 100 years has been about 15 times higher on average than pre-industrial levels (Steinnes and Sjobakk 2005). Coal combustion is the major Hg sources from human activity (US EPA 1997; European Commission 2001), especially in China where coal is the main energy source accounting for 68% of energy production in 2003 (CESY 2005). Therefore, Hg emitted from coal combustion is an important contributor to atmospheric mercury budgets and plays important role in the Hg cycle in China.

Hg emitted from coal combustion mainly consists of gaseous elemental mercury (GEM), gas oxidized mercury (RGM) and particle-bound mercury (PHg). The degree of Hg speciation in power generation depends on operating

conditions such as: the type of coal, flue gas temperature and components, and the configuration of air pollution control devices etc. The atmospheric residence time of GEM is 0.5 - 2 yr, which makes transportation on a hemispherical scale possible and emissions in any continent can thus contribute to deposition in other continents. Any RGM that is directly emitted to the atmosphere is expected to deposit efficiently on a local or regional scale near major sources because of its solubility (Schroeder and Munthe 1998; Lin and Pehkonen 1999). PHg has a shorter atmospheric lifetime than GEM and will deposit via wet or dry processes within roughly 100 to 1000 kilometers (Keeler et al. 2005). So the composition of Hg speciation decides its processes in the atmosphere.

In this study, we estimate Hg emission from coal combustion in China and calculate its deposition by CMAQ-Hg (Byun and Ching 1999; Bullock and Brehme 2002). The impact of meteorological processes, gas-phase chemistry, aqueous-phase chemistry, aerosol processes, heterogeneous chemistry and deposition processes upon Hg transport, transformation and deposition are considered comprehensively. Two cases of Hg speciation scenarios for coal-fired

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power plants are used to study their influence on Hg deposition.

2. MERCURY EMISSIONS

Coal consumption was 1.637 billion tons in China in 2003. The average Hg concentration in Chinese coal ranges from 0.02 to 0.54 mg kg⁻¹ (Zhang et al. 1999; Wang et al. 2000; Feng et al. 2002; Huang and Yang 2002; USGS 2004). In this work, Hg content of raw coal in China by province is from the US Geological Survey (USGS 2004) (see also Jiang et al. 2005; Streets et al. 2005). The calculation of Hg emission from coal combustion is based on the work of Wang et al. (2000) and Jiang et al. (2005). The basis of Hg emission calculation is described by the equation:

$$Q = \sum_i \sum_j c_{i,j} M_{i,j} R_{i,j} (1 - P_{i,j}) \quad (1)$$

Where Q is the Hg emission; $c_{i,j}$ is the Hg content of coal as burned; $M_{i,j}$ is the amount of fuel consumption; $R_{i,j}$ is the fraction of Hg released to the atmosphere; $P_{i,j}$ is the fraction of Hg removed by emission control devices; j is the combustor type with/without emission control devices; and i is the province. Data relating to coal consumption is from the China Energy Statistical Yearbook for 2003 (NBSC 2005), which is compiled by different sectors at a provincial level. Two Hg speciation scenarios for coal-fired power plants are used: one is from the field measurement work of Chen et al. (2007) (Case 1 in Table 1); another is from US EPA (2002) (Case 2 in Table 1). The compositions of Hg speciation from other types of combustors all come from the US EPA (2002) (Table 1). The distribution of esti-

mated Hg emission from coal consumption in China is shown in Fig. 1. Total Hg emission from coal combustion in China is estimated to be 182.2 t in 2003 (Table 2). RGM takes up 58.4% (Case 1) to 66.8% (Case 2) among the total emitted Hg. PHg emission is also large in China due to heavy burning of coal in residential and small industrial settings without PM controls. Hg emission from coal-fired power plants is 72.4 t, accounting for 39.7% of total Hg emission.

3. MODELS DESCRIPTION

CMAQ (Byun and Ching 1999) is an Eulerian-type model developed in the US Environmental Protection Agency to address tropospheric ozone, acid deposition, visibility. In this model, particulate matter and other pollutant issues in the context of a “one atmosphere” perspective are

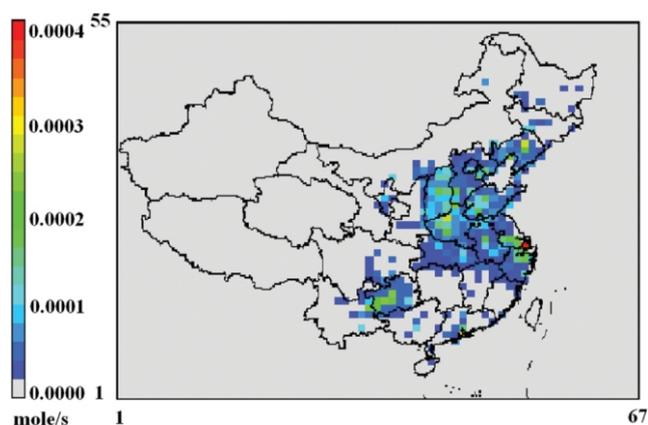


Fig. 1. Distribution of mercury emission from coal consumption in China in 2003.

Table 1. Speciation of total Hg for each major source type (as fraction of the total).

Source category	GEM	RGM	PHg
1. Power plants			
Case 1*	0.40	0.57	0.03
Case 2**	0.20	0.78	0.02
2. Industrial use**			
(a) w/PM control devices	0.2	0.78	0.02
(b) Stoker, w/o PM control devices	0.09	0.03	0.88
(c) Cyclone, w/o PM control devices	0.23	0.35	0.42
(d) Coke production	0.07	0.58	0.35
(e) Coke/burned, w PM control devices	0.20	0.78	0.02
(f) Coke/burned, w/o PM control devices	0.07	0.58	0.35
3. Residential and other uses*	0.09	0.03	0.88

*Chen et al. (2007); **US EPA (2002).

Table 2. Coal consumption and estimated Hg emissions from coal consumption in China in 2003.

		Coal consumption 10 ⁴ ton	GEM ton	RGM ton	PHg ton
Transformation (including: heating supply, coking, coal washing, petroleum and gas works)		37611.38	3.382	28.026	16.912
Farming, forestry, animal husbandry, fishery		1683.33	0.172	0.057	1.680
Industry		35781.21	9.424	36.755	0.942
Construction		577.15	0.053	0.018	0.518
Transportation, postal, and telecommunications		1067.33	0.064	0.021	0.629
Wholesale, retail trade, and catering services		860.42	0.105	0.035	1.024
Residential consumption		8174.71	0.898	0.299	8.785
Electric power plants	Case 1	77976.47	28.963	41.272	2.172
	Case 2		14.481	56.477	1.448
Total	Case 1	163732	43.062	106.483	32.661
	Case 2		28.580	121.688	31.937

confronted in a manner that encapsulates the complex interactions between atmospheric pollutants on regional and urban scales. The CMAQ model was modified by Bullock and Brehme (2002) to include chemistry, transport and deposition of GEM, RGM, and PHg. Transformations of Hg are simulated with four new chemical reactions within the standard CMAQ gaseous chemistry framework and a highly modified cloud chemistry mechanism which includes a compound-specific speciation for oxidized forms of Hg, seven new aqueous-phase Hg reactions, six aqueous Hg chemical equilibria, and a two-way mechanism for the sorption of dissolved oxidized Hg to elemental carbon particles (Bullock and Brehme 2002). This earlier version is updated in a number of areas to improve the underlying science and to address comments from peer reviews (CMAS 2006; Gbor et al. 2006). The three-dimensional meteorological fields used in this study are provided by the Fifth-Generation NCAR/Penn State Mesoscale Model (MM5; Dudhia et al. 1998). The three-dimensional meteorological fields for MM5 were obtained from the European Center for Medium-Range Weather Forecasts (ECMWF) analyzed datasets, and were available every 6 h with 1° resolution.

Wet and dry deposition is simulated for each of the three forms of Hg. Wet deposition rate is calculated based on precipitation information from the CMAQ meteorological processor and physicochemical Hg speciation in the cloud chemistry mechanism. The cloud-water concentration of each pollutant is deposited to the surface based on the simulated rate of precipitation falling from each clouded grid volume during the cloud chemistry time splitting operation. The scavenging of GEM and RGM is based on Henry's Law equilibrium (Bullock and Brehme 2002). The cloud-water concentration of PHg is calculated as the sum of the

concentrations of all sorbed RGM species. The cloud-water concentration of GEM is relatively low compared to the total dissolved and sorbed RGM, and the simulated wet deposition of GEM is minor compared to that of RGM and PHg. Dry deposition rate is calculated based on dry deposition velocity and air concentration information for each of the three forms of Hg. For the Henry's law constant and diffusivity of GEM, we chose 0.11 M atm⁻¹ (Lin and Pehkonen 1999) and 0.1194 cm² s⁻¹ (Massman 1999), respectively. The standard CMAQ dry deposition parameterization for gaseous nitric acid is also used for RGM. PHg dry deposition is simulated based on pre-existing deposition velocity formulations in the standard CMAQ for sulfuric acid. The complete mechanisms with lists of species and reactions in the model are described in detail by Bullock and Brehme (2002) and CMAS (2006).

Hg emission is from coal combustion as stated in the above section, other emissions (including: nitrogen oxides, carbon monoxide, volatile organic compounds etc.) come from the emission inventory of 1:1 specially prepared by scientists at the Center for Global and Regional Environmental Research at the University of Iowa (Streets et al. 2003) to support TRACE-P (Transport and Chemical Evolution over the Pacific) and ACE-Asia (Aerosol Characterization Experiment in Asia). VOCs emissions were apportioned appropriately among the lumped-carbon categories used in CB-IV.

The model domain (shown in Fig. 1) is 5427°E–4455°E for CMAQ centered at (36°N, 105°E) with an 81-km mesh. MM5 and CMAQ have the same model height. For MM5, there are 23 vertical layers in the z coordinates system unequally spaced from the ground to ~23 km, with about 9 layers concentrated in the lowest 2 km of the atmosphere in

order to resolve the planetary boundary layer, while there are 12 levels for CMAQ with the lowest 7 layers being the same as those in MM5.

4. RESULTS AND DISCUSSION

4.1 Simulation Results Analysis

RGM is the main component of simulated Hg deposition in July 2003 (Table 3). It could contribute more than 90% of Hg deposition (Table 3). Two potential reasons for this are: (1) RGM being the main component of Hg emissions from coal consumption; and (2) both dry velocity and wet scavenging of RGM being much higher than that of GEM (Bullock and Brehme 2002; Poissant et al. 2004). After being released into the atmosphere some GEM would be converted to RGM by atmospheric oxidants. The oxidants responsible for such an oxidization process include: ozone, hydroxyl radical, hydrogen peroxide, and reactive halogens such as Cl_2 , Br_2 , BrO , and I_2 . However, the rate constants are very low (Hall 1995; Tokos et al. 1998; Calhoun and Prestbo 2001; Sommar et al. 2001; Ariya et al. 2002; Pal and Ariya 2004a, b), which means GEM has a long atmospheric lifetime (0.5 - 2 yr). Therefore, it is likely that most deposited RGM comes directly from emissions. This means that the greater the proportion of RGM in total Hg emissions, the greater the ratio of Hg deposition to emissions is (Table 3). The ratio of Hg deposition to emission is 38.4% in Case 1, and 43.4% in Case 2.

Simulated distribution of Hg deposition in July 2003 is shown in Fig. 2, together with the monthly mean surface wind field and precipitation map. The results shown in Fig. 2 were obtained from the sum of hourly deposition in July using the emission in Case 1 of Table 2. The patterns of Cases 2 are similar and therefore not shown. Distribution of Hg deposition is the combined effects of Hg pollution levels along with meteorological factors (e.g., wind and precipitation), especially for wet deposition, which is decided mainly by precipitation. The distribution of Hg dry deposition is similar to that of Hg emission, while the distribution of wet deposition is similar to that of precipitation. In July 2003, precipitation in the areas to the north of the Yangtze River is on the high side or close to that of conventional years and severe floods happened in the Huaihe River, while precipitation in the areas to the south of the Yangtze River is on the low side (see also Dong 2003). High precipitation in the Huaihe River led to high wet Hg deposition in these regions (Fig. 2). Regions with high total Hg deposition include: southwestern China (particularly Guizhou Province), the populated and industrial regions of east central and coastal China, and Liaoning Province in the northeast. Hg deposition reached as high as 2 - 6 $\text{g m}^{-2} \text{ month}^{-1}$ in these regions.

Observed Hg deposition in China is at a very high level, especially in cities which produce large Hg emissions (Tan et al. 2000; Feng et al. 2002, 2003). Hg deposition at two

environmental background sites in southwestern China is 4.5 - 4.7 $\text{g m}^{-2} \text{ month}^{-1}$ (Tan et al. 2000), which is higher than Hg deposition observed in North America (Landis and Keeler 2002; NADP 2004) and Japan (Sakata and Marumoto 2005), where Hg deposition is only 1 - 2 $\text{g m}^{-2} \text{ month}^{-1}$. Simulated Hg deposition is in the same order of magnitude with observed Hg deposition at environmental background sites though detailed comparison is not appropriate because simulated Hg deposition in this study is only from coal combustion. Based on the simulations, regions with Hg deposition exceeding 2 $\text{g m}^{-2} \text{ month}^{-1}$ account for about 20% of the total territory. From the work of Streets et al. (2005) we know that Hg emission from coal consumption in China accounts for about 38% of total anthropogenic Hg emissions. Therefore, Hg deposition in China might be at very serious levels.

4.2 Uncertainties

There still exist uncertainties in estimating Hg emission from coal combustion and the simulation of its deposition in this study. The uncertainties are mainly caused by the following factors: (1) Hg content in coals. A number of studies have been carried out to understand Hg emission from coal combustion in China (e.g., Wang et al. 2000; Huang and Yang 2002; Jiang et al. 2005; Streets et al. 2005; Wu et al. 2006), but there still exist great differences among these works. Wang et al. (2000) has estimated that the average mercury concentration in Chinese coals is 0.22 mg kg^{-1} ; while USGS has also measured 331 coal samples from different Chinese regions, suggesting that the mercury concentration ranges from 0.02 to 0.54 mg kg^{-1} , averaged at 0.15 mg kg^{-1} . Based on these investigations, Wang et al. (2000) and Jiang et al. (2005) have calculated the total mercury emissions from coal combustion in China as 213.8 tons (in year 1995) and 161.6/219.5 (based on different databases) tons (in 2000), respectively; (2) Hg speciation from coal combustion. Few field measurements on Hg speciation from coal combustion have been carried out in China, so Hg speciation composition for many source types in this study come from US EPA (2002). Field measurements of Chen et

Table 3. Total Hg emission from coal consumption and simulated deposition in July 2003.

	Emission		Deposition	
	Case 1	Case 2	Case 1	Case 2
GEM	3.657	2.427	0.293	0.184
RGM	9.044	10.335	5.455	6.335
PHg	2.774	2.712	0.198	0.194
Total	15.475	15.475	5.946	6.713

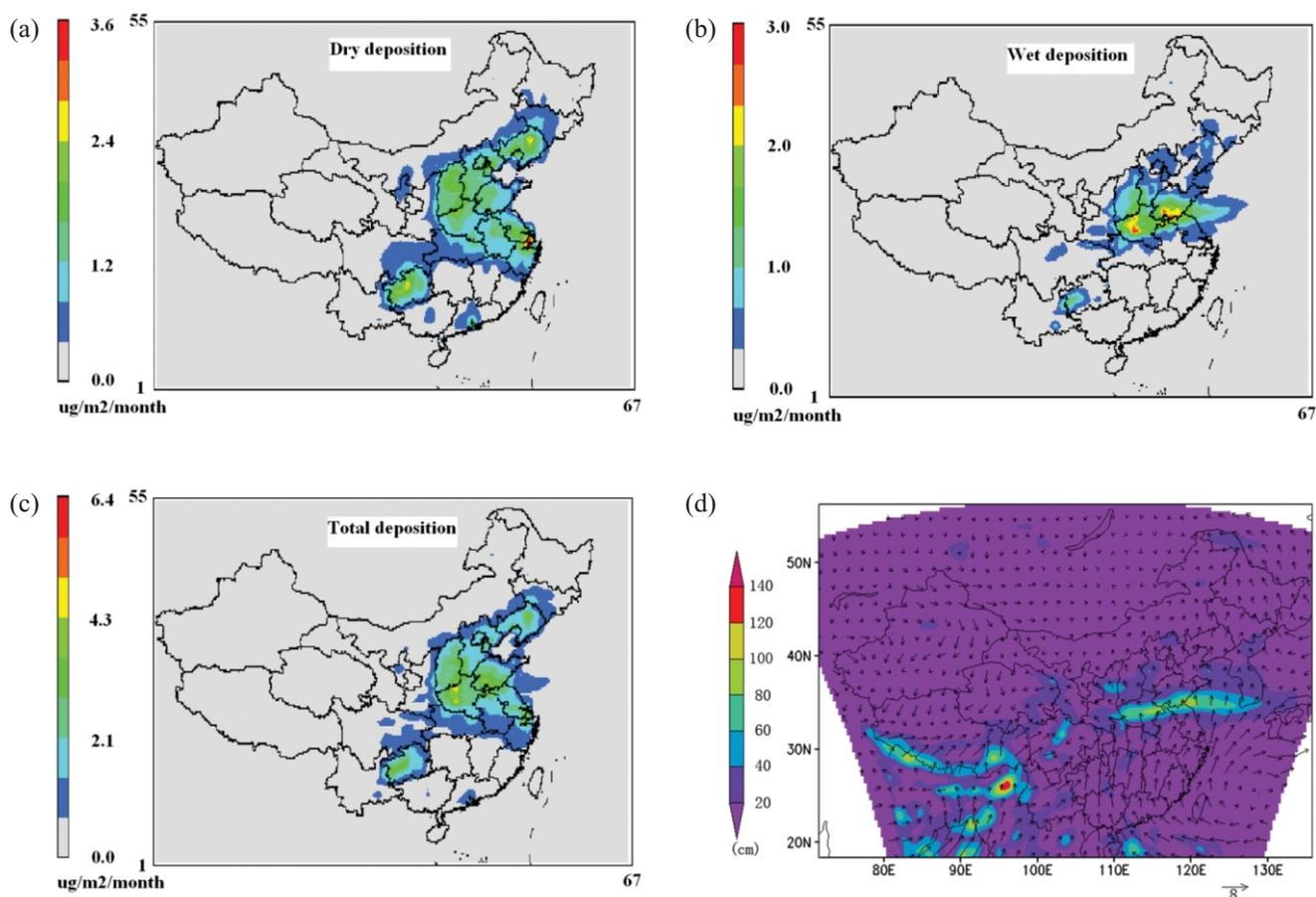


Fig. 2. Simulated distribution of Hg deposition in July 2003 with emission in Case 1 of Table 2. (a) is dry deposition; (b) is wet deposition; (c) is total deposition; (d) is the monthly mean surface wind field and precipitation map of July 2003.

al. (2007) in six power plants of China showed that coal types, especially the chlorine concentration and the basic ash compositions in coal play important roles in the composition of Hg speciation. Therefore Hg speciation composition calculated by the US EPA data does not represent the real situation in China very well; (3) CMAQ-Hg models. Hg chemistry, together with dry and wet deposition processes, might introduce uncertainties into models due to inconsistent kinetic data and a lack of deterministic product identification in the atmosphere (see also Lin et al. 2006).

4.3 Control Measures Discussion

From the above analysis, we know that RGM is the main component in Hg emission from coal combustion and the dominant component in simulated Hg deposition. RGM is soluble and can be removed by 60 - 90% with lime-slurry scrubber solution (Fahlke and Bursik 1995; Lee et al. 2006), so controlling RGM emission might be the most convenient way to decrease Hg emission and deposition. Up to now,

most coal combustors are not equipped with wet flue gas desulfurization (FGD) in China, which tend to dissolve the soluble divalent mercury. Therefore, both Hg emission and deposition would be decreased substantially with the installing of FGD equipment. Based on the Eleventh Five-Year plan of the National Economy and Social Development, more FGD equipment will be installed in coal-fired power plants to control SO₂ emissions (Zhang 2006). So RGM emission from coal combustion should decrease greatly in the near future and Hg deposition will decrease correspondingly.

5. CONCLUSIONS

Hg emission from coal combustion is large in China due to coal consumption, and RGM is the main component in Hg emission from coal combustion. High Hg emissions lead to high Hg deposition. Simulated Hg deposition due to coal combustion reaches 2 - 6 g m⁻² month⁻¹ in southwestern China (particularly Guizhou Province), the populated and

industrialized regions of east central and coastal China, and Liaoning Province in the northeast. RGM is the predominant component in simulated Hg deposition from coal combustion. Its contribution could be above 90%. Most deposited RGM comes from direct emissions. Therefore controlling RGM emission should be the most convenient way to decrease Hg emissions and deposition as lime-slurry scrubber solution can remove RGM by 60 - 90%. Of the total emitted Hg from coal combustion, approximately 40% is deposited in the studied regions in the simulation period and about 60% transported to other regions.

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