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Modeling of atmospheric dispersion of mercury from coal–fired power plants in Japan

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ABSTRACT

The Air Quality Management Division of Ministry of the Environment in Japan selected the maximum level of annual mean air quality standard for mercury as 0.04 μg/m^3. The yearly average atmospheric emissions of mercury from two nearly located point sources, background concentrations of mercury in the atmosphere and one–year meteorological data was used to predict the ambient concentrations of mercury at ground level by atmospheric dispersion modeling. To estimate the mercury concentration in the air of the local area, two different models have been used. The first one is AIST–ADMER model that estimates regional atmospheric distribution of mercury concentration. The second one is METI–LIS model that estimates the atmospheric distribution of mercury concentration in the vicinity of industrial facilities. The annual mean concentration of mercury in the atmosphere was calculated for the central Honshu Island of Japan using the AIST–ADMER model, which served as a background data for the METI–LIS model to calculate atmospheric mercury concentration in the vicinity of industrial facilities. Maximum annual mean atmospheric concentrations of mercury in the vicinity of the two hypothetical coal–fired power plants were calculated as 0.0118 μg/m^3 that was lower than the Japanese annual mean air quality standard for mercury.

Keywords:
Dispersion modeling
Atmospheric mercury concentration
METI–LIS
AIST–ADMER
Mercury emissions

Article History:
Received: 02 June 2011
Revised: 22 January 2012
Accepted: 27 January 2012

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doi: 10.5094/APR.2012.025

1. Introduction

In Japan, mercury was categorized as a hazardous air pollutant (HAP) in 1996 and is on the list of "Substances Requiring Priority Action" published by the Central Environmental Council of Japan (Kida, 2005). The Central Environmental Council prepared the second report "Future direction of measures against hazardous air pollutants" in October 1996, which also proposed that the voluntary action to reduce emissions, as well as an investigation of hazards, atmospheric concentration and pollution sources should be promoted. Although the industrial emissions of mercury in Japan have decreased in recent years (Ito et al., 2006), primarily due to the voluntary reduction of mercury emissions from industrial sources, the concentration distribution of these pollutants in the local atmospheric environment has remained largely unknown (Shirane, 2007).

Mercury is a natural trace component in the environment. Notwithstanding, the bioaccumulation of methylmercury (MeHg) via the food chain, especially through fish, concentrates mercury and poses serious toxicity hazards to the biosphere (Harada, 1995). For that reason, natural and anthropogenic emissions of mercury in the environment (Nriagu and Pacyna, 1988), its transportation and fate (Schroeder and Munthe, 1998; Boening, 2000), and its adverse effects on human health and the ecosystem (Ditri, 1991) have all attracted great attention as facets of a major environmental problem. Stack emissions from coal–combustion power industry includes both vapor and particle–bound phases. Reactive gaseous mercury [RMG or Hg(II)] (Schroeder and Munthe, 1998) can be inorganic (e.g., mercuric chloride, $HgCl_2$) or organic [e.g., methylmercury (MeHg)]. It can also be present as particulate mercury (e.g, mercuric oxide, HgO, or mercury sulfide, HgS). In the global atmosphere, gaseous elemental mercury [GEM or Hg(0)] is the dominant form. Hg(II) typically constitutes a small percentage of total mercury and is predominantly in the gas phase. MeHg concentration in the atmosphere is relatively low, about 10% – 30% lower than total Hg(II) concentrations, according to analysis of precipitation samples (Seigneur et al., 1998). However, Hg(II) becomes methylated in water bodies, where it can bioaccumulate in the food chain. Hg(0) is sparingly soluble in cloud particles and is not removed significantly by wet deposition, and its dry deposition velocity is also believed to be low. As a result, Hg(0) has a long atmospheric lifetime. On the other hand, Hg(II) is quite soluble with cloud particles, so is removed rapidly by wet and dry deposition processes, and has much shorter atmospheric lifetimes (Hedgecock and Pirrone, 2004). Particulate mercury [PM or Hg(p)] is mostly present in the fine fraction of particulate matter ($PM_{2.5}$), although some Hg(p) may be present in coarse PM (Landis and Keeler, 2002).

The concentration of mercury should be estimated both on a regional scale as well as on a local scale, because not only the concentration of mercury in the general environment is important (i.e. the area which includes most of the total population), but also those in the vicinity of industrial sources (i.e. areas of high concentration) should be considered carefully, as particular industrial sources are expected to be associated with relatively high–risk areas. In this study, two different models have been

selected, which were used to assess the extent of exposure: the AIST–ADMER (National Institute of Advanced Science and Technology–Atmospheric Dispersion Model for Exposure and Risk Assessment) estimates regional concentration distribution of hazardous chemical substances (Higashino et al., 2003; Higashino et al., 2004), and the METI–LIS (Ministry of Economy, Trade and Industry–Low–Rise Industrial Source Dispersion Model) estimates the concentration distribution in the vicinity of particular industrial facilities (Kouchi et al., 2004).

Gaseous mercury, including both Hg(0) and Hg(II), were considered as total mercury emissions in the atmosphere, which served as input emission data for these two air pollutant dispersion models. More than 99.5% of mercury in the stack emissions was in the gaseous form (Lindqvist and Rodhe, 1985) and the proportion in particulate form was extremely low in Japan (Yokoyama et al., 2000). Since mercury treatment systems of the coal combustion facilities are very advanced in Japan, Hg(II) emission from the stack is also very low (Takahashi et al., 2008).

The objective of this study was to estimate the concentration of mercury in Japan, whereas the above mentioned two models were used for the assessment of the atmospheric concentration of mercury.

2. Method

2.1. AIST–ADMER model

The AIST–ADMER (Higashino et al., 2003; Higashino et al., 2004) version 1.5e is a series of models and systems designed for estimating the regional atmospheric level of chemicals, developed by the National Institute of Advanced Industrial Science and Technology. The functions of the AIST–ADMER model provide the following calculations and simulations:

- Generation and confirmation of meteorological data
- Generation and confirmation of chemical substance emission data
- Calculation of atmospheric concentrations and deposition of chemicals
- Graphical images of calculation results
- Calculation of resulting histogram
- Population exposure assessment

The purpose of this model is to estimate a long–term, average distribution of chemical concentration in a relatively wide region, such as the Kanto and Kansai areas of Japan. Atmospheric concentration distribution of chemical substances of a 5 km × 5 km square spatial grid for an average of one month to one year can be calculated by this model. Generally, use of models requires preparation of various data, such as meteorological data, creating target substance emission data, and setting calculation param–eters, in order to estimate the atmospheric concentration of chemicals and assess their exposure.

In this study, meteorological input data, calculated monthly for a year, i.e., from January to December 2006, have been used for the AIST–ADMER model calculation. Meteorological input data were produced from AMeDAS (Automated Meteorological Data Acquisition System) (Akasaka and Nimiya, 1986) data, whereas the solar radiation and cloud cover were obtained from individual weather stations.

Simulations calculated from the AIST–ADMER model need information on target substances, such as the amount and geographical location (i.e., latitude, longitude) of emission etc. The AIST–ADMER contains a function for creating the gridded emission data required for the calculation. The methods used for creating gridded emission data can be classified mainly into two types, i.e., point sources, which specify a location using latitude and longitude, and enter the emissions generated from the location, and area sources, which specify emissions for each region or city, and allocate the emissions to calculation grids according to indices such as population, area, industrial statistics, and traffic volume.

The AIST–ADMER model calculation range consists of a number of calculation grids. Total 11 calculation ranges are pre–registered in ADMER in order to cover overall Japanese region (Table 1). Before starting the simulation, it is recommended to select a calculation range that includes target ranges.

***Table 1**. ADMER Calculation range*

Name	Range	Number of grids	Regions
Hokkaido	E 139° 15' 00" – 145° 56' 15" N 41° 17' 30" – 45° 35' 00"	107 × 103	Hokkaido
Tohoku	E 139° 07' 30" – 142° 11' 15" N 36° 45' 00" – 41° 37' 30"	49 × 117	Aomori, Iwate, Miyagi, Akita, Yamagata, Fukushima
Hokuriku	E 136° 07' 30" – 139° 56' 15" N 36° 02' 30" – 38° 35' 00"	61 × 61	Niigata, Toyama, Ishikawa
Kanto	E 138° 18' 45" – 140° 56' 15" N 34° 50' 00" – 37° 12' 30"	42 × 57	Ibaraki, Tochigi, Gunma, Saitama, Chiba, Tokyo, Kanagawa
Chubu	E 135° 22' 30" – 139° 11' 15" N 35° 07' 30" – 37° 05' 00"	61 × 47	Fukui, Yamanashi, Nagano, Gifu
Tokai	E 135° 48' 45" – 139° 15' 00" N 33° 40' 00" – 35° 40' 00"	55 × 48	Shizuoka, Aichi, Mie
Kinki	E 134° 11' 15" – 136° 30' 00" N 33° 22' 30" – 35° 50' 00"	37 × 59	Shiga, Kyoto, Osaka, Hyogo, Nara, Wakayama
Chugoku	E 130° 41' 15" – 134° 33' 45" N 33° 40' 00" – 35° 40' 00"	62 × 48	Tottori, Shimane, Okayama, Hiroshima, Yamaguchi
Shikoku	E 131° 56' 15" – 134° 56' 15" N 32° 37' 30" – 34° 37' 30"	48 × 48	Tokushima, Kagawa, Ehime, Kochi
Kyushu	E 128° 15' 00" – 132° 11' 15" N 30° 55' 00" – 34° 17' 30"	63 × 81	Fukuoka, Saga, Nagasaki, Kumamoto, Oita, Miyazaki,
Okinawa	E 122° 48' 45" – 131° 26' 15" N 24° 00' 00" – 27° 57' 30"	138 × 95	Okinawa

2.2. METI–LIS model

The METI–LIS (Kouchi et al., 2004) is a user–friendly computer model developed originally by Ministry of Economy, Trade and Industry (METI). The METI–LIS model version 2 is now available in English to download from the online site (Kouchi et al., 2004). This model puts special importance to express downdraft effect, which often affects the atmospheric dispersion from lower emission sources, while it gives solutions of simple Gaussian plume and puff formula (Bosanquet and Pearson, 1936; Sutton, 1947; Turner, 1994; Beychok, 2005) for elevated sources. In addition to a short–term estimation with fixed meteorological conditions, a long–term average estimation can be obtained with the model, when hourly meteorological datasets are prepared by the users.

Equation (1) (Sutton, 1932; Sutton, 1947) is used in the METI–LIS model for the transport of pollutants from a point source, such as a smokestack or exhaust outlet. This section deals with the plume–rise height of exhaust gas, methods of determining dispersion parameters, methods of modeling down–wash effects caused by buildings neighboring the emission source, and the applicable conditions of the dispersion model.

For each source and every hour, the origin of the coordinate system calculation is placed on the ground surface at the base of the stack. The *x*–axis is positive in the downwind direction, the *y*–axis is crosswind (normal) to the *x*–axis, and the *z*–axis extends vertically. The user–defined calculation points are converted to each source's coordinate system for the calculation of concentration at each time period. The conversion method in the *x*–axis and *y*–axis direction is described below. The concentration calculated for each source at each calculation point is summed to obtain the total concentration produced by the combined source emissions for that time period (Bosanquet and Pearson, 1936; Sutton, 1947; Turner, 1994; Beychok, 2005).

$$C(x,y,z) = \frac{QV}{2\pi u_s \sigma_y \sigma_z} \times exp\left[-0.5\left(\frac{y}{\sigma_y}\right)^2\right] \quad (1)$$

where C is the concentration of pollutants (g/m³), at any receptor located, x is the downwind distance from the emission source (m), y is the crosswind distance from the emission plume centerline (m), z is the distance above the ground level (m), Q is the pollutant emission rate (g/s), V is the vertical term [Equation (5)], u_s is the horizontal wind velocity along the plume centerline (m/s), σ_z is the dispersion parameter in vertical direction (m), and σ_y is the dispersion parameter in horizontal direction (m).

Table 2. *Atmospheric stability categories*

Wind speed at ground level U (m/s)	Daytime Solar radiation Q (0.01 kW/m²)				Nighttime (Solar radiation Q < 0)
	60 < Q	30 ~ 59	15 ~ 29	1 ~ 14	
U < 2.0	A	A–B	B	D_d	F
2.0–2.9	A–B	B	C	D_d	E
3.0–3.9	B	B–C	C	D_d	D_n
4.0–5.9	C	C– D_d	D_d	D_d	D_n
6.0 < U	C	D_d	D_d	D_d	D_n

Equations (2), (3), and (4) fit the Pasquill–Gifford curves (Venkatram, 1996) approximately, which are used in the METI–LIS model to calculate the dispersion parameters (σ_y and σ_z). The same equations are also used in the ISC (Industrial Source Complex) model (Bowers and Anderson, 1981; Bowers et al., 1982). ISC is a popular steady–state Gaussian plume model and can be used to assess pollutant concentrations from a variety of sources associated with industrial complexes. The approximation equations are the functions of downwind distance from the source and they calculate the lateral dispersion width, σ_y and the vertical one σ_z of Equation (1), respectively. These dispersion widths are contingent on atmospheric stability (Pasquill, 1961), which is determined by meteorological conditions. Table 2 shows the classification method for the atmospheric stability.

The atmospheric stability category can be selected from Table 2 using the data of wind speed and solar radiation in the area of emission sources (Luna and Church, 1972). While 11 different categories (i.e., A–G) can be accepted as the atmospheric stability, those in the approximation in Table 3 are divided into only six categories (i.e., A–F). Table 3 shows the atmospheric stability category mapping between the observed and the approximation situation with the standard values of the power exponent (p). In Table 3, the atmospheric stabilities, A and A–B are unified to A stability, B and B–C are unified to B stability, C and C–D_d are unified to C stability, D_d and D_n are unified to D stability, F and G are unified to F stability. Among the stability categories, (A, A–B, B, B–C, C, C–D_d, D_d) are the daytime and (D_n, F and G) are the nighttime stability categories. The value of the wind profile exponent (p) is used in Equation (6), which can be obtained from Table 3.

The dispersion parameters σ_y and σ_z are used in Equation (1), which can be obtained from Pasquilll–Gifford curves [Equations (2), (3), and (4)]. The values for dispersion coefficients (a, b, c, and d) (Turner, 1967; Turner, 1994) are available in the online technical manual of the METI–LIS model.

$$\sigma_y = 465.11628(X)\tan(TH) \quad (2)$$

where x is the downwind distance (m).

$$TH = 0.017453293[c - d(\ln(x))] \quad (3)$$

$$\sigma_z = ax^b \quad (4)$$

where a, b, c, and d are the dispersion coefficients.

The vertical term, V in Equation (5) (Sutton, 1932; Sutton, 1947) represents the atmospheric distribution of the Gaussian plume in the vertical direction. This term includes the elevation of calculation point and the effects of the height caused by the emitted plume rise (the effective plume–rise height) (Bosanquet and Pearson, 1936; Sutton, 1947; Turner, 1994; Beychok, 2005). Most of the time, the gases that are emitted from the stacks of a power plant are heated and are warmer than the outdoor air. Emitted gases are less dense than the outside air and therefore they are buoyant. A combination of the gas momentum and buoyancy causes the gases to rise. This is referred to as plume rise and allows air pollutants emitted in this stack gas stream to be lifted higher in the atmosphere.

Table 3. *Relationship between the observed atmospheric stability and the approximation index. This approximation simplified 11 different atmospheric stability categories into only six categories (A to F). The standard values of the power exponent p are used in the METI–LIS model to adjust wind–speed, which is similar to the ISC model (Bowers and Anderson, 1981; Bowers and Anderson, 1982) applies these values to rural areas*

Atmospheric Stability	A	A–B	B	B–C	C	C– D_d	D_d	D_n	E	F	G
Approximation	A		B		C		D		E	F	
Rural exponent (p)	0.07		0.07		0.10		0.15		0.35	0.55	

$$V = exp\left[-0.5\left(\frac{Z_r - h_e}{\sigma_z}\right)^2\right] + exp\left[-0.5\left(\frac{Z_r + h_e}{\sigma_z}\right)^2\right] \quad (5)$$

where z_r is the elevation of calculation points of any receptor, which is located z meters above ground level (m), h_e is the effective plume–rise height (m), which is the sum of the physical stack height and the plume rise.

The wind profile power law (Peterson and Hennessey, 1978; Elliott et al., 1986; Robeson and Shein, 1997; Beychok, 2005) [Equation (6)] is a relationship between the wind speeds at one height, and those in another, which converts the observed wind speed to an equivalent wind speed at the actual height of the emission source. The wind speed used in the dispersion equation is the equivalent wind speed at the stack, or release height. If the height of measurement point of the wind speed is lower than the stack height, the power law equation [Equation (6)] will be applied in MET–LIS model. The power law equation is in the form of:

$$u_2 = u_1\left(\frac{z_2}{z_1}\right)^p \quad (6)$$

where u_2 is the wind speed at the stack outlet height (m/s), u_1 is the wind speed at the measurement height (m/s), z_2 is the stack outlet height (m), and z_1 is the wind–speed measurement height (m).

The wind profile exponent, p in Equation (6) is set according to the atmospheric stability. The values shown in Table 3 can be used as average values.

When the point source is a stack, it acts as a drag to the wind, producing a down–wash known as stack–tip down–wash. When the exit velocity of the exhaust gas from the source is less than 1.5 times the velocity of the horizontal wind along the plume centerline, a correction is applied to the stack height corresponding to stack–tip down–wash by using Equation (7). This method adjusts the physical height of the stack as follows:

$$h'_s = h_s + 2d_s\left(\frac{v_s}{u'_s} - 1.5\right) \quad (7)$$

where h'_s is the modified physical stack height (m), h_s is the physical stack height (m), d_s is the stack diameter (m), v_s is the exit velocity of the exhaust gas (m/s), and u'_s is the velocity of the horizontal wind along the plume centerline (m/s).

The Equation (8) is applicable when $v_s \geq 1.5\ u'_s$.

$$h'_s = h_s \quad (8)$$

This modification is not applied when down–wash effects due to a building are calculated.

The METI–LIS model also emphasized on famous "Briggs equations" to calculate buoyancy–induced plume rise due to hot buoyant plumes of bent–over. In general, plume rise equations for bent–over, hot buoyant plumes are based on observations and data involving plumes from typical combustion sources such as the flue–gas stacks from steam–generating boilers burning fossil fuels in large power plants. Therefore, most of the coal combustion power plants in Japan, the stack exit velocities are about 30 m/s and the exit temperatures are about 90°C (Ito et al., 2006). If the gas emitted by the source is comparatively warmer than the ambient temperature, the CONCAWE equation (Briggs, 1965; Briggs, 1968) is applied as follows:

$$h_e = h_s + \Delta h \quad (9)$$

$$\Delta h = 0.175 Q_H^{1/2} u^{-3/4} \quad (10)$$

where h_e is the effective stack height (m), h_s is the physical stack height (m), Δh is the buoyancy–induced plume rise (m), Q_H is the emitted heat quantity (cal/s), and u is the wind speed at top of stack (m).

$$Q_H = \rho C_p Q(T_s - T_A) \quad (11)$$

where ρ is the gas density at 0°C (1.293×10^3 g/m^3), C_P is the isobaric specific heat (0.24 cal/K/g), Q is the exhaust–gas flowrate (Nm^3/s), T_S is the exhaust–gas temperature (°C), and T_A is the ambient temperature (°C, default is 15°C).

This model also includes building downwash, terrain effects, and line source emissions. The METI–LIS model adopted a downwash scheme based on that of the US Environmental Protection Agency's (EPA) Industrial Source Complex (ISC) model, but the parameters in the dispersion widths describing the down–wash effect were improved by incorporating the results of wind tunnel experiments. Another characteristic point of the METI–LIS model different from the ISC model is that the evaluation time which affects the dispersion width especially in the y (crosswind) direction can be adjusted for a simulation of short time dispersion.

3. Atmospheric Mercury Emissions in Japan

3.1. Mercury emission sources

Of the primary anthropogenic sources of mercury emissions to the environment, the principal sources are those where mercury is emitted mainly as an unintentional byproduct. With the exception of mercury mining itself, the atmospheric mercury emissions arise from the mercury that is present as an impurity in the fuel or used raw materials. The main emissions of mercury as byproducts are from the sectors that involve combustion of coal or oil, production of pig iron and steel, production of non–ferrous metals, and cement production. Stationary combustion of coal, and the combustion of other fossil fuels associated with energy or heat production in major power plants, small industrial or residential heating appliances, and various industrial processes are the largest single source category of anthropogenic mercury emissions to the global atmosphere. Although coal does not contain high concentration of mercury, the amount of mercury emissions to the atmosphere from coal–fired industrial facilities indicates that coal burning is one of the largest anthropogenic sources of unintentional mercury emissions to the atmosphere. Mining and industrial processing of ores, particularly in primary production of iron and steel, and non–ferrous metal production (specially copper, lead and zinc smelting) release mercury to the atmosphere due to fuel combustion, the presence of mercury in ores as impurities, and through accelerating the exposure of rock to natural weathering process. Metal production including mining, the production of mercury itself (a relatively minor source) and the production of gold, where mercury is present in ores and used in some industrial processes, are the minor sources of mercury emissions to the atmospheric environment. Meanwhile, one of the major sources of by–product releases of mercury is associated with cement production, where mercury is released primarily as a result of the combustion of fuels (mainly coal but also a range of wastes) to heat the cement kilns (AMAP and UNEP, 2008).

According to recent research reports (Kida et al., 2007; Moritomi, 2008) and emission data provided by the Japan Ministry of Economy, Trade and Industry (METI, 2001–2004), the total amount of mercury released to the atmosphere from Japan was estimated as 24 – 28 Mg/year, taking into account the releases from specified facilities not reported by PRTR (Pollutant Release

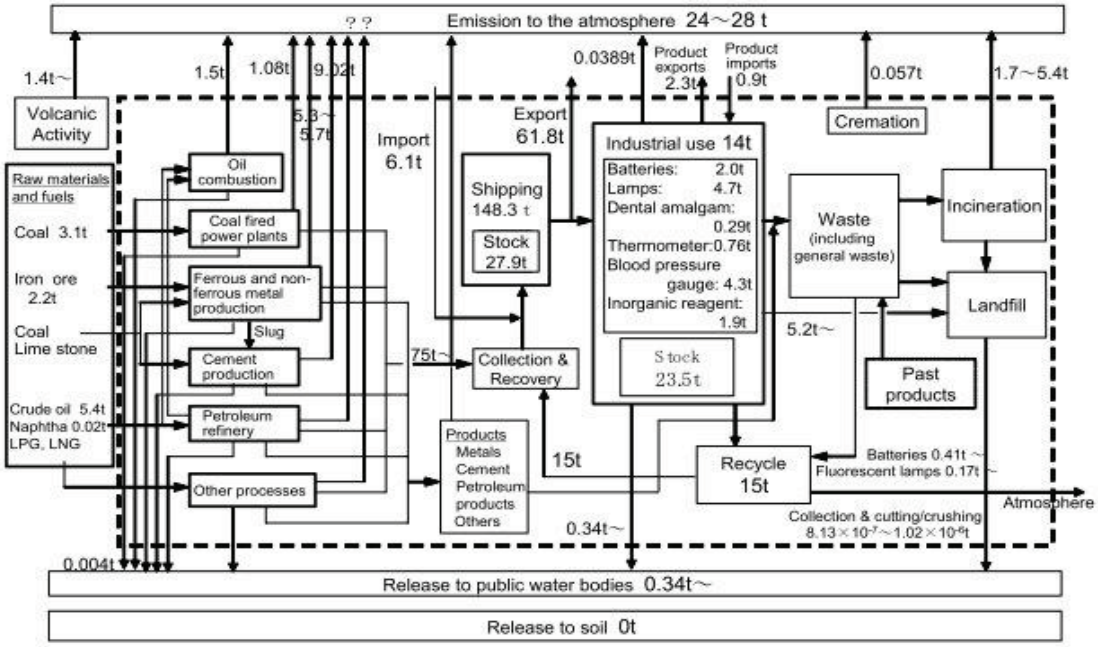

Figure 1. *Material flow diagram of mercury in Japan (Kida et al., 2007; Moritomi, 2008).*

and Transfer Register) (Lerche et al., 2004; Wexler and Harjula, 2005). In the combustion category, coal–fired power plants, industrial oil combustion boilers, incinerators of medical waste, sewage sludge and other wastes are considered to be significant mercury emission sources. Among the heavy industrial production units, primary ferrous and non–ferrous metal production as well as cement production are thought to be major contributors of atmospheric mercury emissions in Japan. Atmospheric mercury emissions in Japan are calculated to be 0.190–0.225 g/year/person (METI, 2001–2004; Kida et al., 2007; Moritomi, 2008). Figure 1 shows the material flow of mercury, which depicts the net mercury load to the atmosphere in Japan from primary anthropogenic sources.

3.2. Estimation of mercury emissions

In this study, the amount of atmospheric mercury emissions from different point sources and area sources in Japan were estimated according to the report on the mercury emissions inventory of Japan (Kida et al., 2007; Moritomi, 2008). Total coal consumption data for 2005 was considered as a calculation basis to estimate the mercury emissions to the atmosphere in Japan, whereas the emission of atmospheric mercury in 2006 is almost similar to that of 2005. The power plant of electrical capacity 1 000 MW consumes 360 Mg/hour coal and the mean concentration of mercury in that coal was 0.045 ppm (Ito et al., 2006). Since about 30% of the total mercury of feed coal goes to the atmosphere from the stack of the coal combustion power industries (Moritomi, 2008) in Japan, the mean emission rate of mercury to the atmosphere was 4.4 µg/KW h. The power plant of electric capacity 1 000 MW emits mercury to the atmosphere is (360 Mg coal/hour) × (0.045 g Hg/Mg coal) × (365×24 hour/year) × 0.3 = 42 600 g/year (42.6 kg/year). The coal combustion rate has been used as a basis to calculate the amount of mercury emissions to the atmosphere from coal combustion power industries in Japan. The emission factors were derived from estimates of the annual emission rate and the total production capacity for each plant in 2006. Total productions are 69.5×10^6 Mg and total emissions of mercury to atmosphere are 5.7 Mg in the industrial sector of iron works, total productions are 79×10^6 Mg and total atmospheric mercury emissions are about 9.8 Mg in the industrial sector of cement plants, total productions are 9 057 Mg and total atmospheric mercury emissions are 0.3 Mg in the industrial sector of chemical plants in 2006 in Japan (JCOAL, 2005; Kida et al., 2007). A simple unitary calculation method was applied to calculate atmospheric mercury emissions from each point source of iron works, cement plants, chemical complexes. The data of yearly production capacity and yearly mercury emissions for each industrial sector were considered as the basis of calculation (METI, 2001–2004; JCOAL, 2005; Kida et al., 2007; Moritomi, 2008). For example, mercury emissions from a specific cement industry = {(total mercury emissions from cement industries in Japan) × (production capacity of that industry)} ÷ total cement production capacity in Japan. The geographical locations of mercury emission sources from coal–fired industrial facilities can be easily pointed out on the map from the website of Japan Coal Energy Center (JCOAL, 2005). In this study, yearly municipal and medical waste has also been considered as a big source of atmospheric mercury emissions in Japan. Total atmospheric mercury emissions from municipal and medical waste are about 1.7–5.4 Mg (Kida et al., 2007) in Japan, that have been distributed to the local atmosphere of each region on the basis of the prefectural population density (METI, 2001–2004; Kida et al., 2007; Moritomi, 2008).

Burning of fossil fuels (primarily coal) is the largest single anthropogenic source of atmospheric mercury emissions, although the emissions from combustion of medical and municipal waste, and industrial waste have a significant release of mercury to the atmosphere in Japan. It is very difficult to find out the actual locations and amounts of mercury emissions in Japan from industrial point sources, since the lack of reliable information on industrial emission assumptions and technologies to calculate mercury emissions as well as confidentiality. In this study, the coal–fired industrial facilities such as power plants, iron works, cement plants, chemical complexes, and oil or gas combustion heavy industries are considered as large emission sources of atmospheric mercury in Japan. Mercury emissions from municipal and medical waste from different areas in Japan have also taken into consideration for the AIST–ADMER model as important area sources of mercury emissions. To calculate the regional atmospheric concentration distribution of mercury, about 28 Mg/year (Kida et al., 2007; Moritomi, 2008) of mercury emissions to the atmosphere have been distributed hypothetically throughout Japan.

4. Study Area

In this study, the industrial source complexes are considered as mercury emission sources, which are located in the central region of the Honshu island of Japan. Total nine regions (Aichi, Mie, Gifu, Fukui, Ishikawa, Niigata, Nagano, Gunma and Toyama) have been selected for the AIST–ADMER model simulation to calculate the regional distribution of mercury concentration. There are different types of heavy and medium–scale industrial facilities located in this area that are in operation.

On the other hand, a small domain (the blue rectangle in Figure 3) in Aichi Prefecture of Japan was selected as a site to calculate the ambient air concentration of mercury in the vicinity of two major industrial sources using the METI–LIS model. Among the two hypothetical power plants inside of the small domain, the plant–1 (the solid black circle in Figures 4, 5, 6) is located in Nagoya area (35° 1' 39.38" N, 136° 51' 54.98" E) and plant–2 (the solid red circle in Figure 4, 5, 6) is located in the Hekinan area (34° 50' 6.80" N, 136° 57' 44.75" E). The two sources are about 20 km apart from each other, and they are located in the coastal area of Japan.

5. Results

5.1. Regional concentration level

The input emission data for the AIST–ADMER model was compiled from the survey results of the Pollutant Release and Transfer Register (PRTR) of 2005. Moreover, mercury emission inventory work, prepared by Kida et al. (2007) and Japan Coal Energy Center (JCOAL, 2005) was applied as an input data for the AIST–ADMER model. Table 4 shows the input parameters for the AIST–ADMER model.

The mercury concentrations in the small domain (blue rectangle) of the Figure 3 served as a background concentration for the METI–LIS model to determine the mercury concentration in the vicinity of two nearly located hypothetical power plants. Since the maximum ambient concentration of mercury inside of the blue rectangle of the Figure 3 was 2.934 ng/m^3 (0.002934 μg/m^3) and minimum concentration was 0.66 ng/m^3 (0.00066 μg/m^3), the background concentration was determined to be (0.66 + 2.934)/2 = 1.797 ng/m^3 = 1.8 ng/m^3 (0.0018 μg/m^3). The values of the background mercury concentrations in different areas are obtained from the Figure 3, which can be compared with the monitoring survey data of hazardous air pollutants in 2006 prepared by the Japan's Ministry of the Environment (MOE, 1997; MOE, 1998–2009). The yearly average mercury concentration data of 12 air quality monitoring stations in Aichi and Mie regions, provided by Japan's Ministry of the Environment (MOE) along with the AIST–ADMER simulation result, are illustrated in Table 5. The magnitudes of mercury concentration calculated by the AIST–ADMER model are slightly overestimated relative to the observed results of 7 monitoring stations, while they are underestimated at 5 monitoring stations, suggesting that the industrial emissions or emission from biomass burning at underestimated monitoring sites were not significantly considered in this study and this needs to be improved. The distribution of the air quality monitoring stations for mercury and the characteristics of each site can refer to the presentation of Suzuki (2008) in Vietnam. Figure 2 shows the geographical locations of 12 monitoring stations. The two hypothetical coal–fired power plants that have been considered for the METI–LIS simulation are located inside of the small domain (blue rectangle) of Figure 2.

Table 5 shows a comparative evaluation of annual mean mercury concentration in 12 monitoring stations with the simulation results of the AIST–ADMER model (Figure 3). Since, most of the monitoring stations are located far away from the industrial point sources in Japan the simulation result of the METI–LIS model cannot be compared with the monitoring data in Table 5.

Table 4. *Input parameters for the AIST–ADMER model*

Start of calculation	January 2006
End of calculation	December 2006
Washout ratio	1
Half life (days)	365
Emission pattern	Yearly average emission

Figure 2. *Map of observation sites in Aichi and Mie regions and two industrial point sources in Aichi region. The red solid circle on the map shows the location of 12 monitoring stations. The blue solid circle (34° 50' 6.80" N, 136° 57' 44.75" E) inside the rectangle shows a point source location of mercury emission and the black solid circle (35° 1' 39.38" N, 136° 51' 54.98" E) shows another point source of mercury emission.*

Table 5. *Monitoring data of mercury concentration in 2006 provided by Ministry of Environment in Japan (MOE, 1997–2004; MOE, 1997)*

Name of the region	Number	Name of the monitoring station	Geographical location	One–year mean concentration (ng/m^3)		Observed concentration range (ng/m^3)
				The observed mean value	The AIST–ADMER simulation mean	
Aichi	1	Nagoya City (Chikusaku)	E 136° 56' 52" N 35° 9' 57"	0.73	1.63	0.20–1.7
	2	Nagoya City (Nakagawaku)	E 136° 51' 17" N 35° 8' 30"	2.1	2.32	1.8–2.4
	3	Toyohashi City (Oosaki)	E 137° 20' 36" N 34° 42' 58"	2.5	1.92	1.6–4.6
	4	Toyohashi City (Futagawa)	E 137° 26' 20" N 34° 43' 32"	1.8	0.21	0.98–2.5
	5	Okazaki City	E 137° 11' 15" N 34° 55' 51"	2.5	2.08	0.51–3.9
	6	Toyota City (Central)	E 137° 4' 44" N 35° 1' 59"	2.0	2.24	1.4–2.8
	7	Toyota City (North)	E 137° 3' 29" N 35° 1' 0"	1.7	2.06	0.81–2.6
	8	Komaki City	E 136° 55' 6" N 35° 17' 35"	1.8	1.43	1.0–2.3
Mie	9	Yokkaichi (North)	E 136° 38' 29" N 35° 0' 30"	2.4	2.65	1.9–3.2
	10	Yokkaichi City (Center)	E 136° 37' 28" N 34° 57' 59"	1.9	2.48	1.2–2.9
	11	Matsusaka City	E 136° 32' 25" N 34° 33' 43"	2.2	1.76	1.6–2.7
	12	Kuwana City	E 136° 41' 6" N 35° 3' 43"	2.1	2.34	1.5–2.5

Figure 3 shows the annual mean distribution of atmospheric mercury concentrations in the central Honshu island of Japan calculated by the AIST–ADMER model. The results established that the atmospheric mercury concentration was relatively high in major urban areas such as Nagoya and Yokkaichi, as emissions from industrial facilities, medical and municipal wastes tend to be concentrated in these densely populated areas. The annual mean concentration of atmospheric mercury was calculated to be less than 2.934 ng/m^3 (0.002934 μg/m^3) in major industrial areas, greater than 0.0263 ng/m^3 (0.000263 μg/m^3) in nonindustrial areas, which was calculated by the AIST–ADMER model in this study. The AIST–ADMER is a regional dispersion model, which can calculate wide–area chemical transportation considering several point sources, line sources and area sources in Japan. The simulation result shows that the mercury concentration calculated by the AIST–ADMER mode was diluted and always less than that of the result of the METI–LIS model, because of its regional scale chemical transportation scheme to calculate the atmospheric concentration of chemical substances. In some cases, concentrations were calculated to be 5 ng/m^3 (0.005 μg/m^3) – 10 ng/m^3 (0.01 μg/m^3) in the vicinity of major industrial point sources simulated by the METI–LIS model. Most of the cases, the concentration of mercury calculated by the METI–LIS model is slightly higher than observations, because METI–LIS generally calculate the pollutant concentration in the vicinity of industrial point sources for a small domain.

5.2. Concentration level near industrial sources

The ambient concentration of mercury in the vicinity of two major industrial sources was predicted by the METI–LIS model. Mercury releases to the atmosphere from these two hypothetical coal–fired power plants were calculated on the basis of mercury emission factor (Kida et al., 2007; Moritomi, 2008; METI, 2011). The selected site for the METI–LIS simulation had a calculation domain of 25 km × 25 km with a grid spacing of 200 m, which included the two largest point sources corresponding to the 3 km × 3 km calculation grids of the AIST–ADMER model. There are 5 units in power plant–1 and 6 units in power plant–2, whereas the capacity of each unit is 1 000 MW. Power plant–1 with 5 000 MW electric capacity is emitting 213 kg mercury/year into the local atmosphere. Similarly, the capacity of power plant–2 is 6 000 MW and it is emitting 256 kg/year mercury to the atmosphere. The amount of mercury emissions to the atmosphere from plant–1 and plant–2 have been considered as input data for the METI–LIS model. These two large coal combustion facilities in this area are the significant sources of mercury emissions to the atmosphere in Japan, emitting about 1.7% of mercury into the air every year (Kida et al., 2007). To evaluate the effect of mercury emissions from the two hypothetical coal–fired power plants in the local atmosphere, stack gas dispersion was calculated by the METI–LIS model. Table 6 shows the operational conditions of these two power plants. The specifications of each power plant provided in Table 6 are also very important input data for the METI–LIS model. Since there was no emission of Hg(p) from the coal combustion power plants in Japan, the effect of gravitational sedimentation, and the amounts of dry and wet depositions were not considered in this study. Necessary assumptions for mercury emissions and the specifications of each point source were determined from the report of Japan Coal Energy Center for 2005 (JCOAL, 2005) and the research work by Ito et al. (2006). It was assumed that the emission factors were constant for 365 days, 24 hours a day. AMeDAS (JMA, 2006) data were used as meteorological input data for the METI–LIS model. Source contributions from other sources (e.g., mobile sources or point sources located outside of the calculation domain) were not included in the input data for the METI–LIS model. Source contributions from other sources were calculated with the AIST–ADMER model and were superposed onto the simulation results of the METI–LIS model as the background concentration data of mercury in the atmosphere.

Figures 4 – 6 show the distribution of mercury concentrations in the vicinity of the two hypothetical power plants in winter, summer, and one–year average (2006), which were calculated using the METI–LIS model. The mark of the solid black circle (latitude 35° 1' 39.38" N, and longitude 136° 51' 54.98" E) with

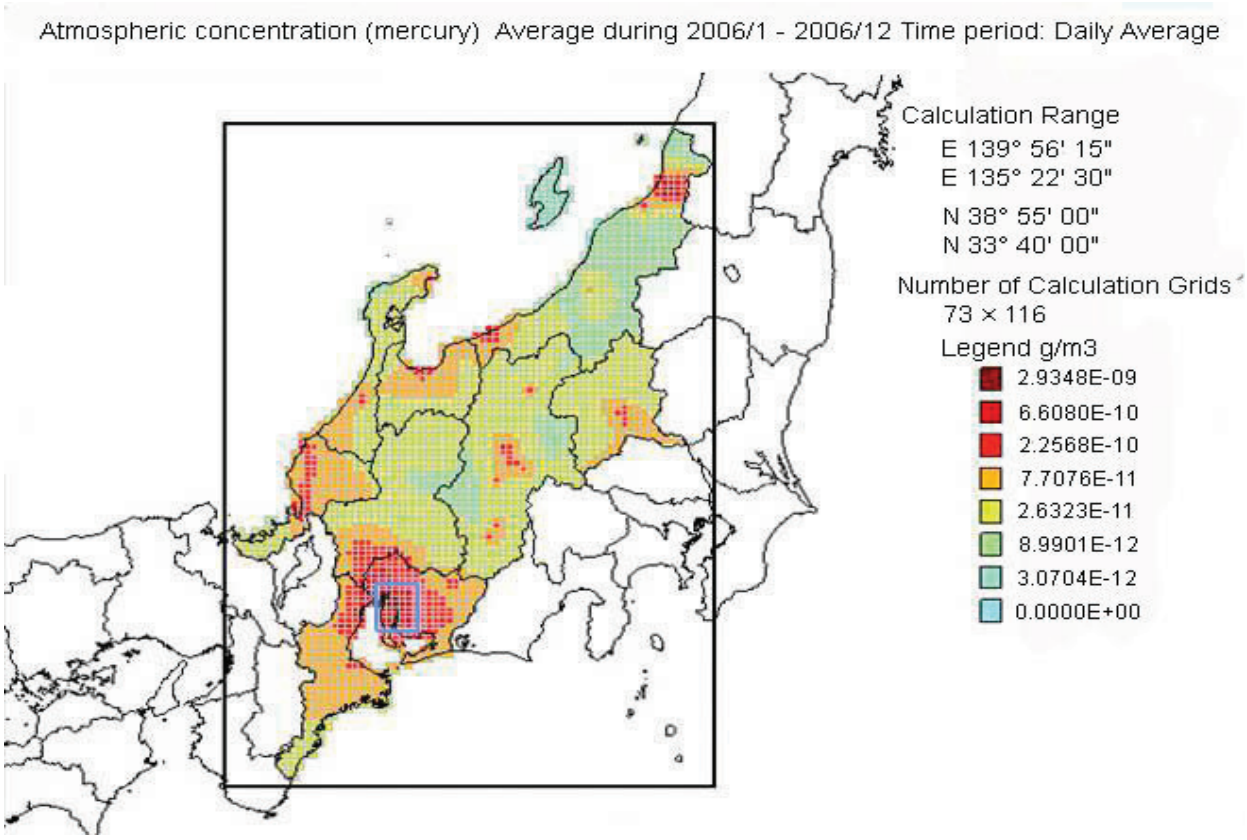

Figure 3. *The annual mean concentration distribution of atmospheric mercury calculated with the AIST–ADMER model in 2006. Nine areas (Aichi, Mie, Gifu, Fukui, Ishikawa, Niigata, Nagano, Gunma, and Toyama) are designated on the map. The blue rectangle was used for the METI–LIS model as a local domain, which served as background concentration data in the calculation areas for the METI–LIS model.*

Table 6. *Specifications of hypothetical coal–fired power stations (Ito et al., 2006). The plant–1 has five units and the Plant–2 has six units. The production capacity of each unit is 1 000 MW*

Operation condition	**Plant–1 (35° 1' 39.38" N, 136° 51' 54.98" E)**	**Plant–2 (34° 50' 6.80" N, 136° 57' 44.75" E)**
Output	1 000 MW × 5	1 000 MW × 6
Coal consumption	360 Mg/h × 5	360 Mg/h × 6
Height of stack	200 m	200 m
Stack gas temperature	90 °C	90 °C
Discharge velocity	30 m/s	30 m/s
Volume flow rate (wet)	3 400 000 Nm^3/h × 5	3 400 000 Nm^3/h × 6
Availability factor (annual)	100% (365 days/year)	100% (365 days/year)

0.213 Mg/year mercury emissions and the mark of the solid red circle (latitude 34° 50' 6.80" N, and longitude 136° 57' 44.75" E) with 0.256 Mg/year emission represents the industrial source location on the Figures 4–6. In winter, the mean distribution of mercury concentrations were calculated to range between 0.0068 and 0.0118 μg/m^3 near industrial sources. On the other hand, mean distribution of mercury concentrations ranged between 0.0028–0.0068 μg/m^3 in the same locations in summer, that were much lower than those of winter due to the effect of the boundary–layer meteorological conditions in coastal areas of Japan. In coastal regions, sea and land breezes can be important factors affecting the wind speed and direction. During the summer, the temperature difference between the sea surface and the land surface is much greater than that during the winter (Steve, 1995; JetStream, 2008). During the summer, the effect of the sea and land breeze causes a strong wind flow in the coastal ground level, driving the pollutants far away from their sources. As a result, the concentration of mercury is relatively low in the summer near the industrial point sources. Besides, the thermal circulations of wind in winter are very low, which causes a higher concentration of mercury in the vicinity of the industrial sources.

The annual concentration distribution of mercury calculated by the METI–LIS model establishes that some people living in certain areas near industrial point sources were exposed to a little higher concentration of mercury compared to general population but the levels of mercury meets the air quality standard of Japan's Ministry of Environment. Figure 7 shows the annual wind rose plot, which gives a succinct view of how wind speed and direction are typically distributed at the location between the two point sources in 2006. The annual mean concentration was estimated not to exceed 0.04 μg/m^3 near an industrial source (Kida, 2005), whereas a similar concentration level was found in different seasons.

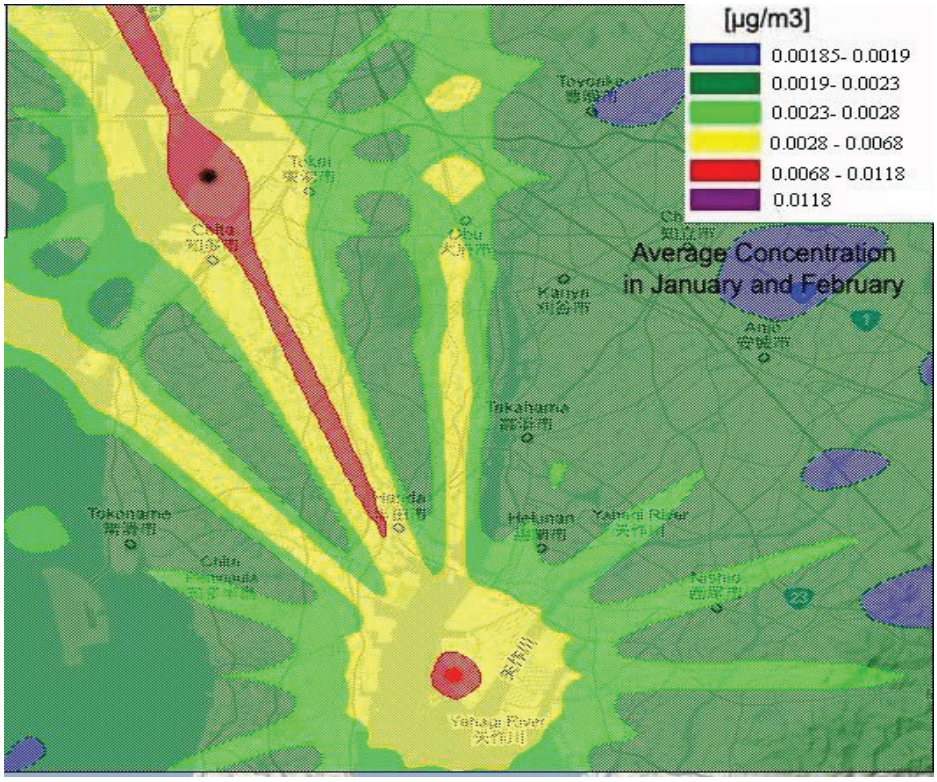

Figure 4. *The average concentration distribution of mercury vicinity of two large point sources, calculated with the METI–LIS model in January and February, 2006. The mark of the solid black circle and red circle represents the industrial source locations in the figure.*

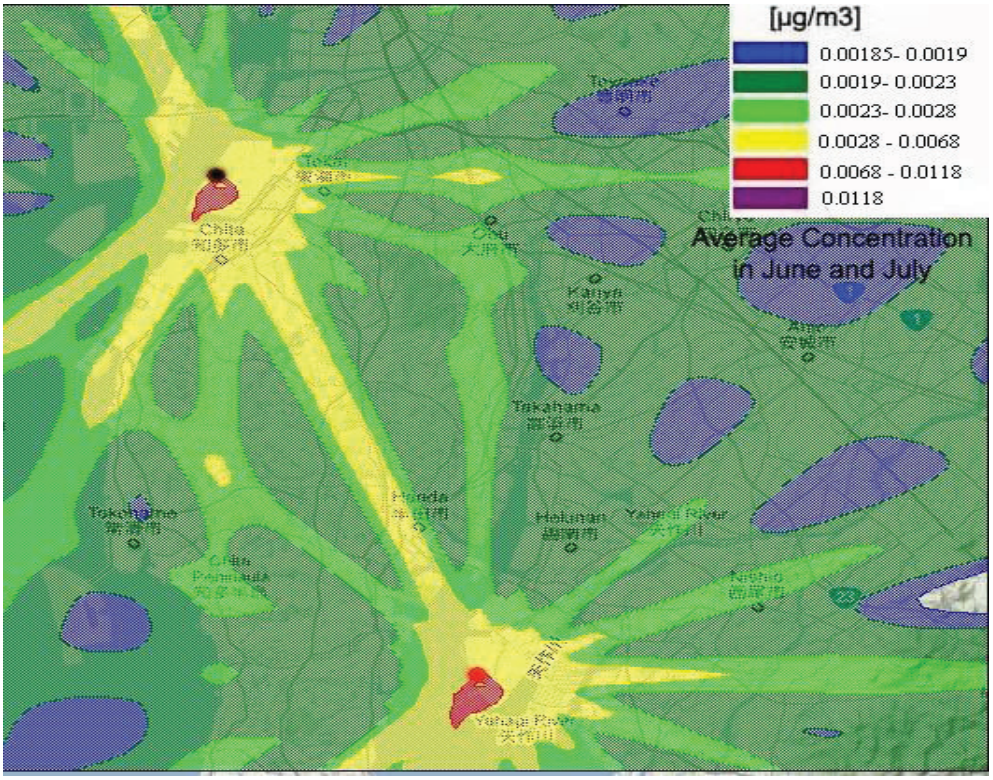

Figure 5. *The average concentration distribution of mercury vicinity of two large point sources, calculated with the METI–LIS model in June and July, 2006. The mark of the solid black circle and red circle represents the industrial source locations in the figure.*

6. Conclusions

In Japan, mercury was categorized as a hazardous air pollutant (HAP) in 1996 due to its high carcinogenic potential. The national government initiated a number of programs to establish emission evaluations and assessments of ambient concentrations. Reduction efforts of mercury emissions has been started on a community basis under public (local governments) and private partnership of industries in Japan which ongoing since 2005 under the support of a voluntary reduction program for emissions. In 2003, Japan initiated the PRTR system, such that the emission data regarding mercury from various sources could be made available in near

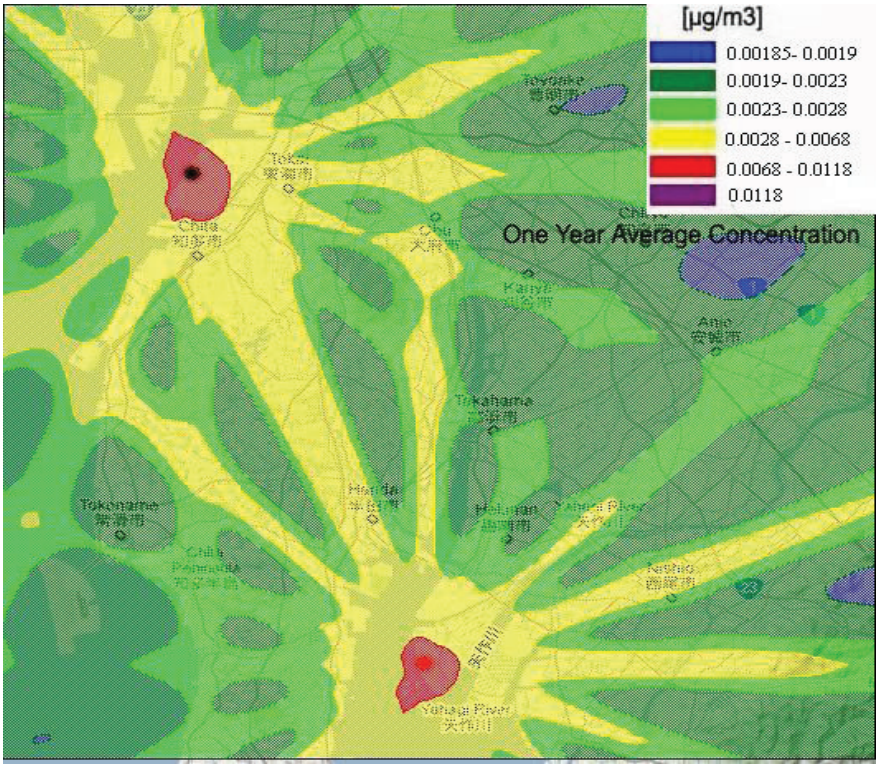

Figure 6. *One–year average concentration distribution of mercury vicinity of two large point sources, calculated with the METI–LIS model in 2006. The mark of the solid black circle and red circle represents the industrial source locations in the figure.*

future. However, the precise amounts remain somewhat uncertain due to ambiguities in the estimation methodologies employed to evaluate mobile sources. The main sources of mercury emissions to atmosphere in Japan are coal–fired cement plants, accounting for over 30% of the total emissions in the year 2006. On the other hand, industrial emissions from primary ferrous metal production and coal–fired power plants had a significant contribution of atmospheric mercury emissions in Japan in 2006. The assessment of mercury concentrations in the local atmosphere in Japan was performed using two different atmospheric dispersion models, i.e., the AIST–ADMER and the METI–LIS. The results of the present study indicated that the annual mean ambient concentrations of mercury in residential areas generally amounted to be less than 0.22 ng/m^3 (0.00022 µg/m^3), but there are no sites that exceed 0.04 µg/m^3 near industrial point sources. Though it is unrealistic to expect the Gaussian models to predict the real situation of mercury concentration in the local atmosphere, the major purposes of the present assessment was to conduct a methodology of comprehensive analysis of exposure and atmospheric distribution of mercury concentration, and thereby to develop a detailed picture of current air quality assessment of the different industrial areas of Japan.

In the present study, small–scale and medium–scale dispersion models for the different regions in the coastal area of Japan were used. The results show a reasonable agreement with the monitoring data with respect to predicting local atmospheric concentrations of mercury. Although there are many models have been dedicated to the modeling of mercury transport in the atmosphere of global and regional scales in the last decades, not many studies have been conducted to investigate the transport pathway of mercury from point sources. Readily available tools and data combined with these two dispersion models provide an accurate representation of the air quality at a lower cost than the existing air quality monitoring systems in Japan. The dispersion models that applied to the regions of Japan in this study, remove the assumptions for uniform air quality within the vicinity of a monitoring station. The preliminary results of the present study are encouraging as air dispersion models providing emission data for assessing air quality in the different regions in Japan.

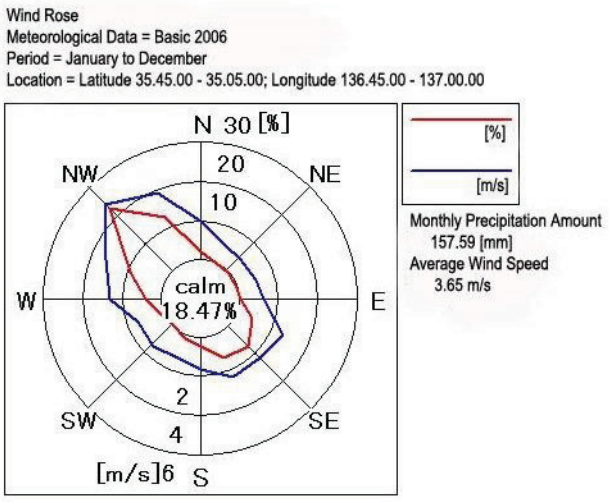

Figure 7. *The annual wind rose of the point–source area in 2006. Blue and red lines indicate the annual mean wind speed (m/s) and the frequency (%) of each direction, respectively.*

Acknowledgments

We wish to thank Professor Kobayashi Tomonao of the Faculty of Engineering of Gifu University for sharing resources and data with us. The deepest gratitude is also expressed to Naher Meherun the late wife of first author and a former doctoral student of the Graduate School of Agriculture, Gifu University, who passed away in February 2010.

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