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## CASE REPORT

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# Exposure Assessment of Mercury and Its Compounds by Dispersion Modeling: A Case Study in the Sea of Japan Coastal Area

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**Abstract:** In Japan, mercury and its compounds were categorized as hazardous air pollutants in 1996 and are on the list of “Substances Requiring Priority Action” published by the Central Environmental Council of Japan. The Air Quality Management Division of the Environmental Bureau, Ministry of the Environment, Japan, selected the current annual mean environmental air quality standard for mercury and its compounds of  $0.04 \mu\text{g}/\text{m}^3$ . Long-term exposure to mercury and its compounds can have a carcinogenic effect, inducing eg, Minamata disease. This study evaluates the impact of mercury emissions on air quality in the coastal area of the Sea of Japan. Average yearly emission of mercury from an elevated point source in this area with background concentration and one-year meteorological data were used to predict the ground level concentration of mercury. The annual mean concentration distribution of mercury and its compounds were calculated for the middle part of Honshu Island, which served as a background level of mercury concentration for the coastal are of the Sea of Japan. To estimate the concentration of mercury and its compounds in air of the local area, two different simulation models have been used. The first is the National Institute of Advanced Science and Technology Atmospheric Dispersion Model for Exposure and Risk Assessment (AIST-ADMER) that estimates regional atmospheric concentration and distribution. The second is the Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model (METI-LIS) that estimates the atmospheric concentration distribution in the vicinity of facilities.

**Keywords:** dispersion modeling, atmospheric mercury concentration, environment, mercury emission, Sea of Japan

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## Introduction

In Japan, mercury and its compounds were categorized as hazardous air pollutants in 1996 and are on the list of “Substances Requiring Priority Action” published by the Central Environmental Council of Japan.<sup>1</sup> The Central Environmental Council published its second report entitled “Future Direction of Measures against Hazardous Air Pollutants” in October 1996, which also proposed that voluntary action to reduce emission, as well as investigation of hazards, atmospheric concentration, and sources of pollutants, should be promoted. Although the industrial emission of mercury and its compounds in Japan has decreased in recent years, primarily due to voluntary reduction from industrial sources, the risks of exposure to this pollutant have remained largely unknown.

Mercury is a natural trace component in the environment. Notwithstanding, the bioaccumulation of methyl mercury via the food chain, especially through fish, concentrates the mercury and poses serious toxicity hazards in the biosphere.<sup>2</sup> For that reason, natural and anthropogenic emissions of mercury in the environment,<sup>3</sup> its transportation and fate,<sup>4,5</sup> and its adverse effect on human health and the ecosystem<sup>6</sup> have all attracted great attention as aspects of a major environmental problem. Stack emissions from coal-combustion power industry include both vapor and particle-bound phases. Hg (II)<sup>7</sup> can be inorganic (eg, mercuric chloride, HgCl<sub>2</sub>) or organic (eg, methyl mercury, MeHg). It can also be present as particulate mercury (eg, mercuric oxide, HgO, or mercury sulfide, HgS). In the global atmosphere, Hg (0) is the dominant form. Hg (II) typically constitutes a small percentage of total mercury and is predominantly in the gas phase. The MeHg concentration in the atmosphere is negligible, about a factor of 10%–30% lower than the Hg (II) concentration, according to analysis of precipitation samples.<sup>8</sup> However, Hg (II) becomes methylated in water bodies, where it can bioaccumulate in the food chain. Hg (0) is sparingly soluble 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, so is removed rapidly by wet and dry deposition processes. Particulate mercury is mostly present in the fine fraction of particulate matter, although some particulate mercury may be present in coarse particulate matter.<sup>9</sup>

The exposure concentration of mercury and its compounds should be estimated both on a regional scale as well as on a local scale, not only because the concentration of mercury and its compounds in the general environment is important (ie, the area which includes most of the total population), but also because those in the vicinity of industrial sources (ie, areas of high concentration) are expected to be associated with relatively high-risk areas. In this study, two different models were used to assess the extent of exposure, ie, AIST-ADMER (National Institute of Advanced Science and Technology Atmospheric Dispersion Model for Exposure and Risk Assessment), which estimates regional concentration and distribution of hazardous chemical substances,<sup>10,11</sup> and METI-LIS (Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model) which estimates the concentration and distribution in the vicinity of facilities.<sup>12</sup>

Gaseous mercury, including both Hg (0) and Hg (II), were considered as input data for total mercury emission for this two air pollutant dispersion models, whereas more than 99.5% of the mercury in the stack emissions was in gaseous form and the proportion in particulate form was extremely low in Japan.<sup>13</sup> Since the flue gas treatment systems of the coal combustion facilities are very excellent in Japan, Hg (II) concentration from the stack is also very low.

This study was designed to estimate the concentration of mercury and its compounds in the coast area of Japan, whereas the above two models were used for the exposure assessment of mercury and its compounds.

## Methods

### AIST-ADMER model

AIST-ADMER<sup>10,11</sup> version 1.5e is a series of models and systems designed for estimating atmospheric concentration of chemicals and assessing their exposure, developed by the National Institute of Advanced Industrial Science and Technology. The functions include:

- Generation and confirmation of meteorological data
- Generation and confirmation of chemical substance emission data
- Calculation of atmospheric concentration and deposition of chemicals

- Graphical images of calculation results
- Calculation result 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. Exposure assessment data of a 5 km × 5 km square spatial grid with a resolution of six time zones for an average of one month can be calculated. Generally, using models requires preparation of various data, such as obtaining meteorological data, creating target substance emission data, and setting calculation parameters, in order to estimate the atmospheric concentration of chemicals and assess their exposure.

Meteorological input data edited for AIST-ADMER is required for calculating its simulation. In this study, basic meteorological data, calculated monthly for a year, ie, from January to December 2006, have been used for AIST-ADMER calculations. Basic meteorological data were produced from AMeDAS data, whereas solar radiation and cloud amount were obtained from individual weather stations.

Simulation calculated by the AIST-ADMER needs information on target substances, such as the amount of emission and location of emission. AIST-ADMER contains a function for creating the emission grid data required for calculation. The methods used for creating emission grid data can be classified mainly into two types, ie, point sources, which specify a location using latitude and longitude, and enter the emission generated from the location, and area sources, which specify emission for each prefecture or city, and allocate the emission to calculation grids according to indices such as population, area, industrial statistics, and traffic volume.

The AIST-ADMER calculation range consists of a number of calculation grids. A unit of calculation range always carries out AIST-ADMER operations, such as creation of AIST-ADMER meteorological and emission grid data and performing the calculations. During the simulation period, it is recommended to select a calculation range that includes target ranges. In total, 11 calculation ranges are obtained by dividing the overall Japanese region preregistered in AIST-ADMER. In addition, an arbitrary calculation range can also be created. The general flow process for analysis using AIST-ADMER is shown in Figure 1.

#### Preparation for calculation

Creation and selection of target calculation range

Creation of meteorological data in target range

Creation of emission grid data for target substance



#### Performing calculation

Confirmation of data and parameters to be used for calculation

Creation of a calculation case

Performing calculation



#### Confirmation and analysis of calculation results

Confirmation of calculation results

Analysis using calculation results

**Figure 1.** General flow of analysis process using National Institute of Advanced Science and Technology Atmospheric Dispersion Model for Exposure and Risk Assessment (AIST-ADMER).

## METI-LIS model

METI-LIS<sup>12</sup> is a user-friendly computer model developed originally by the Ministry of Economy, Trade and Industry. METI-LIS version 2 is now available in English for downloading from an online site. This model gives special importance to expressing the downdraft effect, which often affects dispersion from low emission sources, and gives solutions of a simple Gaussian plume and puff formula for elevated sources. In addition to short-term estimation using fixed meteorological conditions, a long-term average estimation can be obtained using the model when hourly meteorological datasets are prepared by users.

The METI-LIS model is based on a Gaussian plume equation<sup>14-17</sup> that assumes steady-state conditions. This equation is used to model emissions from a point source, such as a smokestack or exhaust outlet. This section deals with the base Gaussian dispersion equation, the plume-rise height of exhaust gas, methods of determining dispersion parameters, methods of modeling downwash effects caused by buildings

neighboring the emission source, and the application conditions for the dispersion model.

The steady-state Gaussian plume model is used as a dispersion equation for point sources. For each source and each hour, the origin of the calculation's coordinate system is placed at ground level 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 calculation of concentration at each time period. The conversion method in the  $x$  axis and  $y$  axis directions 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.<sup>14–17</sup>

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

where  $c$  = concentration in the  $x$ ,  $y$ , and  $z$  directions ( $\mu\text{g}/\text{m}^3$  ppb, ppm, or other units),  $q$  is the pollutant emission rate ( $\text{m}^3 \text{ N}/\text{sec}$ ), whereas  $\text{m}^3 \text{ N}$  is the volume of a gas under standard conditions ( $0^\circ\text{C}$ , 1 atmosphere),  $N$  is the symbol for normal,  $v$  is the vertical term (Equation 2),  $u_s$  is the mean wind speed at release height ( $\text{m}/\text{sec}$ ), and  $\sigma_y$ ,  $\sigma_z$  are the dispersion parameters in the lateral and vertical directions ( $\text{m}$ ).

The vertical term,  $V$ , represents the distribution of the Gaussian plume in the vertical direction. This term includes the calculation point elevation and the effects of height caused by the emitted plume rise (effective plume-rise height).<sup>14–17</sup>

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

where  $z_r$  is the elevation of the calculation point ( $\text{m}$ ) and  $h_e$  is the effective plume-rise height ( $\text{m}$ ).

The wind profile power law<sup>18–20</sup> (equation 3) is the relationship between wind speeds at one height, and those at another, which converts the observed wind speed to an equivalent wind speed at the actual height of the source. The wind profile of the atmospheric boundary layer (surface to around 2000 m) is generally

logarithmic in nature, and is best approximated using the log wind profile equation that accounts for surface roughness and atmospheric stability, and is used in the METI-LIS model. The power law equation is of the form:

$$u_s = u_{\text{ref}} \left( \frac{h_s}{z_{\text{ref}}} \right)^p \quad (3)$$

where  $u_s$  is the wind speed at the stack outlet height ( $\text{m}/\text{sec}$ ),  $u_{\text{ref}}$  is the wind speed at the measurement height ( $\text{m}/\text{sec}$ ),  $h_s$  is the stack outlet height ( $\text{m}$ ), and  $z_{\text{ref}}$  is the wind speed measurement height ( $\text{m}$ ). The wind profile exponent,  $p$ , is set according to stability. The values shown in Table 1 can be used as average values.

When the source emitting air pollutants is a stack, the stack's tower acts as a drag to the wind, which produces a downwash, known as stacktip downwash. When the exit velocity of the exhaust gas from the source is less than 1.5 times the wind speed, a correction is applied to the stack height corresponding to the stacktip downwash. This method adjusts the height of the physical stack as follows:

$$h'_s = h_s + 2d_s \left( \frac{v_s}{u_s} - 1.5 \right) \quad (4)$$

Equation 4 is applicable when  $v_s < 1.5 u_s$

$$h'_s = h_s \quad (5)$$

Equation 5 is applicable when  $v_s \geq 1.5 u_s$  where  $h'_s$  is the modified physical stack height ( $\text{m}$ ),  $d_s$  is the stack diameter ( $\text{m}$ ), and  $v_s$  is the exhaust gas exit velocity ( $\text{m}/\text{sec}$ ). This modification is not applied when downwash effects due to building are calculated.

**Table 1.** Standard values of power exponent  $P$  used in wind-speed adjustments, whereas the ISC model<sup>24,25</sup> applies these values to rural areas.

Stability category	Rural exponent $P$
A	0.07
B	0.07
C	0.10
D	0.15
E	0.35
F	0.55

If the gas emitted from the source is not significantly warmer than the ambient temperature and the wind speed is slow, a buoyancy-induced plume rise can be disregarded. This treatment is allowed for exhaust vents and ventilation towers that account for most of the sources of hazardous chemical substances.

In the METI-LIS model, plume rise equations for bent over, hot buoyant plumes are based on observations and data involving plumes from typical combustion sources, such as flue gas stacks from steam-generating boilers burning fossil fuels in large power plants. Therefore, the stack exit velocities were probably in the range of 20–100 feet/sec (6–30 m/sec), with exit temperatures in the range of 250 °F–500 °F (120 °C–260 °C). If the gas emitted by the source is comparatively warmer than the ambient temperature, the CONCAWE equation<sup>21,22</sup> is used as follows:

$$h_e = h_s + \Delta h \quad (6)$$

$$\Delta h = 0.175 Q_H^{1/2} u^{-3/4} \quad (7)$$

where  $h_e$  is the effective plume-rise height (m),  $h_s$  is the physical source height (m),  $\Delta h$  is the buoyancy-induced plume rise (m),  $Q_H$  is the emitted heat quantity (cal/sec), and

$$Q_H = \rho C_p Q (T_s - T_A) \quad (8)$$

where  $\rho$  is the gas density at 0 °C ( $1.293 \times 10^3$  g/m<sup>3</sup>),  $C_p$  is the isobaric specific heat (0.24 cal/kg/g),  $Q$  is the exhaust-gas volume per unit time (m<sup>3</sup> N/sec),  $T_s$  is the exhaust gas temperature (°C), and  $T_A$  is the ambient temperature (°C, default being 15 °C).

Equations that approximately fit Pasquill-Gifford curves<sup>23</sup> are used to calculate the dispersion parameters. This model uses the approximation equations shown in Table 2. The same equations are also used in the Industrial Source Complex (ISC) model.<sup>24,25</sup> These approximation equations are functions of the downwind distance from the source and find the lateral dispersion width ( $\sigma_y$ ) and the vertical width ( $\sigma_z$ ) of

equation 1, respectively. These dispersion widths are contingent on atmospheric stability,<sup>26</sup> which is determined by meteorological conditions. While 11 different categories can be accepted as atmospheric stability, those in the approximation equations are redivided into only six categories (ie, A–G). Table 2 shows the atmospheric stability category mapping between observed and approximation situation. Atmospheric stability category can be classified by wind speed and solar radiation, which are also available at different online sites.<sup>27</sup> In Table 2, the atmospheric stabilities,  $D_d$  and  $D_n$ , are the D stabilities during the daytime and night-time, respectively. The six atmospheric stability categories can be calculated from parameters used in Equation 1 to calculate  $\sigma_y$  and  $\sigma_z$  from Pasquill-Gifford curves, whereas the parameters are available from different online sources.<sup>28</sup>

This model also includes building downwash, terrain effects, and line source emissions. The METI-LIS model adopts a downwash scheme based on that of the US Environmental Protection Agency (EPA) ISC model, but the parameters in the dispersion widths describing the downwash effect are improved by incorporating the results of wind tunnel experiments. Another characteristic in which the METI-LIS differs from the ISC is the evaluation time, which affects the dispersion width especially in the y (cross-wind) direction, and can be adjusted for a simulation of short time dispersion observation.

## Atmospheric mercury emissions in Japan

Mercury release into the atmosphere as reported by the Pollution Release and Transfer Register<sup>29</sup> is shown in Table 3 for registered emissions and Table 4 for estimates from coal-fired power plants. Mercury discharge from coal-fired power plants was estimated from the mercury content of coal and coal consumption.

## Mercury emission sources

According to the Kida research report and data provided by the relevant industries,<sup>30,31</sup> the total amount of mercury released into the atmosphere by Japan

**Table 2.** Relationship between observed atmospheric stability and approximation equation index.

Atmospheric stability	A	A–B	B	B–C	C	C–D	$D_d$	$D_n$	E	F	G
Approximation equation	A		B		C		D		E	F	

**Table 3.** Registered mercury releases into atmosphere (kg/year) according to the Pollution Release and Transfer Register.<sup>29</sup>

Fiscal year	Amount of emission in atmosphere	Total emission
2001	325	4642
2002	98	4283
2003	14	14,400
2004	32	1772
5-year average	98	5601

was estimated to 24–28 metric tonnes per year, taking into account the release from specified facilities not reported by the Pollution Release and Transfer Register. In the combustion category, coal-fired power plants, industrial oil combustion boilers, medical waste incinerators, sewage sludge, and other wastes are considered to be significant mercury emission sources. In the manufacturing category, primary ferrous and nonferrous metal production, as well as cement production, are thought to be major contributors to atmospheric mercury emission in Japan. Per person emission of atmospheric mercury in Japan is 0.19–0.225 g/year.<sup>29–31</sup>

### Mercury emission assumptions

For this research, emission of mercury in Japan was estimated according to the mercury emission of Japan inventory report by Kida.<sup>30</sup> Coal consumption data for 2005 have been considered as the basis to measure mercury emission into the atmosphere in Japan, whereas the emission of atmospheric mercury in 2006 is almost similar to that of 2005. To produce 1000 MW of power, the amount of coal consumption is 305 MT/hour whereas the mean concentration of mercury in coal was 0.045 ppm, the

**Table 4.** Estimates of mercury releases from coal fired power plants (kg/year).

Fiscal year	Release into atmosphere	Release to public water body	Total
2001	766.7	3.5	770.2
2002	814.2	3.7	817.9
2003	890.0	4.0	894.0
2004	934.6	4.2	938.8
2005	981.6	4.4	986.0
5-year average	877.4	4.0	881.4

mean emission rate was 4.4 µg/kW/hour, and the mean emission ratio of mercury from stack is 27% of the total mercury of feed coal in the coal combustion power industry.<sup>32</sup> In the coal combustion power industries, the capacity in MW and coal combustion rate has been used as a basis of calculations of atmospheric mercury emission. In the sector of iron works, total production is  $69.5 \times 10^6$  MT and total atmospheric mercury emission is 5.7 MT, in the sector of cement plants, total production is  $79 \times 10^6$  MT and total atmospheric mercury emission is 35 MT, in the sector of chemical plants, total production is 9057 MT and total atmospheric mercury emission is 0.03 MT in Japan.<sup>33</sup> To calculate atmospheric mercury emission from each point source of iron works, cement plants, and chemical complexes, the yearly production capacity and yearly mercury emission from each sector has been considered as a basis of calculation by using simple unitary methods.<sup>29,31,33</sup> Yearly municipality and medical waste have also been considered as a large source of atmospheric mercury emission in Japan, whereas the emission of mercury into atmosphere has been distributed in each prefecture on the basis of population density in the present study.<sup>29–31</sup>

### Study area

The industrial source complexes considered in the present study were located (latitude 37.01.47 N and longitude 137.50.53 E) in the coastal area of the Sea of Japan Sea, which is the central region of Honshu, Japan's main island. A total of nine prefectures (Aichi, Mie, Gifu, Fukui, Ishikawa, Niigata, Nagano, Gunma, and Toyama), as a calculation range for AIST-ADMER, were considered for simulation of the distribution of regional mercury concentration. There are several heavy and medium scale units of different types of facilities in operation in this area. On the other hand, a small location in the coastal area of the Sea of Japan was selected as the site for calculation of the ambient concentration of mercury in the vicinity of a major industrial source (latitude 37.01.47 N and longitude 137.50.53 E) of mercury emission in Japan using the METI-LIS model, where this area has an estimated population of 48,653 and a density of 65.2 persons per km<sup>2</sup> as of 2006. The total land area is 746.24 km<sup>2</sup>.

## Results

Mercury emissions from different industrial sources, along with their source characteristics and meteorological data, are described in this section.

### Meteorological data

Meteorological data from 2006 were used for computer simulation by the Automated Meteorological Data Acquisition System (AMeDAS),<sup>34</sup> which provides hourly data at distance intervals of approximately 17 km throughout Japan, because the 2006 weather data are available for use by AIST-ADMER and METI-LIS. One-year meteorological data have been prepared for AIST-ADMER, consisting of four meteorological elements, including temperature, amount of precipitation, wind direction and speed, sunshine duration, and snow depth for each hour at different point locations for the whole of Japan. On the other hand, single point meteorological data in the vicinity of large coal combustion facilities in the coastal areas of the Sea of Japan were used for the METI-LIS model. One-year meteorological data, with wind direction, wind speed, temperature, sunlight ratio, and solar radiation were prepared for METI-LIS at hourly intervals.

### Emission data

Burning of fossil fuel (primary coal) is the largest single source of atmospheric mercury emission from human sources, accounting for 45% of total anthropogenic emission, although the emissions from combustion of medical, municipality, and industrial waste account for significant release of mercury into the atmosphere in Japan. It is very difficult to identify actual locations and amounts of mercury emission in Japan from point sources, because of a lack of reliable information about industrial assumptions and technologies used for calculating mercury emission, as well as confidentiality issues. In this study, the geographical location of some coal-firing facilities, such as power plants, iron works, cement plants, chemical complex, and heavy oil or gas combustion industries, are considered as large sources of atmospheric mercury emission in Japan, based on the report of the Japan Coal Energy Center,<sup>33</sup> which provided the input data for AIST-ADMER. Mercury emissions from municipality and medical wastes have also been taken into consideration as area sources of

prefectural mercury emission data for AIST-ADMER. To calculate the regional distribution of mercury concentration hypothetically, about 28 tonnes/year of mercury have been distributed throughout Japan. In addition, the amount of mercury emission from the point source of the coastal area in the Sea of Japan was calculated on the basis of production capacity and, sector-wise, total mercury emission throughout Japan was calculated using the unitary method.<sup>33</sup>

### Assessment of exposure

In this study, two different atmospheric dispersion simulation models were used to assess the distribution of release of mercury into the regional atmospheric environment and in the vicinity of industrial sources. The first, ie, AIST-ADMER, is a computerized model with a set of systems providing an estimation of atmospheric concentration of chemical substances and assessment of human exposure. The purpose of AIST-ADMER is to estimate the chemical concentration in relatively broader areas, such as Kanto, Kansai, or Chubu, as averages over a long period of time. This model can assess exposure as a monthly average in a 5 km × 5 km mesh and six different time zones. The second model, ie, METI-LIS, estimates the local concentration near industrial sources and emphasizes the importance of expressing the downdraft effect, which often affects dispersion from low emission sources, and gives solutions using simple Gaussian plume and puff formulae for elevated sources. In addition to short-term estimation using fixed meteorological conditions, a long-term average estimation can be obtained with the model by preparing hourly meteorological datasets.

### Regional concentration level

A regional scale atmospheric concentration of mercury in Japan was estimated for a 5 km × 5 km grid using AIST-ADMER. The input emission data were compiled from the results of the Pollutant Release and Transfer Register survey of 2005 (Tables 3 and 4) and a mercury emission inventory by Kida<sup>30</sup> and Japan Coal Energy Center.<sup>33</sup> Table 5 shows the input parameters for the AIST-ADMER. Mercury in the atmosphere primarily exists in the elemental gaseous form, ie, Hg (0), generally at about 95%,<sup>35</sup> and 5% of the total amount is typically present as divalent reactive gaseous mercury (Hg (II))

**Table 5.** Input parameters for the National Institute of Advanced Science and Technology Atmospheric Dispersion Model for Exposure and Risk Assessment.

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

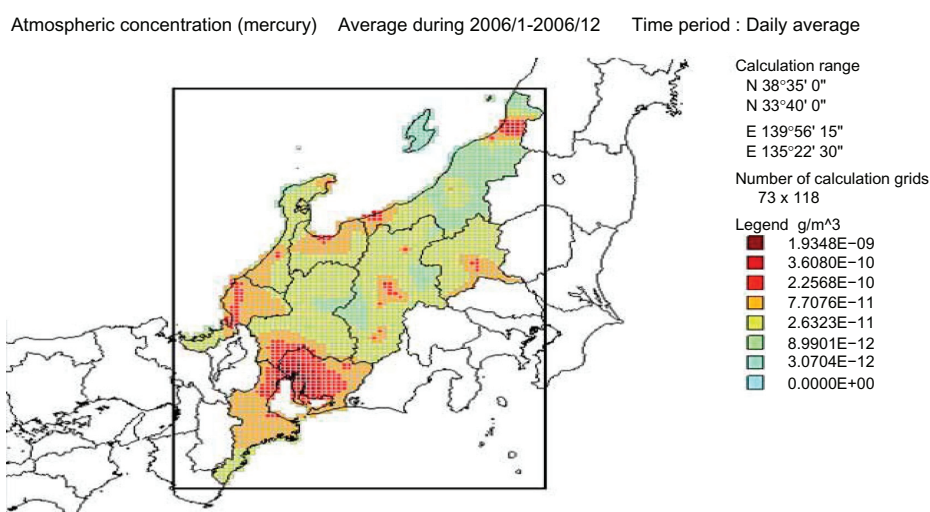
and particulate mercury. Hg (0) is believed to have an atmospheric lifetime of about one year, while Hg (II) and particulate mercury has much shorter atmospheric lifetimes.<sup>36</sup> The background concentration was determined to be 0.077 ng/m<sup>3</sup>. The value for the background concentration was selected from Figure 2 in the coastal area in the Sea of Japan and also comparing the monitoring survey results of hazardous air pollutants monitoring in 2006 by the Japanese Ministry of the Environment.<sup>37–41</sup>

Figure 2 shows the calculated annual distribution of concentration of atmospheric mercury in the central Honshu area in Japan. The atmospheric mercury concentration was relatively high in major urban areas such as Nagoya, Yokkaichi, because emissions from industrial facilities tend to be concentrated in these densely populated areas. The annual mean concentration of atmospheric mercury was calculated to be less than 0.225 ng/m<sup>3</sup> in industrial areas, 0.0263 ng/m<sup>3</sup> in nonindustrial areas, and sometimes the concentration

was greater than 1 ng/m<sup>3</sup> in the vicinity of major industrial point source areas.

## Concentration level near industrial sources

The ambient concentration of atmospheric mercury in the vicinity of major industrial sources was estimated using the METI-LIS. The coastal area in the Sea of Japan (latitude 37.01.47 N and longitude 137.50.53 E) was selected as the sites for the calculation; this site had a calculation domain of 6.5 km × 4.5 km with a grid spacing of 200 m, which included the largest point source located centrally. These domains corresponded to the 3 km × 3 km calculation grids of AIST-ADMER. The amount of emission from the point source of this area is about 0.35 tonnes/year. A large coal combustion facility in this area is one of the big sources of mercury emission into the atmosphere in Japan, pouring 3%–5% of mercury into the air every year.<sup>30</sup> It is assumed that the emission point was located at the center of the industrial yard, and that emissions are released from a height of 100 m, because no other detailed information regarding the specific location of the sources within factories was available. The emission from each point source was determined on the basis of total yearly mercury emission amount, total annual production capacity, and per unit annual production capacity in specific industrial sectors, as published in the Kida report for



**Figure 2.** Annual mean concentration distribution of atmospheric mercury calculated with the National Institute of Advanced Science and Technology Atmospheric Dispersion Model for Exposure and Risk Assessment (AIST-ADMER) in 2006. Nine areas (Aichi, Mie, Gifu, Fukui, Ishikawa, Niigata, Nagano, Gunma, and Toyama) are designated in the map, which provided the background concentration data in the calculation areas for the Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model (METI-LIS).

2007<sup>30</sup> and in the Japan Coal Energy Center report for 2005.<sup>33</sup> For example, mercury emission from a specific cement industry = {(total mercury emission from cement industries in Japan) × (production capacity of that industry)} ÷ total cement production capacity in Japan. It was assumed that the emission factors were constant for 365 days a year, 24 hours a day. AMeDAS<sup>34</sup> data were used as meteorological input data for the METI-LIS. Source contributions from other sources (eg, mobile sources or point sources located outside of the calculation domain) were not included in the input data for the METI-LIS. Source contributions from other sources were calculated with the AIST-ADMER and were superposed onto the results of the METI-LIS study as the background concentration.

Figures 3–5 show the distribution of mercury concentration at the coastal area in winter season, summer season, and one year average (2006), which were calculated using the METI-LIS simulation model. United place marks (latitude 37.01.47 N and longitude 137.50.53 E) on these figures represent industrial source location, with emission of 0.35 tonnes per year. In the winter season, the mean concentration of mercury was calculated to be 0.05–0.01  $\mu\text{g}/\text{m}^3$  near an industrial source. On the other hand, the mean mercury concentration was calculated to be 0.01–0.005  $\mu\text{g}/\text{m}^3$  in the summer season, which is much lower than that in the winter season due to the effect of boundary layer conditions in the

coastal area. In coastal regions, sea and land breezes can be important factors in the prevailing winds found in a given location. In the summer season, the temperature difference between the sea surface and the land surface is much greater than that during the winter season.<sup>42,43</sup> During summer, the effect of sea and land bridging causes a strong wind flow at ground level which driven pollutant matter far away from the source. As a result, the concentration of mercury is relative lower in the summer season. Also, in the winter season, thermal circulation of wind is very low, which causes a higher concentration of mercury in the vicinity of the source.

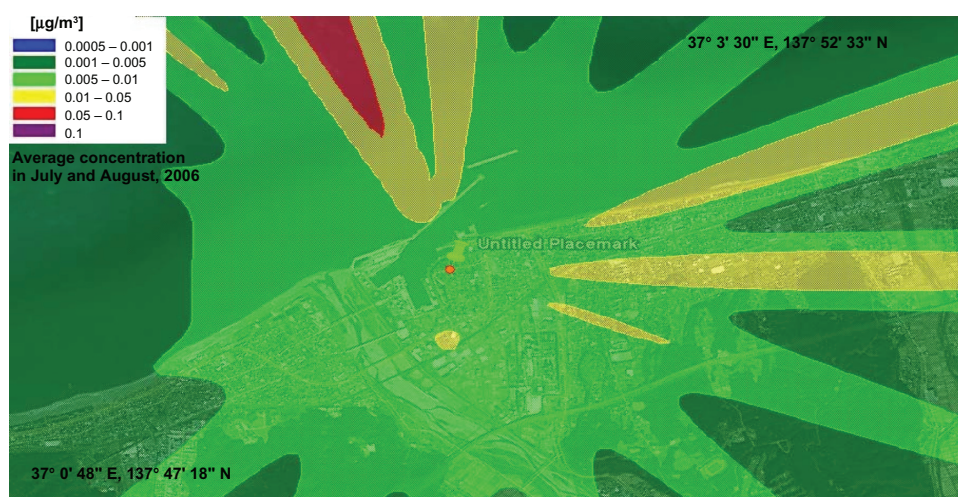
According to the results calculated in the METI-LIS model, although some people living in certain areas near industrial point sources were exposed to a significantly higher concentration of mercury than was the general population, the mercury concentration meets the air quality standard of the Japanese Ministry of the Environment. Figure 6 shows the annual wind rose plot, which gives a succinct view of how wind speed and direction are typically distributed at the location near the point source in 2006. The annual mean concentration was estimated not to exceed 0.04  $\mu\text{g}/\text{m}^3$  near the industrial source,<sup>1</sup> whereas a similar concentration level was found in different seasons.

## Discussion

In Japan, mercury was categorized as a hazardous air pollutant in 1996 due to its high carcinogenic potential.



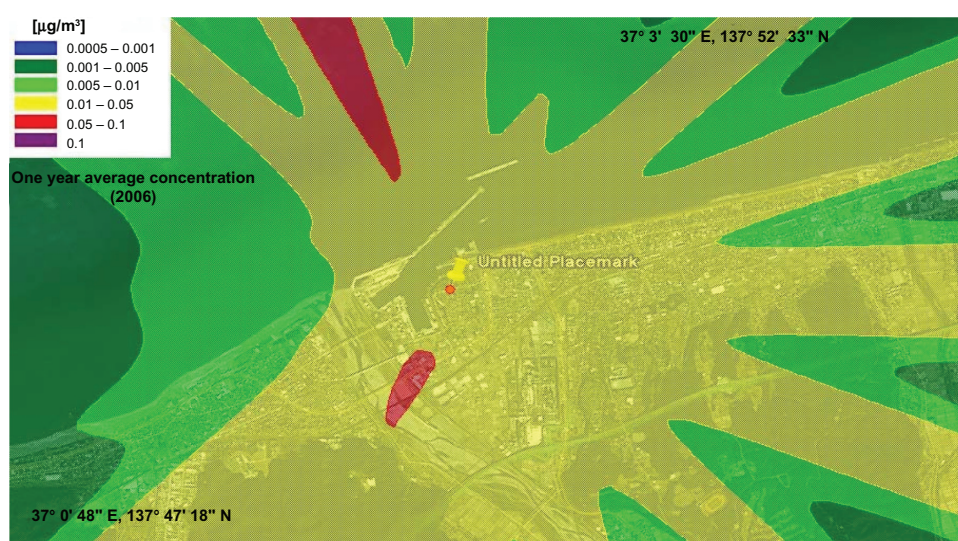
**Figure 3.** Average concentration distribution of mercury vicinity of a large point source in the coastal area in Japan Sea, calculated with the Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model (METI-LIS) in January and February, 2006. The united place mark represents industrial source locations.



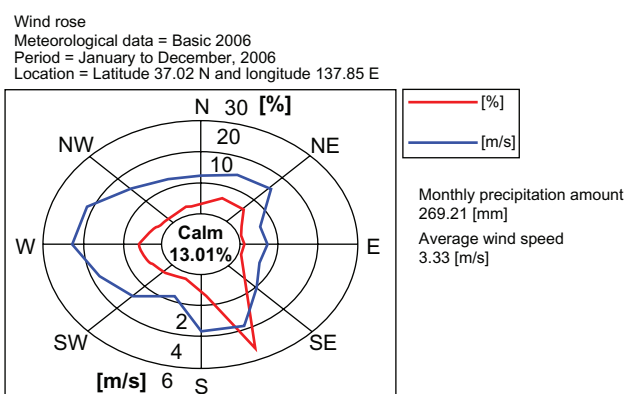
**Figure 4.** Average concentration distribution of mercury vicinity of a large point source in the coastal area in Japan Sea, calculated with the Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model (METI-LIS) in July and August, 2006. The united place mark represents industrial source locations.

The national government initiated a number of programs to evaluate emissions and assess ambient concentrations. Efforts to reduce mercury emissions started at a community level as part of public (local government)-private partnerships in Japan industry which have been ongoing since 2005 with support of a voluntary emissions reduction program. In 2003, Japan initiated the Pollutant Release and Transfer Register system, so emissions data for mercury from various sources could be made available. However, the precise amounts remain somewhat uncertain due to ambiguities in the estimation methodologies

used to evaluate mobile sources. The main source of mercury emission in Japan is from coal-fired cement plants, accounting for over 35% of total emissions in 2006. On the other hand, industrial emissions from primary ferrous metal production and coal combustion power plants made a significant contribution to atmospheric mercury emission in Japan in 2006. The assessment of exposure to ambient mercury concentration in Japan was performed using two different atmospheric dispersion models, ie, AIST-ADMER and METI-LIS. The present results indicate that the annual mean mercury concentration in residential



**Figure 5.** One-year average concentration distribution of mercury vicinity of a large point source in the coastal area of the Sea of Japan, calculated with the Ministry of Economy, Trade and Industry Low Rise Industrial Source Dispersion Model (METI-LIS) in 2006. The united place mark represents industrial source locations.



**Figure 6.** The annual wind rose of the point source area in 2006. Blue and red lines indicate annual mean wind speed (m/sec) and the frequency (%) of each direction.

areas generally amounted to less than  $0.22 \text{ ng/m}^3$ , but there are no sites that exceed  $0.04 \text{ } \mu\text{g/m}^3$  near industrial point sources. Although it is unrealistic to expect Gaussian models to predict the real world, the major purpose of the present assessment was to conduct a comprehensive analysis of exposure and distribution of mercury concentration, and thereby develop a detailed picture of current mercury exposure in the different industrial areas of Japan.

In the preliminary study reported here, small-scale and a medium-scale dispersion models of the different prefectures in the coastal area in the Sea of Japan were devised. The results show reasonable agreement with the monitoring data with respect to predicting localized atmospheric mercury concentration. 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 systems in Japan. A dispersion model applied to the prefectures of Japan removes the assumption of uniform air quality within the vicinity of a monitoring station. The preliminary results of the present study are encouraging as an air dispersion model providing emission data for assessing air quality in the different prefectures in Japan.

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