

# Development of the Method for Estimating the Emission Source of Air Pollutants

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**Key Words:** Exposure assessment, plume model, emission source information, effective stack height

## Abstract

For risk assessment of chemicals in a community around an emission source, we have developed the method for estimating the spatial distribution of long-term average concentrations of chemicals without information on the emission source. In our method, this distribution is estimated by using concentrations measured simultaneously at only five sampling locations. First, the source information was estimated from the measured data. Second, effective stack height ( $h_e$ ), which varied according to meteorological conditions, was transformed for the corresponding weather. Last, the long-term average distribution was estimated by sigma equation of the plume model. The source information was estimated by solving the simultaneous equations acquired from the plume equation with a random number from the normal distribution whose average was the measured concentration or wind direction.  $h_e$  was estimated by transforming to the height corresponding to the given atmospheric stability and wind speed by Briggs buoyancy-dominated plume rise. The estimated annual distribution acquired by the method mentioned above is in good agreement with those estimated by the ISCLT3 dispersion model, which was used in the case of known emission conditions.

## 1. Introduction

A community around an emission source, e.g., factories, is exposed to hazardous air pollutants (HAPs) emitted from the factories continuously over a long time with a low concentration. For risk assessment on human health of HAPs, it is necessary to know the distribution of the average concentrations of the HAPs.

Measurements of chemicals and model simulations are both well-known methods of determining the concentrations of chemicals<sup>1,2)</sup>. We can obtain true concentration values with

some uncertainty by direct measurements. However, since the concentration of chemicals in the atmosphere varies with emission and meteorological conditions, the measured value is the concentration only at the time when and location where the samples were taken. Therefore, much time, money, and many hands must be expended to obtain the distribution of the long-term average concentrations of chemicals by this method because of the necessity of many measurements. On the other hand, we can estimate the concentration of chemicals at an arbitrary time and location by model simulation. In order to use a model simulation, information of emission source, such as source location, emission rate, and other emission and meteorological conditions, is necessary. However, in Japan, it is difficult to obtain this information except for those people affiliated with the emission source.

Generally, most of the environmental measurements have been carried out in the restricted term, and resulted in the few data. These data are usually used for exposure assessment. However, because of the temporal and spatial variation of concentration, the amount of exposure estimated from measured concentration is not always correct. If the annually averaged concentration could be estimated from these few temporal data, the certainty of the exposure assessment would be greater.

We have developed the method for estimating the spatial distribution of an annually averaged concentrations using a few measured values at restricted times and places without any information concerning the emission source, using a combination of measurements and a model simulation.

## **2. Strategy**

In order to estimate the spatial distribution of annually averaged concentrations by model simulation, information on the emission source is necessary. Therefore, we developed the method for estimating such information on the emission source from temporary measured values and meteorological conditions at the time when samples were taken.

The estimated information based on the values at the time when measured samples were taken. Since some of the information varies with meteorological conditions, we developed the method for transforming emission conditions for various meteorological conditions.

The spatial distribution of an annually averaged concentration could be acquired from this estimated information using a common atmospheric dispersion algorithm that can yield the distribution using the frequency of meteorological conditions.

## **3. Estimation of Source Information**

### **3.1. Model selection**

To estimate the information on the emission source, we chose the plume model<sup>3,4)</sup> as the atmospheric dispersion model, due to the following reasons.

- The plume model is applicable for steady-state dispersion.
- The sigma equation of the plume model is easier than those of other dispersion models.

### 3.2. Estimation of source information at the time when samples were obtained

Under continuous emission, the chemical concentration at locations along the downwind direction is given by the sigma equation of the plume model<sup>3,4)</sup> as follows;

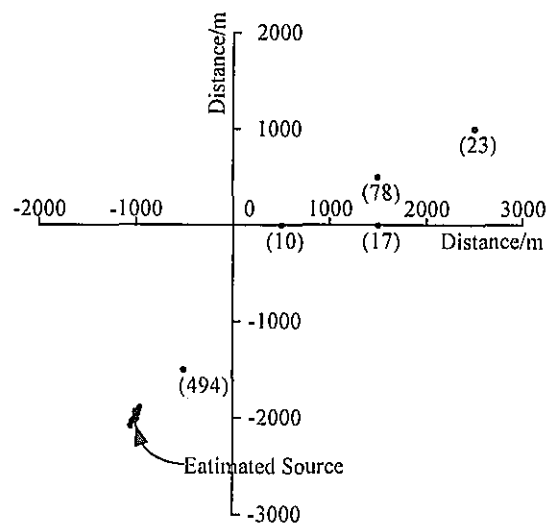
$$C = \frac{Q}{2\pi \cdot u_s \cdot \sigma_x \cdot \sigma_y} \exp\left(-\frac{1}{2} \frac{Y^2}{\sigma_y^2}\right) \left[ \exp\left(-\frac{1}{2} \frac{(z_r - h_e)^2}{\sigma_z^2}\right) + \exp\left(-\frac{1}{2} \frac{(z_r + h_e)^2}{\sigma_z^2}\right) \right] \quad (1)$$

where  $C$  is the concentration at the receptor,  $Q$  is the emission rate,  $u_s$  is the wind speed at the stack height,  $h_e$  is the effective stack height,  $Y$  is the crosswind distance from the source to the receptor, and  $z_r$  is the receptor height above ground level.  $\sigma_y$  and  $\sigma_z$  are dispersion parameters for horizontal and vertical directions, respectively.

Generally, stack height is much taller than human height. For the purpose of human risk assessment,  $z_r$  is negligible.  $C$  and the locations where samples were taken are known through "measured".  $\sigma_y$  and  $\sigma_z$  are functions of downwind distance,  $X$ . The necessary information for estimating the spatial distribution are five parameters:  $X$ ,  $Y$ ,  $Q$ ,  $u_s$ , and  $h_e$ .  $Q/u_s$  is treated as a single parameter because it is not possible to split  $Q$  and  $u_s$ .

Four simultaneously measured concentrations and locations were used to solve the simultaneous equations from sigma equation of the plume model. In addition, the data from one sampling point were used for validation.

The measured concentration includes uncertainty. And the wind direction includes error when we use a 16-direction system, for example, north wind does not exactly blow from strict north. Therefore, the distribution of the possible estimated emission locations was calculated from a random number within the normal distribution whose average was the measured concentration or wind direction. **Fig.1** illustrates an example of estimating the source information from five concentrations and locations calculated by the ISCST3<sup>5,6)</sup>.



**Fig.1** Source Estimation from concentration surrounding the emission source. Points with a value given in parentheses are measured concentration in  $\mu\text{g}/\text{m}^3$ .

Parameters; the area type is rural, the wind speed is 4 m/s, the wind direction is SW, and the atmospheric stability is C.

The emission location was determined by selecting a location from within the estimated distribution. Thereafter,  $h_e$  and  $Q/u_s$  could be calculated from the source location.

The wind power law is usually adapted in order to calculate wind speed at a certain height, for example, stack height, from the wind speed at a reference measurement height<sup>4,6</sup>. The power law equation is

$$u_s = u_{ref} \left( \frac{h_s}{z_{ref}} \right)^p \quad (2)$$

where  $u_{ref}$  is the observed wind speed,  $z_{ref}$  is the reference measurement height,  $h_s$  is a certain height at which wind speed is necessary and  $p$  is the wind profile exponent.

Since  $h_s$  is lower than  $h_e$ , it can be estimated from  $h_e$  by the person taking the exposure assessment. When  $h_s$  is determined,  $Q$  can be calculated from simultaneous equations mentioned above.

### 3.3. Estimation of effective stack height under variable meteorological conditions

The values of  $u_s$  and  $h_e$  varies with meteorological conditions.  $u_s$  can be easily calculated from the wind speed aboveground using the wind power law.

$h_e$  is expressed as the sum of  $h_s$  and the height the stack gas rises after emission from the stack, called plume rise. The stack gas rises due to the momentum of emission and the buoyancy of the stack gas itself. The rise due to momentum is not dependent on meteorological conditions, whereas the rise due to buoyancy varies with meteorological conditions. However,  $h_e$  cannot easily be calculated using only the parameters discussed above.

Therefore, we developed the method for estimating  $h_e$  under various meteorological conditions. In this study, we used Briggs plume rise equation<sup>6,7</sup> in order to calculate  $h_e$ .

#### 1) Atmospheric Stability

Atmospheric stability is the factor which most influences buoyancy and depression force of stack gas in the atmosphere. Thus, plume rise is affected by atmospheric stability. We examined the relationship between plume rises under neutral and other situation.

When stack-tip downwash is neglected and plume rise is dominated by buoyancy, the relationship between plume rise is expressed as follows:

$$Pe = Pe_D \left( \frac{h_s}{z_{ref}} \right)^{p_D - p} \quad (\text{unstable}) \quad (3)$$

$$\log Pe = \frac{4}{9} \log Pe_D + \left( \log 2.6 - \frac{4}{9} \log 21.425 - \frac{1}{3} \log s + \frac{1}{9} \log u_{ref} \right) + \left( \frac{4}{9} p_D - \frac{p}{3} \right) (\log h_s - \log z_{ref}) \quad (\text{stable and } F_b < 55) \quad (4)$$

$$\log Pe = \frac{5}{9} \log Pe_D + \left( \log 2.6 - \frac{5}{9} \log 38.71 - \frac{1}{3} \log s + \frac{2}{9} \log u_{ref} \right) + \left( \frac{5}{9} p_D - \frac{p}{3} \right) (\log h_s - \log z_{ref}) \quad (\text{stable and } F_b \geq 55), \quad (5)$$

where,  $Pe$  is the plume rise under the given stability,  $Pe_n$  is the plume rise at neutral,  $s$  is the stability parameter, and  $F_b$  is the buoyancy flux parameter, calculated by

$$F_b = gv_s d_s^2 \left( \frac{\Delta T}{4T_s} \right) \quad (6)$$

where,  $g$  is the acceleration due to gravity,  $v_s$  is the stack gas exit velocity,  $d_s$  is the stack inside diameter, and  $\Delta T = T_s - T_a$ ,  $T_s$  is the stack gas temperature, and  $T_a$  is the ambient temperature.

We examined the condition under which  $F_b$  became greater than 55. This only occurred when the stack inside diameter was very large, gas exit velocity was very high, and gas exit temperature was very high. Under ordinary conditions,  $F_b$  was less than 55. Thus, we used the equations (3) and (4) in order to estimate  $h_e$ .

When plume rise is dominated by momentum, the equations (3) and (4) cannot be used for estimation of  $h_e$ . Plume rise is dominated by momentum only when gas exit temperature does not exceed the ambient temperature. Under ordinary conditions, plume rise is dominated by buoyancy.

When stack-tip downwash is considered, plume rise is calculated to be higher. Stack-tip downwash can be calculated by

$$\text{Stack - tip downwash} = 2d_s \frac{v_s}{u_s} \quad (7)$$

In Briggs plume rise equation, stack-tip downwash is considered only when 1.5 times of  $u_s$  is larger than  $v_s$ . Under that condition stack-tip downwash is calculated as a value smaller than  $3d_s$ . In most cases, the stack height is estimated to be much higher than  $3d_s$ . Since we cannot know the emission conditions, stack-tip downwash was neglected in our method.

## 2) Wind speed aboveground

When we consider the effect of wind speed aboveground, we should examine the relationship between the plume rises at various wind speed. We propose the plume rise factor. The plume rise factor is calculated as follows;

$$\text{Plume rise factor} = \frac{\text{Plume Rise at the given wind}}{\text{Plume Rise at 1 m / s wind}} \times u_{ref} \quad (8)$$

When stack-tip downwash can be neglected, for example, when  $u_s$  is much slower than  $v_s$ , the plume rise factor is equal to 1 under neutral and unstable situation, and  $u_{ref}^{2/3}$  under stable situation. In this study, we neglected stack-tip downwash for reason discussed in 3.3.1). Plume rise was estimated using the factor mentioned above.

## 3) Estimation method of $h_e$

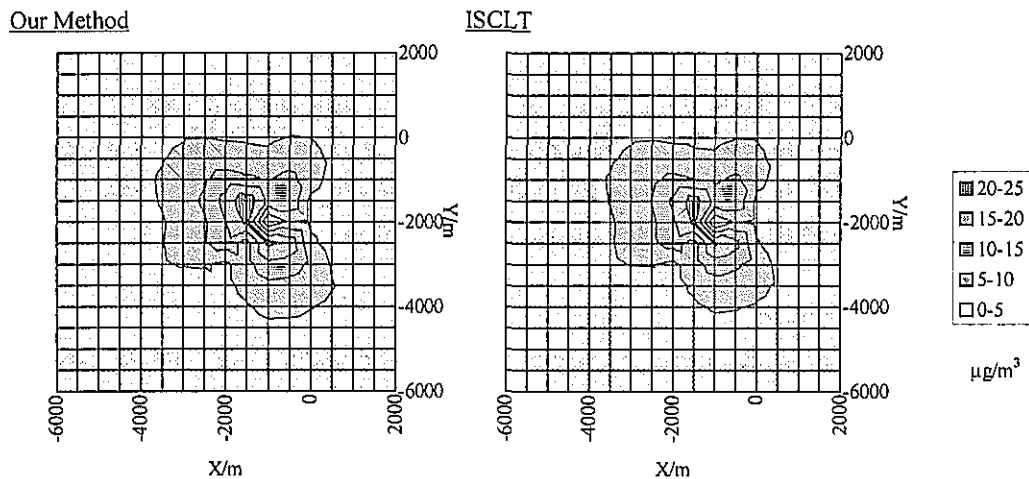
Based on the result above, we decided on the estimation method of  $h_e$  as follows:

- (1) Plume rise at the time the source information is estimated is calculated from the effective stack height and the estimated stack height.
- (2) Using the relationship to the wind speeds, plume rise is calculated at the stability when source information was estimated and a 1m/s wind.

- (3) Using the relationship to the stabilities, plume rise is calculated at the neutral stability and a 1m/s wind.
- (4) Using the relationship to the stabilities, plume rise is calculated at the desired stability and a 1m/s wind.
- (5) Using the relationship to the wind speeds, plume rise is calculated at the desired stability and wind speed.
- (6) Effective stack height at the desired stability and wind speed is calculated by the plume rise calculated above and the stack height.

#### 4. Estimation of Spatial Distribution of Annually Averaged Concentration

The spatial distribution can be estimated from the emission location and  $h_e$  obtained by the method developed above. **Fig.2** illustrates the estimation from the source information acquired in **Fig.1**. In addition, **Fig.2** illustrates the result estimated by the ISCLT3<sup>5,6)</sup>, which was used in the case of known emission conditions, using the emission information to calculate the concentrations and the locations in **Fig.1**. The distribution estimated by our method is good agreement with those estimated by the ISCLT3 dispersion model.



**Fig.2** Annual distribution results estimated by our method and by the ISCLT3.

##### Parameters

Annual meteorological frequencies were obtained from Environmental Surveillance Center of Ichihara City.

Our method: parameters are cited in **Fig.1**.

ISCLT3: the source location is (-1000, -2000), the emission rate is 20 g/s, the stack height is 25 m, the stack gas temperature is 400 K, the stack gas exit velocity is 12 m/s, and the stack inside diameter is 0.4 m.

#### 5. Examples of Distribution Estimated from the Simultaneously Measured Concentrations and the Locations

Using the method discussed above, we estimated the source information and the distribution of annually averaged concentrations of the chemicals from the measured data.

**Table I** The data used to estimate source information and distribution of annually averaged concentrations of chemicals

Location No.	Concentration ( $\mu\text{g}/\text{m}^3$ )	Error (%)	Location*	
			X (m)	Y (m)
1	1214	3	-6262.5	-5062.5
2	11.0	20	-5087.5	-3350
3	23.8	20	-4912.5	-2662.5
4	2.9	20	-5350	-3737.5
5	25.0	20	-4250	-250

Object: Ethylene

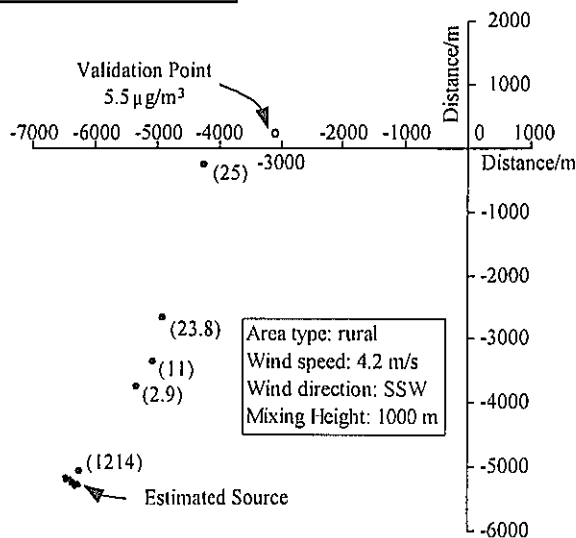
\*Location is expressed on a Cartesian grid whose origin is at the Environmental Surveillance Center of Ichihara City.

**Table I** shows the concentrations of ethylene and the sampling locations in Ichihara City on Mar. 4, 1996. We estimated the source information and the distribution of ethylene in 1995 from these data.

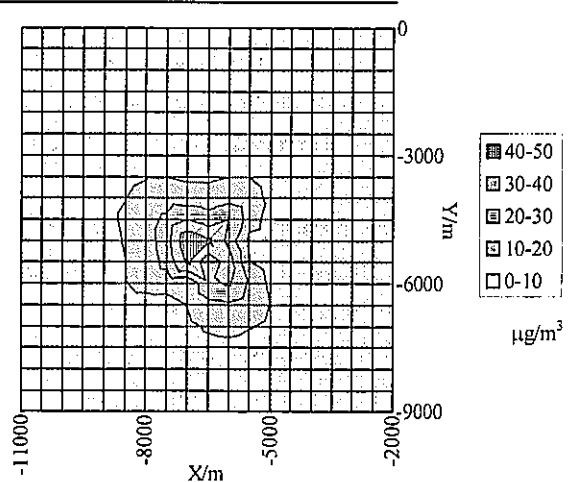
**Fig.3** shows the results of estimation of the source and the distribution of annually averaged concentrations of ethylene in 1995 using the data above. We estimated that the location of the source is (-6361.62, -5260.75). Estimation of the location of the source allowed  $h_c$  to be calculated as 26.92 meters. We estimated  $h_s$  as 25 meters.  $Q$  was estimated as 36.51 g/s from these results.

In order to ascertain the above results, we estimated the concentration at another locations from the above results using the plume model, and compared the estimated concentration with the measured one. The measured concentration of ethylene at the location (-3100, 237.5) was  $5.5 \mu\text{g}/\text{m}^3$  and that estimated was  $3.65 \mu\text{g}/\text{m}^3$ . We judged that the estimated concentration is in

### Source Estimation



### Annual Distribution Estimation



**Fig.3** The results of estimation of the Source and the distribution of annually averaged concentration (annual distribution) from measured data.

Parameter: meteorological frequencies are at Ichihara in 1995.

good agreement with the measured value.

## 6. Conclusion

For the risk assessment of HAPs to the community around the emission source, we have developed the method for estimating the spatial distribution of chemicals around the emission source. In our method, the spatial distribution of annually averaged concentrations can be estimated with the concentrations measured simultaneously at only five locations, even without information on the emission source. The spatial distribution of the concentration of chemicals was in good agreement with those estimated by the ISCLT3, which is used in the case of known emission conditions.

## 7. Acknowledgement

This work has been supported by CREST (Core Research for Evolutional Science and Technology) of the Japan Science and Technology Corporation (JST). Meteorological conditions of Ichihara were provided by Environmental Surveillance Center of Ichihara City.

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