Approach to risk assessment for air pollutants using PRTR data and atmospheric dispersion model

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Key Words: PRTR, ISC (Industrial Source Complex Model), simulation model, air pollutants, benzene, NOx

Abstract

In order to utilize the emission data of PRTR in risk assessment for human health, the environmental simulation models for estimating the environmental concentrations using emission data are required. In Japan, the environmental concentrations of the target substances, such as SOx and NOx, of Total Pollutant Load Control have been estimated to a high accuracy, using simulation models (Total Control Model). However the Total Control Model is considered to be unsuitable for risk assessment and communication because the program source code and calculation conditions are closed to the public. In this study, the NOx concentration was calculated by simulation using the ISC (Industrial Source Complex Model), which is used in health risk assessment and whose source code is opened to the public. The results were compared with the results obtained using the Total Control Model. It was shown that the error between the value calculated using the Total Control Model and our modeling approach using ISC was 10% or less on average in each region, and that the approach using ISC was adaptable for roughly estimating NOx concentration. As a result of the calculation using the emission data of benzene reported by the PRTR Pilot Project of Kawasaki city in fiscal 1998, the ratio of the calculated concentration to the measured concentration of benzene was under 1/4 of that of NOx. Reasons for underestimating benzene concentration compared to NOx concentration were discussed.

1. Introduction

In Japan, the PRTR Law (commonly used name) was promulgated in July 1999 and will be enforced in fiscal 2001 (JEA, 1999). After enforcement, the emission data of many chemical substances into air, water and soil will become available. In order to evaluate and manage the risk due to air pollutants, simulation models must be used to examine whether or not the relation between reported emission data and atmospheric concentration is reasonable. However, since the emission data of chemical substances into the environment have rarely been reported in detail, the consistency between emission data and environmental concentration has also not been examined. As an exception, for SOx and NOx, the consistency between the emission data and environmental concentrations has been examined using an atmospheric dispersion model in the target area, because SOx and NOx are target substances of Total Pollutant Load Control in Japan. Hereafter, the atmospheric dispersion model used for Total Pollutant Load Control will be called "the Total Control Model" for simplicity.

However, there are two reasons why the Total Control Model seems to be unsuitable for evaluating the risk based on the PRTR data. The one reason is that information necessary for the calculation is not available to the public. Calculation using the Total Control Model is usually entrusted to consulting companies by the central or local government, and the result is then reported. However, since the source code and the input parameters of the model used are not available to the public, recalculation or verification of the results can not be carried out by outsiders. In risk communication, it is necessary to release all information used in the calculations, because simulation models used to evaluate the risk should be usable by people from various viewpoints. Another reason is that input parameters for calculation are too detailed. In particular, the information on location and emission rate of individual sources is given in detail. However, the emission data reported by PRTR is expected to be composed of various types of data estimated by various methods, and thus is unacceptable for the Total Control Model.

In this study, we attempted to estimate the environmental concentration of benzene, which is a known human carcinogen (U.S. EPA, 1998), by using the emission data reported in PRTR and a simulation model with a source code available to the public. ISC (Industrial Source Complex Model) (U.S. EPA, 1995) was used as the framework of our modeling approach. ISC is an atmospheric dispersion model developed by the U.S. EPA for the risk assessment of air pollutants, and has been verified and revised several times. The source code of ISC is available to the public and can be downloaded for free from the web site (http://www.epa.gov/scram001/).

This study is composed of two parts. The first is the examination of the accuracy of our modeling approach by comparing the atmospheric NOx concentrations calculated using our model to those calculated using the Total Control Model. The second is the estimation of the atmospheric concentrations of benzene, by our modeling approach verified by NOx data and the emission data of benzene reported by the PRTR Pilot Project, and the discussion of the result. Since emission sources and spatial distributions of concentrations of benzene are known to be

similar to those of NOx (Kajihara et al., accepted), benzene is suitable for our modeling approach using NOx data.

2. Calculation condition

There are two types of model in ISC; one is called the LT (Long-Term Model) for estimating long-term average concentration and the other one is the ST (Short-Term Model) for estimating short-term average concentration. In this study, ISC-LT was selected because annual average concentration was predicted in the report of the Total Control Model, which was object of comparison. As an example of the Total Control Model for comparison with our modeling approach, the reports from Kawasaki city (Kawasaki city, 1998) and Tokyo Metropolis (Tokyo Metropolitan Government, 1997) concerning NOx were used.

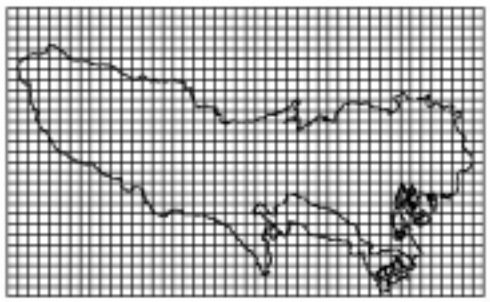

Figure 1 Blank map of Kawasaki and Tokyo with 2×2 km grid system

Emissions of NOx in each ward and source in Kawasaki city are shown in Table 1(a) (Kawasaki city, 1998). Although the source parameters have been set in more detail in the report of Kawasaki city (Kawasaki city, 1998) than in Table 1(a), specific values were not given in it. In this study, emission data for the calculation was prepared with a 2×2 km grid resolution (See Figure 1) using emission data for each ward. Uniform emission was assumed in each mesh. Kawasaki ward was divided into coastal and inland regions, since large-scale factories were considered to be concentrated in the coastal region. The source height was assumed to be 100m in the coastal region and 50m in regions other than the coastal region for factories, and 1.5m for sources other than factories. Since there is no sufficient description concerning sources for the municipalities surrounding Kawasaki city in the report (Kawasaki city, 1998), emissions from the sources in Kawasaki city only were considered in the calculation. The calculation points (receptors) were set at the center of each mesh. The parameters of dispersion, source, meteorology and land in our modeling approach are shown in Table 2.

The main differences between calculation conditions in our modeling approach and in the Total Control Model are shown in Table 3. In our modeling approach, parameters such as the types of dispersion equations, source parameters and meteorological parameters, were considerably simplified.

In the case of Tokyo, calculation conditions were set to be the same as in the case of Kawasaki city. Emissions of NOx in Tokyo Metropolis (Tokyo Metropolitan Government, 1997) are shown in Table 1(b), and NOx emissions in each municipality are shown in the Appendix. The source height of aircraft in Tokyo Metropolis was assumed to be 50m.

Table 1(a) NOx emission in Kawasaki City (fiscal 1993) [a)] (ton/yr)

Ward	Area (km^2)[b)]	Factories, [c)]	Automobiles	Vessels [d)]	Domestic [e)]	Total
Kawasaki	39.21	11,307	1,342	624	78	13,351
Saiwai	10.05	28	296		78	402
Nakahara	14.70	62	273		78	413
Takatsu	16.36	124	618		78	820
Tama	20.49	17	338		78	433
Miyamae	18.61	17	766		78	861
Asao	23.28	79	136		78	293
Total	142.70	11,634	3,768	624	548	16,574

a) The report for the countermeasures hereafter of nitrogen oxides and suspended particulate matters in Kawasaki city (Kawasaki city, 1998). Emission data for each ward was obtained by personal communication with Kawasaki City Environment Bureau. b) Handbook of municipalities across the nation in fiscal year 1998 (Self-government association of municipalities, 1998). c) Unknown emission in each ward, the total emission of 18 ton from small boilers and incinerators was divided into 5 ton for Kawasaki ward, 2ton for the other wards. d) The total emission from vessels was grouped into Kawasaki ward. e) The total emission from domestic was divided equally in each ward.

Table 1(b) NOx emission in Tokyo Metropolis (fiscal 1995) [a)] (ton/yr)

Area (km^2)[b)]	Factories	Automobiles	Vessels	Domestic	Aircrafts	Construction machines	Total
1776.3	9,895	44,999	1,482	9,437	1,827	7,428	74,888

a) The basic investigative report on the countermeasures against reduction of nitrogen oxides (Tokyo Metropolitan Government, 1997). b) Excluding the area of island regions. Handbook of municipalities across the nation in fiscal year 1998 (Self-government association of municipalities, 1998).

Table 2 Dispersion, source and meteorological parameters used in our modeling approach [a)]

Parameters	Parameter value or explanation
Dispersion equation	Plume equation
Dispersion parameters	Briggs urban dispersion parameters
Meteorological data	Hourly data (1997) from Tokyo observatory Annual frequency distributions were applied to whole object area
Stability	Calculated based on the Pasquill Stability Categories for every hour
Mixing heights (m) [b)]	800(A), 600(B), 500(C), 300(D), ∞(E), ∞(F) (values in parenthesis are stability levels)
Annual average temperature (℃)	16.7

Table 2 continued

Parameters		Parameter value or explanation
Source height (m)	NOx	Factories（Coastal region, Kawasaki） … 100
		Factories（Other wards, cities） … 50
		Other sources … 1.5
	Benzene	Factories … 10
		Other sources … 1.5
Source type		Area source (2×2 km) for all the sources
Receptor location, height		Location … Center of each mesh
		Height … 1.5 m
Terrain		Flat terrain was assumed
Land-use (surface roughness)		No revision
Reaction in ambient air		Neglected

a) The ISC model was used for the framework of our modeling approach. Not shown above; the ISC default conditions were used. b) ISC assumes unlimited vertical mixing under the conditions of stability levels E and F.

Table 3 Comparison of the calculation conditions between Total Control Model and our model

Conditions, Parameters	Total Control Model [a)]	Our model
Dispersion equation	Plume equation (under typical wind condition) Puff equation (under calm or low wind conditions)	Plume equation
Dispersion parameters	P-G parameters (used for plume equation) [b)] Turner parameters (used for puff equation) [b)]	Briggs urban parameters
Source parameters	Major sources : Set the locations and emissions for individual sources Minor sources : Area sources	Area source (2×2 km) for all sources
Meteorological conditions	Divided into several meteorological blocks	One condition for whole area

a) (Kawasaki city, 1998; Tokyo Metropolitan Government, 1997; JEA, 1995). b) Revised value by Japan Environment Agency (JEA, 1995).

3. Calculation results and validation of calculation condition

The distribution of calculated concentrations in Kawasaki city was higher in the eastern area (coastal area) and lower in the western area, as shown in Figure 2. Concentrations of NOx measured at the nine sites in Kawasaki city and concentrations calculated by our modeling approach and Total Control Model at the same sites are shown in Table 4. The ratios of the concentrations calculated by our modeling approach to those by the Total Control Model were 0.61-1.33. The average concentrations measured at nine sites was 16.5ppb in our model and 18.2ppb in the Total Control Model, and the ratio of the two averages was 0.91. The coefficient of determination between the concentration calculated using our model and the Total Control Model was 0.54, which showed significant correlation ($p = 0.05$) by Students' t-test. The ratio of the average calculated concentration to the measured one was only about 30% for both models. Since Kawasaki city has a small land area and slender landform, and lies

between two big cities, Tokyo Metropolis and Yokohama city, the ambient concentration in Kawasaki city seems to be considerably affected by sources in the outside the city.

The spatial distribution of calculated concentrations in Tokyo Metropolis is shown in Figure 3. Concentrations of NOx measured at nine sites in Tokyo and concentrations calculated using our model and the Total Control Model for the same sites are shown in Table 5. The ratios of the concentrations calculated using our model to those obtained using the Total Control Model were 0.29-1.29. There is one site at which the ratio is extremely low. This site seems to be affected by a nearby emission source. When data for this site were disregarded, the ratio of (our model)/(the Total Control Model) was in the range of 0.65-1.29. The average concentration of nine sites was 39.1ppb in our model and 40.5ppb in the Total Control Model, and the ratio of two averages was 0.96. The coefficient of determination between the calculated concentrations obtained using our model and the Total Control Model was 0.52, which showed significant correlation ($p = 0.05$) by Students' t-test. The ratio of calculated to measured concentrations was about 70% which was higher than the ratio calculated for Kawasaki city for both models. Since Tokyo Metropolis has a fairly large land area and numerous emission sources, atmospheric concentrations in Tokyo seem to be contributed to mostly by sources inside Tokyo.

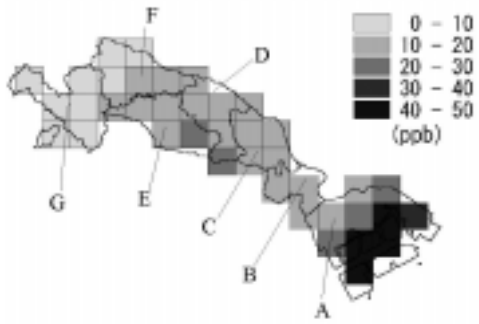

Figure 2 Calculated NOx concentration in Kawasaki
A: Kawasaki, B: Saiwai, C:Nakahara, D: Takatsu, E: Miyamae, F: Tama, G: Asao

Table 4 Measured and Calculated NOx concentrations in ambient air in Kawasaki (fiscal 1993) (ppb)

Monitoring stations (general environment)	Measured concentration	Calculated concentration [a)]	
		This study	Total Control Model
Daishi, Kawasaki	62.8	24.5	19.7
Tajima, Kawasaki	62.9	23.2	27.6
Kokusetsu Kawasaki, Kawasaki	62.8	19.3	16.7
Saiwai, Saiwai	66.5	17.1	27.9
Nakahara, Nakahara	60.7	11.4	15.8
Takatsu, Takatsu	62.9	17.9	13.4
Miyamae, Miyamae	62.4	20.0	26.1
Tama, Tama	51.7	9.1	8.8
Asao, Asao	39.0	6.2	7.6
Average	59.1	16.5	18.2

a) Sources only in Kawasaki city were considered

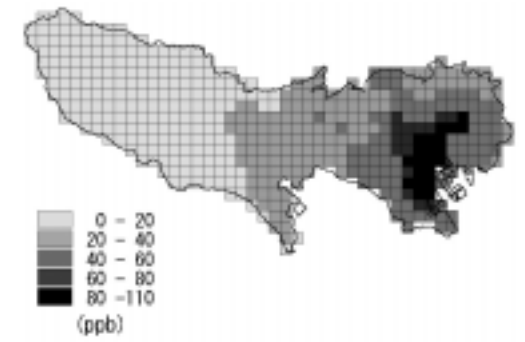

Figure 3 Calculated NOx concentration in Tokyo

In this study, sensitivity analysis of each parameter is insufficient. Therefore, the sensitivity of each parameter is described qualitatively. The most sensitive parameter was the type of source. In this study all sources are given as area sources, but when point sources were assumed, the calculated concentration decreased several times. The mixing height set for each atmospheric stability is shown in table 2; they also influence the result. The source height and calculation point heights also affect the result. The assumption that the heights of the sources were 100m or 50m for factories and 1.5m for the sources other than factories is consistent with common sense.

Table 5 Measured and calculated NOx concentrations in ambient air in Tokyo (fiscal 1995) (ppb)

Monitoring stations (general environment)	Measured concentration	Calculated concentration[a)]	
		This study	Total Control Model
Kokusetsu Tokyo, Shinjuku	64.9	69.0	65.0
Higashikoujiya, Ota	67.4	56.5	43.9
Setagaya, Setagaya	55.3	53.4	41.3
Hikawa, Itabashi	70.7	51.2	53.3
Haruecho, Edogawa	57.9	45.4	45.8
Honcho, Koganei	47.7	30.0	27.9
Nishiterakata, Hachioji	20.1	14.5	11.6
Narahashi, Higashiyamato	40.9	15.3	23.4
Katakura, Hachioji	65.3	16.9	52.3
Average	54.5	39.1	40.5

a) Sources only in Tokyo Metropolis were considered

As mentioned above, average concentrations calculated using our model were consistent with those calculated using the Total Control Model in an error range of about ± 10%. The correlation between the calculated concentrations using the two models for each site was also significant. Therefore, our modeling approach is applicable for roughly estimating NOx concentrations. The difference between the results of our modeling approach and those of the Total Control Model seems to be caused by the roughness of the input parameters in our model.

4. Environmental concentrations calculated using PRTR data

The PRTR Pilot Project was started in 1997 and enforced in four regions (Kawasaki city and the Shonan region in Kanagawa prefecture, the west Mikawa region in Aichi prefecture, Kitakyushu city in Fukuoka prefecture) in 1998. Kawasaki city, where the applicability of our model to NOx was verified, was selected as a target area for the calculation using the PRTR emission data.

Emission data of benzene in the coastal region, inland region and hill region of Kawasaki city were reported in the PRTR Pilot Project in 1998, as shown in Table 6. Emission in each mesh was set by the same method as the case of NOx. The calculation conditions, except for some parameters of source height, were the same as in the case of NOx. The height of factory emission sources for benzene was set to be 10m, which was lower than that for NOx, because benzene is used as a solvent or chemical raw material and is not considered to be emitted from stacks (Table 2).

Table 6 Benzene emission in Kawasaki city (fiscal 1997)[a)]

(ton/yr)

Regions	Factories	Auto-mobiles	Two-Wheelers	Vessels	Rail-ways	Gas stations	Total
Coastal	43.5	2.6	2.7	1.7	0	2.0	52.5
Inland	0	3.4	8.5	0	0	2.9	14.8
Hill	0	3.2	6.6	0	0	3.0	12.8
Total	43.5	9.2	17.8	1.7	0	7.9	80.1

a) PRTR Pilot Project Report in fiscal year 1998 (JEA, 1999)

Concentrations of benzene measured and calculated by our modeling approach at four sites in Kawasaki city are shown in Table 7. The calculated concentrations were about 4-7% of the measured concentrations. The average of concentrations calculated for the same sites was 0.25 μg/m^3, though the average measured concentration for the four sites in Kawasaki city was 4.13 μg/m^3. The ratio of the calculated to measured values was only 6%, which was 1/4 or less of the ratio for NOx.

Table 7 Measured and calculated benzene concentrations in ambient air in Kawasaki city (fiscal 1997) (μg/m^3)

Monitoring stations (general environment)	Measured concentrations	Calculated concentrations [a)]
Daishi, Kawasaki	5.19	0.32
KokusetsuKawasaki, Kawasaki	3.60	0.25
Nakahara, Nakahara	3.85	0.27
Tama, Tama	3.88	0.17
Average	4.13	0.25

a) Sources only in Kawasaki city were considered

The following three points can be considered as the causes of the underestimation of benzene concentration in comparison with NOx concentration. 1) Because benzene is more stable than NOx in atmosphere, an emission source far from receptor can affect the concentration. 2) The difference in the equations used for determining the dispersion width between the our model and the Total Control Model. 3) The total benzene emission reported in the PRTR Pilot Project is too low.

5 Discussion on the calculated result using the PRTR data

5.1 Difference in reactivity between benzene and NOx

The atmospheric half-life for benzene is reported to be 5.3 days, assuming an average hydroxyl radical concentration (WHO, 1993). The transformation of NOx depends on the strength of solar radiation. Typical transformation rates of NOx under Japanese weather conditions were reported to be from 0.6 %/h to 5.3 % /h (JEA, 1995), which correspond to 0.5 days to 4.5 days for the half-life if a 1st-order reaction is assumed. In our previous study (Kajihara et al., accepted), for the relationship between atmospheric concentrations of benzene and NOx measured in 1997, the following regression equation was obtained.

$$Y = 0.067 X + 0.91 \quad (1)$$

Here, X is the NOx concentration in ppb and Y is the benzene concentration in μg/ m^3. The Y-intercept of 0.91 μg/m^3 for this equation seems to reflect the fact that the half-life of benzene is longer than that of NOx. In addition, background concentrations were measured to be about

1 μg/ m^3 (JEA, 1998). On the other hand, the background concentration of NOx measured at general air pollution monitoring stations were reported to be 0-4ppb (JEA, 1995). Therefore, 1 μg/ m^3 in the measured average concentration of 4.13 μg/m^3 is considered to be the background concentration that cannot be reproduced by our model. The ratio of the calculated concentration of 0.25 μg/ m^3 to the value of 3.13 μg/ m^3 calculated by subtracting the background concentration from the measured concentration, was about 8%. However, this ratio is still considerably less than the 28% in the case of NOx. The underestimation of benzene concentration compared to NOx concentration could not be explained by only the difference between the reactivities of the two substances.

5.2 Discussion on dispersion parameters

The lateral and vertical standard deviations of concentration distributions in the dispersion equation, σ_y and σ_z, strongly affected the concentration calculated using the model. In order to derive σ_y and σ_z for an urban area, the Briggs equation (U.S. EPA, 1995) was used in our model, and the Pasquill-Gifford equation (P-G equation) (Kawasaki city, 1998; Tokyo Metropolitan Government, 1997) was used in the Total Control Model, as shown in Table 3. σ_y and σ_z derived using Briggs equation are larger than those derived using P-G equations under almost all conditions.

When σ_z increases, the pollutants emitted near the ground surface are more dilute and the pollutants emitted from higher sources, in contrast, are likely to reach the ground. In contrast, the total benzene emission was set to be from the height of 1.5m or 10m and 70 % of NOx emission was set to be emitted from 50m or 100m in Kawasaki city. In short, the source height of NOx is higher than that of benzene on the whole. Therefore, benzene concentration was considered to be estimated lower than NOx concentration when the Briggs equation was used as compared to when the P-G equation was used. As a result, the difference in the equation used to determine σ_z and the difference in the emission source heights are considered to be two causes of the underestimation of benzene concentration compared to NOx concentration. The evaluation of the degree of influence of the differences mentioned above will be considered in the future.

5. 3 Discussion on the PRTR emission data

As shown in Table 6, the contributions to the total benzene emission reported in the PRTR Pilot Project for Kawasaki city are 54% for factories and 34% for automobiles. These two account for the greater part of the total emission. Emission from factories in the Pilot Project was estimated based on the values reported by the factory owners. However only 55% of chemical manufacturers, whose emission of benzene accounts for 91% of the total emission from factories, responded to the survey.

The emission from automobiles was estimated by the Environment Agency in the Pilot Project. Total emission, emission factors (emission per trip) and traffic volume of each

automobile type reported in the Pilot Project are shown in Table 8. Values in parenthesis are those substituted by values for other automobile types, since there were no measured values. There are few reports on the benzene emission factor for automobiles other than gasoline passenger cars in Japan. Some reported emission factors for gasoline passenger cars are shown in Table 9. The emission factor of 0.6mg/km for gasoline passenger cars reported in the Pilot Project is several or ten times smaller than that measured in the 10・15 mode, at which the combustion condition of engines is mild.

Table 8 Emission of benzene from automobiles and traffic volume in Kawasaki city reported in the PRTR Pilot Project [a)]

Automobile types	Gasoline						Diesel				LPG	Two-wheelers
	Passenger cars	Light trucks	Lightweight trucks, buses	Mediumweight trucks, buses	Heavyweight trucks, buses		Passenger cars	Lightweight trucks, buses	Mediumweight trucks, buses	Heavyweight trucks, buses	Passenger cars	
Emission factors (mg/km)	0.62	21	(0.62)	2.1	7.4		1.0	(1.0)	4.3*	5.8	(0.62)	133.9
Traffic volume (million vehicle-km/yr)	1310[b)]	152	136	250	81		198	59	95	567	-[b)]	133
Emission (ton/yr)	0.81[b)]	3.2	0.08	0.53	0.60		0.20	0.06	0.41	3.3	-[b)]	17.8

Data in parenthesis were substituted values of other automobile types because of the lack of data. Data with * were obtained from only one experimental car. a) PRTR Pilot Project Report in fiscal year 1998 (JEA, 1999). b) Traffic volume and emission of LPG passenger cars were included in those of gasoline passenger cars.

In this study, the emission factor for gasoline passenger cars was assumed to be 4.5mg/km which was the average of the measured values in the 10・15 mode, as shown in Table 9. The amount of benzene emitted from automobiles is considered to be proportional to the total amount of emitted hydrocarbons, since benzene is a kind of hydrocarbon. For the total amount of hydrocarbons, emission factors have been measured for each automobile type and each driving condition, because total hydrocarbon is a target substance for regulation (Nomura Research Institute, 1998). In this study, the amount of benzene emission from all gasoline and LPG automobiles was estimated to be 56.4 ton/year under the assumption that benzene in the exhaust gas of gasoline and LPG automobiles is proportional to emitted hydrocarbons. For two-wheelers and diesel automobiles, the emission factors reported in the Pilot Project were used. Total benzene emission from all automobiles was estimated to be 78.1 ton/year, which was 2.9 times higher than the 27.0 ton reported in the Pilot Project.

It seems that the underestimated benzene emission in the PRTR Pilot Project may be one of the reasons for the underestimation of benzene concentrations compared to NOx concentrations.

6 Conclusions

Annual average concentration of NOx in Kawasaki city and Tokyo Metropolis was calculated by the modeling approach using the ISC as the framework. The ratio of the calculated concentration to the measured concentration is 28% for Kawasaki city and 72% for Tokyo Metropolis. It was shown that the contribution of sources outside Kawasaki city is large. It was also shown that the error between the values calculated using the Total Control Model and the model using ISC became 10% or less on average, and that the model using ISC is applicable for roughly estimating NOx concentration. As a result of the calculation using emission data of benzene reported in the the PRTR Pilot Project of Kawasaki city, the ratio of the calculated concentration to the measured concentration was 6%, 1/4 that of NOx (28%). As the major reasons for the underestimation of benzene concentration compared to NOx concentration, the following three points were considered: 1) effect of the difference between reactivities of NOx and benzene, 2) effect of dispersion parameters used in the dispersion equations and height of emission sources, 3) effect of the emission data reported in the PRTR Pilot Project.

Table 9 Benzene emission factors of gasoline passenger cars (experimental data)

(mg/km)

Report organization	displacement (cc)	Driving mode 10・15	11
Petroleum Council [a)]	1000-2000	6.0 (4)	50 (1)
JARI [b)]	2000	0.22 (1)	27.3 (1)
PEC [b)]	2000	2.71 (1)	13.2 (1)

The values in parenthesis are the number of experimental cars. All of the experimental cars are equipped with catalytic converters. Benzene content in gasoline was 2.3 Vol. % in all experiments. JARI: Japan Automobile Research Institute. PEC: Petroleum Energy Center, Advanced Technology and Research Institute. a) (Petroleum Council, 1996), b) (Natural Resources and Energy Agency, 1998)

7 Acknowledgements

This work was supported by CREST (Core Research for Evolutional Science and Technology) of Japan Science and Technology Corporation (JST).

8 References

Japan Environment Agency: About PRTR Pilot Project Report in fiscal year 1998 (1999 a) (in Japanese)

Japan Environment Agency Air Quality Bureau: Manual for total pollutant load control of nitrogen oxides (1995) (in Japanese)

Japan Environment Agency Air Quality Bureau: The results of hazardous air pollutants monitoring by local government in fiscal year 1997 (1998) (in Japanese)

Japan Environment Agency Environmental Health Department: PRTR Pilot Project Report in fiscal year 1998 (1999 b) (in Japanese)

Kajihara, H., Ishizuka, S., Fushimi, A., Masuda, A., Nakanishi, J.: Environmental Engineering and Policy

(accepted)
Kawasaki City: The report for the countermeasures hereafter of nitrogen oxides and suspended particulate matters in Kawasaki City (1998) (in Japanese)
Natural Resources and Energy Agency: (personal communication) (1998) (in Japanese)
Nomura Research Institute: Investigative report on primary unit and total emission of automobile exhaust gas (1998) (in Japanese)
Petroleum Council The Expert Committee for the Quality of the Petroleum Products: The report for the desirable quality hereafter of petroleum products (1996) (in Japanese)
Self-government association of Municipalities: Handbook of municipalities across the nation in fiscal year 1998 (1998) (in Japanese)
Tokyo Metropolitan Government: The basic investigative report on the countermeasures against reduction of nitrogen oxides (1997) (in Japanese)
Tokyo Metropolitan Government: Investigative report on total amount of traffic and emission of automobile exhaust gas (1996) (in Japanese)
U.S. EPA: Carcinogenic Effects of Benzene: An Update. Washington DC: Office of Research and Development, EPA/600/P-97/001F (1998)
U.S. EPA Office of Air Quality Planning and Standards: User's Guide for the Industrial Source Complex (ISC3) Dispersion Models, EPA-454/B-93-003a (1995)
World Health Organization: Environmental Health Criteria 150, Benzene (1993)

Appendix NOx emission in Tokyo Metropolis (fiscal 1995) [a)] (ton/yr)

City, Ward	Area (km^2) [b)]	Factories	Automo-biles [c)]	Vessels [d)]	Domestic [e)]	Aircrafts [f)]	Construction machines	Total
Chiyoda ward	11.64	216.1	1716.5		195.6		91.6	2219.8
Chuo ward	10.15	78.9	1195.6	247.0	303.9		334.2	2159.6
Minato ward	20.34	183.6	1677.0	247.0	462.2		388.5	2958.3
Shinjuku ward	18.23	158.2	1183.6		523.9		124.0	1989.7
Bunkyo ward	11.31	51.0	630.9		222.6		77.1	981.6
Taito ward	10.08	19.7	1049.5		299.7		85.7	1454.5
Sumida ward	13.75	68.6	2123.0		166.8		151.0	2509.3
Koto ward	39.24	833.4	1459.0	247.0	299.9		401.7	3241.0
Shinagawa ward	22.69	2376.1	1902.5	247.0	231.3		390.8	5147.7
Meguro ward	14.70	135.3	639.3		260.0		91.2	1125.8
Ota ward	59.46	605.6	2469.1	247.0	378.1	1827.0	375.5	5902.4
Setagaya ward	58.08	203.8	3193.3		501.9		323.0	4222.0
Shibuya ward	15.11	68.5	1480.7		374.2		100.0	2023.4
Nakano ward	15.59	14.9	694.1		249.6		89.8	1048.4
Suginami ward	34.02	125.7	1045.1		289.7		150.2	1610.8
Toshima ward	13.01	46.9	634.7		48.9		82.2	812.7
Kita ward	20.59	58.8	674.7		68.4		191.9	993.7
Arakawa ward	10.20	14.7	435.8		124.3		73.5	648.3
Itabashi ward	32.17	557.9	2445.7		76.2		151.6	3231.4
Nerima ward	48.16	173.0	1799.9		130.1		224.1	2327.0
Adachi ward	53.20	426.7	2393.5		127.1		343.1	3290.5
Katsushika ward	34.84	515.4	1474.0		196.0		188.1	2373.6
Edogawa ward	49.86	190.5	2462.2	247.0	159.3		409.0	3468.0
Hachioji city	186.31	241.5	1969.1		145.5		324.8	2680.9
Tachikawa city	24.38	86.8	365.9		38.9		95.0	586.6
Musashino city	10.73	57.6	232.4		297.6		76.3	663.9
Mitaka city	16.50	55.1	323.4		428.6		69.7	876.8
Oume city	103.26	24.5	504.0		154.1		143.0	825.6
Fuchu city	29.34	108.6	898.5		46.3		173.6	1227.0
Akishima city	17.33	177.4	326.3		110.1		55.6	669.4
Chofu city	21.53	162.0	571.9		136.9		107.2	978.1
Machida city	71.64	153.4	696.7		164.5		191.8	1206.4
Koganei city	11.33	4.0	209.4		141.0		53.3	407.8
Kodaira city	20.46	170.9	304.2		116.5		71.7	663.3
Hino city	27.53	428.1	645.9		46.3		93.2	1213.5
Higashimurayama city	17.17	63.6	313.0		63.8		70.7	511.2
Kokubunji city	11.48	36.1	142.2		138.0		66.6	382.9
Kunitachi city	8.15	32.6	262.7		37.2		30.3	362.8
Tanashi city	6.80	13.7	152.8		267.6		41.3	475.3
Hoya city	9.05	3.2	175.2		230.9		49.0	458.2
Fussa city	10.24	2.7	146.1		56.9		38.6	244.3
Komae city	6.39	4.7	105.8		171.3		44.6	326.4
Higashiyamato city	13.54	18.5	103.0		28.8		45.2	195.4
Kiyose city	10.19	11.7	115.0		308.0		31.4	466.1
Higashikurume city	12.92	120.6	172.9		242.0		48.3	583.9
Musashimurayama city	15.37	70.6	262.6		60.5		36.4	430.1
Tama city	21.08	78.4	399.6		99.3		60.9	638.2
Inagi city	17.97	111.6	152.1		56.7		33.2	353.5
Hamura city	9.91	253.9	157.7		54.8		36.3	502.7
Akiruno city	73.34	60.1	146.2		102.6		109.5	418.4
Mizuho city	16.83	127.5	250.4		2.0		27.6	407.5
Hinode town	28.08	22.6	62.9		0.3		35.4	121.2
Hinohara village	105.42	0.2	4.8		0.3		60.1	65.3
Okutama town	225.63	69.3	46.6		0.0		89.9	205.8
Total	1776.32	9894.9	44999.0	1482.0	9437.0	1827.0	7248.0	74887.9

a) Tokyo Metropolitan Government, 1997. b) Self-government association of municipalities, 1998. c) The total emission from automobiles (Tokyo Metropolitan Government, 1997) was divided into each ward using the ratio written in the report in fiscal 1994 (Tokyo Metropolitan Government, 1996). d) The total emission from vessels was divided equally into six coastal wards (Edogawa, Koto, Chuo, Minato, Shinagawa, Ota). e) The total emission from domestic was divided into each ward according to the ratio of GHP (Gas Heat Pump) (Tokyo Metropolitan Government, 1997). f) The total emission from aircraft was grouped into Ota ward where Tokyo International Airport lies in.