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Emission inventory of VOCs from mobile sources in a metropolitan region

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Abstract−Based on methodologies developed by US EPA, European EMEP/CORINAIR, and Australian NPI, and the former emission inventory in Korea, two methods were applied to 151 villages in northeastern Seoul, Korea to estimate emission of VOCs from line and area vehicle sources depending on vehicle types with different fuel types. A discharge coefficient method for the line source on the Eastern main road was calculated by multiplying the emission amounts per unit of mileage, and a fuel exhaust coefficient method for the area vehicle sources on other roads was determined as multiplying the emission rates by the actual consumption of excess fuel. Results indicated the methods could be adequate for estimating the amounts of mobile emissions when limited information on mobile emission is available. The methods can be used to develop the emission model for all VOCs emission sources (point and non-point sources), which provides input data of atmospheric models.

Key words: Mobile, Vehicle, Emission, Inventory, Metropolis, Road, Toluene, VOCs, Benzene, Xylene, Styrene

INTRODUCTION

Air pollution due to O_3 precursors, volatile organic compounds (VOCs) and NO_x, has increased, primarily as the result of the rapid increase in number of automobiles. Mobile sources are of concern in terms of developing strategies for reducing ground level ozone across the world. Aardenne et al. [1999] reported data for NO_x emissions in Asia during the period 1990-2020 due to anthropogenic activity based on the RAINS-ASIA methodology. The rapid growth of NO_x emission was directly related to the dynamic nature of energy use between road transport and non-road transport in Asia. In 1990 NO_x emissions due to transport made up 80-65% of the total emission in Bangkok, Delhi, Jakarta, Manila, and Seoul.

It has been known that mobile emissions are also a major source of VOCs in urban areas away from industrial sources [Chan et al., 2002]. Chan et al. [2003] reported on the characteristics and concentrations of VOCs in roadside microenvironments in metropolitan Hong Kong and compared their data with the most developed cities in China, Germany, UK, Greece, Canada, and Brazil. Hong Kong was dominated by aromatic VOCs and VOC concentrations, especially toluene and, to a lesser extent, benzene. Chlorinated VOCs in Hong Kong were a little higher than those in other cities. Tsai et al. [2003] and Bong et al. [2003] reported that isopentane, toluene, m,p-xylene, n-pentane, 2-methylpentane, 3-methylpentane, benzene, n-heptane, and methylheptane were the major VOC components in motorcycle engine exhaust. Singer and Harley showed that fuel use estimates and emission factors measured from over 60,000 on-road vehicles could be used to calculate stabilized exhaust emissions of 550±90 metric tons day^{-1} of VOC in California's South coast Air Basin during the summer of 1997. These values were higher than official inventory estimates for the same period by factors 3.5±0.6 for VOC. Meanwhile, the high degree of uncertainty and a severe underestimation of a VOC emission inventory reported by the National Research Council [NRC, 1992] may be partially responsible for the limited success of VOC control strategies across the U.S. during the last two decades in solving ground level ozone problems [McLaren and Singleton, 1996].

To address these questions and to estimate the amounts of emitted VOCs from vehicles, a new emission inventory technique was adopted on the basis of two parameters, a fuel exhaust coefficient and a discharge coefficient of the distance covered in a given time (mileage). The present study is a part of an integrated environmental management (IEM) project supported by the Korean Ministry of Environment and ECO-Technopia-21 (Core Environment Technology Development Project for Next Generation) during the period 2001-2004. The new EI technique was applied for the northeast of Seoul, over all of the Jung-Rang stream, in Korea. Based on the previous studies cited above and the priority substance list developed in the first step of the IEM project [Kim et al., 2005; Chah et al., 2003; Yi, 2002], Benzene, Toluene, Xylene, and Styrene (BTXS) were selected as the major aromatic compounds in the present study.

1. Methodology

During the fuel combustion process of road vehicles, amounts of air pollutants, emitted into the air, depend on the type of vehicle, speed, fuel use, etc. In order to estimate emissions of mobile sources on two different road types, the main road and other roads, this study used two different methodologies developed on the basis of the US EPA, European EMEP/CORINAIR, Australian NPI, and the previous development of an emission inventory in Korea. Two major methods, in estimating the amounts of pollutants emitted from a vehicle, are the fuel exhaust coefficient and the discharge coefficient of the distance covered in a given time (mileage). The former is determined as multiplying the emission rates (per unit fuel use)

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by the actual consumption of excess fuel, while the latter is calculated by multiplying the emission amounts per unit of mileage (unit distance covered) by the actual distance covered (mileage).

1-1. Discharge Coefficient Method

The discharge coefficient method uses the actual distance covered (mileage) to estimate the emission amounts of the pollutants when it is impossible to determine the amount of the fuel consumption by the limited mileages over the target region. Based on current traffic volume data, the multiplication of the average traffic volume and the road extension distance (RL) within the target block determines the annual total mileage of the vehicle model within the target area. Meanwhile, the mileage of the vehicle model is determined by dividing the total mileages by the current registered vehicles. Because emission coefficient data for each VOC for the total vehicle models are not currently available, the present study applies the European emission coefficient for each VOC. The following equation is used to determine the VOC emission rate (Kg yr^{-1}), E;

$$E=\frac{1}{1000}\times\sum_{i}\left(\mathrm{TV}\times\frac{\mathrm{VN}_i}{\mathrm{tVN}}\times\mathrm{RL}\times\mathrm{EF}_i\times\mathrm{R}\right) \qquad (1)$$

where, i is the vehicle model, TV is the annual traffic volume (vehicles year^{-1}), VN_i is the number of registered vehicles by the vehicle model, tVN is total registered vehicles, RL is the road extension (Km vehicle^{-1}), EF_i is the VOC emission coefficient due to the vehicle model (g km^{-1}), and R is the mass composition ratio of the VOCs. Note that EF_i is estimated by the average speed within the block as a function of vehicle speed.

1-2. Fuel-exhaust-coefficient Method

Using registered vehicles due to the vehicle model related to the fuel type and their fuel consumption, the fuel-exhaust-coefficient method estimates the mobile emission rates (kg yr^{-1}) depending on the fuel consumption as follows:

$$E=1000\times\sum_{i}\left(\mathrm{DVN}_i\times\frac{\mathrm{VN}_i}{\mathrm{tVN}_i}\times\mathrm{F}_i\times\mathrm{d}_i\times\mathrm{R}\right) \qquad (2)$$

where, i is the vehicle model related to the fuel type and business use, DVN_i is the registered vehicles for the vehicle model for each village in the target area, VN_i is the registered vehicles for the vehicle model in the target area, tVN_i is the total registered vehicles in the target area, F_i is the annual fuel consumption (kl vehichle^{-1} yr^{-1}), d_i is the fuel density (kg L^{-1}), and R is the mass composition ratio of VOCs (kg kg^{-1}). All information for this method was precisely gathered for each village.

2. Application of Emission Inventory Technique

All 151 villages (so called "Dong"), over the Jung-Rang stream located in northeastern Seoul, were selected for developing the emission inventory of a metropolitan region. The Jung-Rang stream is the longest stream (45.3 km) in Seoul and flows into the Han River. Fig. 1 shows the 3-dimensional topography of the target area for this study. The target area has the characteristics of a local environment causing air pollution by non-point emission sources (line and area sources) as well as by point sources such as incineration. The road and the traffic conditions in this area are a representation of metropolitan Seoul. In Seoul, traffic activity, vehicles and traffic volume have greatly increased, and road traffic volumes continue to increase due to the availability of low cost fuel vehicles. Registered LPG vehicles have increased from 126,948 in 1998 to 228,996 in 2000 [data, Seoul]. The traffic volumes of LPG vehicles are about 22.7% for private automobiles.

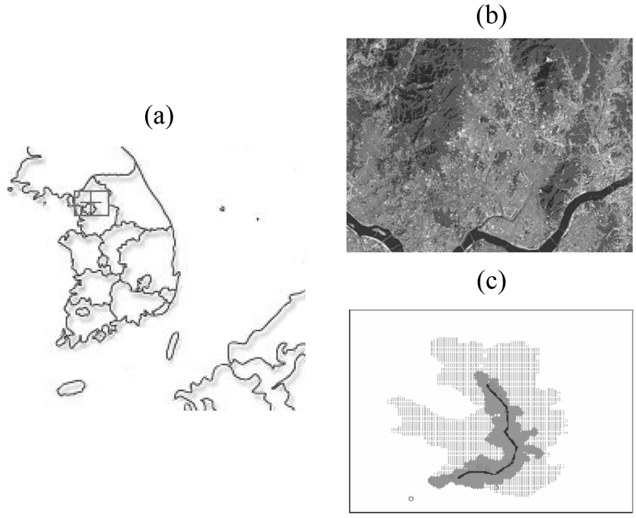


Fig. 1. The target area map: (a) a map of South Korea, (b) a picture of Northeastern Seoul taken by Satellite, and (c) locations of the Eastern Main road (solid line), other roads in the 34 districts of the seven counties (■), VOCs measurement sites (○), and the target area, seven counties, of the present study (▦).

The mobile sources in the target area include road mobile emission and rail-road mobile emission. In the case of a railroad vehicle, hazardous compounds are emitted from the fuel combustion during operation. The railroads include the Kyong-Won Track, the Kyong-Choon Track, the Jung-Ahng Track, and the Mang-U Track distributed along Jung-Rang stream and aggregate from the Cheong-Ryang-Ri via the Hui-Kyong. However, the amounts of railroad emissions are comparatively much lower than those from road mobiles. On that account, the focus of the present study was on road mobile sources, which were divided into two parts: the Eastern main road sources and other road sources as shown in Fig. 1. The Eastern main road is a representative road in the area and runs from Yong-Bi Gyo (bridge) to Nok-Cheon Gyo along the Jung-Rang stream. Other roads are located in seven districts (SungDong-Gu, GwangJin-Gu, DongDaeMoon-Gu, JungRang-Gu, SungBook-Gu, DoBong-Gu, and No-Won-Gu) including 34 villages (among all 151 villages in the seven districts) along the Eastern main road over the Jung-Rang stream.

RESULT AND DISCUSSION

The Eastern main road (through 25 villages) was chosen and considered as a line source for the emission inventory of mobiles on the road over the Jung-Rang stream. For the main-road source, the discharge coefficient method of the distance in Eq. (1) was applied to determine the emission rate of VOCs because fuel consumption or traffic volume in the target area was not known with certainty. Table 1 shows the information required in Eq. (1) to determine the discharge coefficient as a function of vehicles' category; the vehicles include passenger cars (PC), duty-cars (DC), and duty-trucks (DT) with three different size types (light, medium, and heavy) and with three different fuel types (gasoline, diesel, and LPG). Infor-

Table 1. Emission factors for calculating emission rates according to the two methods given in Eqs. (1) and (2)

Categories of vehicles			Vehicle registration in Seoul§	Fraction of vehicles registration in Seoul	Emission coefficient of total VOC (g/km)*	Mass composition of VOCs kg/kg
Passenger car	Gasoline		3,493,589	0.833	$19.079V^{-0.693}=1.44$	0.00616
	Diesel				$4.61V^{-0.937}$	0.00258
	LPG		103,431	0.025	$26.3V^{-0.865}=1.05$	0.00959
Duty-car	Light	Gasoline	9,532	0.002	$19.079V^{-0.693}=1.44$	0.00616
		Diesel	181,664	0.043	$4.61V^{-0.937}=0.14$	0.00303
		LPG	63,195	0.015	$26.3V^{-0.865}=1.05$	0.00877
	Medium	Gasoline			$0.000677V^2-0.1170V+5.4734$	0.00616
		Diesel	7,534	0.002	$0.000066V^2-0.0113V+0.6024=0.25$	0.00303
	Heavy	Gasoline				0.00616
		Diesel	12,510	0.003	$43.647V^{-1.0301}=0.94$	0.00303
Duty-truck	Light	Gasoline	11,214	0.003	$0.000677V^2-0.1170V+5.4734=1.79$	0.00616
		Diesel	237,798	0.057	$0.000066V^2-0.0113V+0.6024=0.25$	0.00258
		LPG	12,967	0.003	$26.3V^{-0.865}=1.05$	0.00841
	Medium	Gasoline			7	0.00616
		Diesel	34,006	0.008	$40.120V^{-0.8774}=1.53$	0.00258
	Heavy	Gasoline			7	0.00616
		Diesel	27,003	0.006	$40.120V^{-0.8774}=1.53$	0.00258
			4,194,443			

§data from The Ministry of Construction and Transportation [1999].

*European EMEP/CORINAIR data: V is an average speed (km/hr) given the district, V=41.46 km/hr in the East main road in the year 2000.

Table 2. Ratio of VOC mass composition ($\times10^{-2}$) (Europe EMEP/CORINAIR data)

Chemicals	Gasoline	Diesel	LPG	Chemicals	Gasoline	Diesel	LPG
Ethane	1.8	1	3	1-Hexene	0.4		
Propane	1	1	44	1,3 Hexene	0.4		
n-Butane	5.5	2		Alkanes C>7	0.2	2 (1)	
i-Butane	1.5			Benzene	3.5	2	
n-Pentane	3.2	2		Toluene	7	1.5	
i-Pentane	7			o-Xylene	2	0.5	
Hexane	6			m,p-Xylene	4	1.5	
Heptane	5			Ethylbenzene	1.5	0.5	
Octane	7			Styrene	0.5		0.1
Nonane	2			1,2,3-Trimethylbenzene	1		
Alkanes C>10	3	30 (1)		1,2,4-Trimethylbenzene	4		
Ethylene	7	12	15	1,3,5-Trimethylbenzene	2		
Acetylene	4.5	4	22	Other aromatics C9	3		
Propylene	2.5	3	10	Aromatics C>10	6	20 (1)	
Propadiene				Formaldehyde	1.1	6	4
Methylacetylene	0.2			Acetaldehyde	0.5	2	2
1-Butene	1.5			Other Aldehydes	0.2	1.5	
1,3 Butadiene	0.5	2		Acrolein	0.2	1.5	
2-Butene	0.5			2-Butenal		1	
1-Pentene	0.5			Benzaldehyde	0.3	0.5	
2-Pentene	1	1		Acetone	1	1.5	
					100	100	100

mation concerning vehicles was adopted from the Ministry of Construction and Transportation in Seoul [1999]. Using the European EMEP/CORINAIR data, the emission coefficients for total VOC (g km^{-1}) were determined as a function of average speed V for the main-road zone, which was 41.46 km hr^{-1} for the East main road in the year 2000. The ratios of VOC mass compositions in Table 2 include BTXS values (Benzene, Toluene, Xylene, and Styrene).

The fuel-exhaust-coefficient method in Eq. (2), for estimating

Table 3. Vehicles registered in each county over the target area

	Passenger car			Duty-car			Duty-truck		
	Gasoline	Diesel	LPG	Gasoline	Diesel	LPG	Gasoline	Diesel	LPG
Sung-Dong	22432.73	912.3	1582.98	91.37	2320.11	1735.52	135.72	5955.15	615.13
Kwang-Jin	8249.39	335.49	582.13	36.31	922	689.68	43.65	1915.48	197.86
Dong-Dae-Moon	14294.02	581.32	1008.66	62.44	1585.53	1186.02	77.14	3385.18	349.67
Jung-Rang	18762.02	763.02	1323.95	91.35	2319.55	1735.1	120.48	5286.46	546.06
Sung-Book	4009.06	163.04	282.9	17.85	453.17	338.99	22.26	976.84	100.9
Do-Bong	5492.1	223.35	387.56	18.95	481.14	359.91	10.87	476.87	49.26
No-Won	20672.52	840.72	1458.76	71.39	1812.68	1355.94	61.04	2678.31	276.65
All	93911.84	3819.24	6626.94	389.66	9894.18	7401.16	471.16	20674.29	2135.53

Table 4. Annual Fuel consumption (k*l* vehicle^{-1}): Ministry of Construction and Transportation in Seoul [1998]

Passenger car					
Gasoline		Diesel		LPG	
Non-business	Business	Non-business	Business	Non-business	Business
1.39	11.4	1.39	11.4	1.49	11.4
Duty-car					
Gasoline		Diesel		LPG	
Non-business	Business	Non-business	Business	Non-business	Business
4.68	37.92	4.68	37.92	4.68	37.92
Duty-truck					
Gasoline		Diesel		LPG	
Non-business	Business	Non-business	Business	Non-business	Business
2.54	12.66	2.54	12.66	2.54	12.66

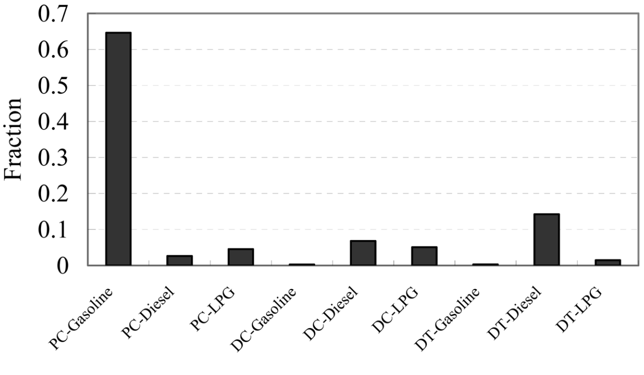
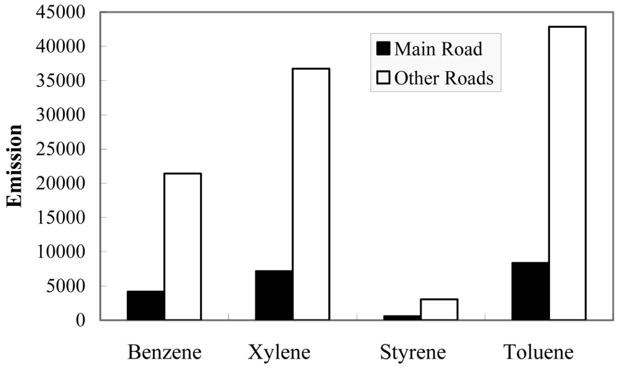


Fig. 2. Registered vehicles related to fuel types in selected counties near Jung-Rang stream.

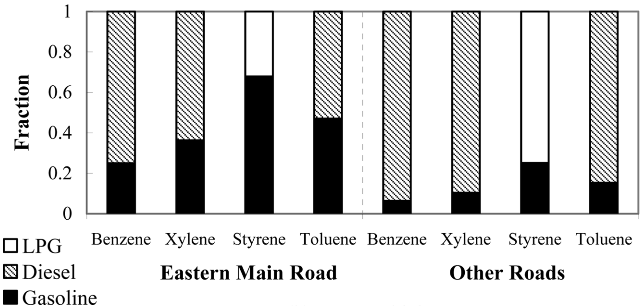
the mobile emission from other roads over Jung-Rang stream except the Eastern main road, was applied as a function of emission factors such as the number of registered vehicles, annual fuel consumption, and so on. Data for registered vehicles and the annual fuel consumption in the selected seven districts were obtained by using the Seoul data, as reported by the Ministry of Construction and Transportation [1998] and those data are given in Tables 3 and 4. The vehicles, as given in Table 4, can be classified due to vehicle model related to the fuel type. Most vehicles are gasoline powered PC which makes up almost 70%, while the diesel powered DVs comprise 20% of the total vehicles over the target area (see Fig. 2). To calculate the emission rates on other roads, we used the registered vehicles and the annual fuel consumption as shown in Tables 3 and 4, respectively. The annual fuel consumption was obtained by multiplying the fuel densities (734.5 g L^{-1} for gasoline, 839.8 for diesel, and 578.0 for LPG), and the ratio of VOCs mass compositions shown in Table 2.

Based on data from Table 1 to Table 4, Fig. 3 shows the emission estimates of major VOCs, BTXS, emitted from mobile sources on the Eastern main road (the line source) and on other roads (the area sources). On the Eastern main road, PC makes up more than 80% of the total vehicles and the emission rates from gasoline powered light-PC are dominant factors. Fig. 3(a) shows values for BTXS emitted from all gasoline-powered PC, and are in the order; toluene, xylene, benzene, and styrene. The emission rates for toluene are over 8 T yr^{-1}, xylene emissions are over 7 T yr^{-1}, those for benzene are about 4 T yr^{-1}, and emitted styrene is very low. On other roads, the emissions from gasoline powered PCs are almost 87% of the total emissions and the BTXS from the gasoline powered PCs are ordered as shown in Fig. 3(a). When comparing the results with those from the Eastern main road, the BTXS values are in the same order but the amounts of material emitted are five times greater.

In Fig. 3(b), BTXS emission rates from Duty-Vehicles (Duty-Car and Duty-Truck) are illustrated according to fuel type. The gasoline-powered DV has the same order of BTXS emission as a gas-



(a) Gasoline powered PC



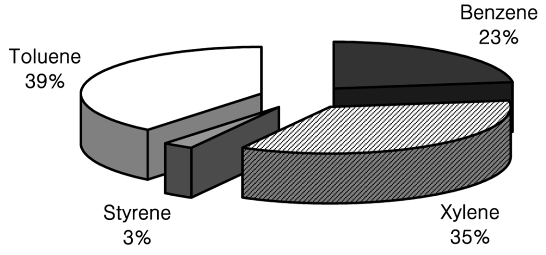
(b) Duty-Vehicle

Fig. 3. VOC emissions estimation due to mobile sources on other roads over all the target area using fuel exhaust coefficient as a function of vehicle model and fuel type (a) Emission rate (kg yr^{-1}) from Gasoline powered Passenger Cars and (b) fraction of emission rates from Duty-Vehicle.

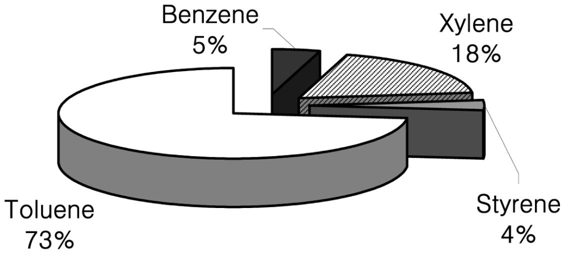
oline-powered PC, but the amounts of pollutants emitted are very low compared to a gasoline-powered PC. For the case of the Eastern main road, benzene and xylene are the main components due to diesel-powered DV, while styrene levels are largely dependent on gasoline and less so, about 30%, due to LPG. Toluene is contributed equally by gasoline-powered DV and diesel-powered DV. Fig. 3(b) also shows that the fraction of emission by diesel-powered DV on other roads is 80-90% of the total DV, and this value can be compared to the 60-80% for the Eastern main road sources.

The total mobile emissions in the target area were obtained by summing the Eastern main road vehicle emissions and other-road vehicle emissions. For both emissions rates from the Eastern main road sources and other road sources, toluene is the dominant compound, almost equivalent to xylene, following benzene, and styrene, in that order. The amounts of pollutants emitted from other road sources are five times greater when compared to those from the Eastern main road mobile sources; the BTXS emission from the Eastern main road sources is approximately 14% of the total mobile sources, and about 86% of the total mobile sources are caused by other road emission sources.

Fig. 4 shows the fraction difference between BTXS mobile emissions and the fraction of BTXS concentration. BYXS were measured under ambient conditions during the project period 2002 from 10 measurement sites located within the target area, as shown in Fig. 1, and these BTXS concentrations were used to estimate the distribution of VOCs concentrations. Fig. 4(a) indicates that the fraction of toluene emitted is 39% of the total BTXS emission, while



(a) Mobile Emission



(b) Ambient Air Concentration

Fig. 4. Comparison of BTXS between emission estimation and ambient concentration measured during the 2002 from the sites marked in Fig. 1(c).

the fraction of ambient toluene concentration is over 70% of the total ambient BTXS concentrations in Fig. 4(b).

1. Summary and Conclusion

Two emission inventory (EI) techniques were used to estimate the amounts of pollutant VOCs emitted from vehicles as a part of an integrated environment management (IEM) project supported by the Ministry of Environment in Korea and ECO-Technopia-21 (Core Environment Technology Development Project for Next Generation). These methodologies for the mobile emission inventory were developed on the basis of the US EPA, European EMEP/ CORINAIR, Australian NPI, and using the emission inventory developed in Korea. The EI techniques were also applied to the northern parts of Seoul, over the Jung-Rang stream. Aromatic compounds, benzene, toluene, xylene, and styrene (BTXS) were chosen as the major VOCs in an urban atmosphere based on the literature.

The two methods for estimating the amounts emitted from mobile sources are the fuel exhaust coefficient and the discharge coefficient of the distance covered in a given time (mileage). The former is determined by multiplying the emission rates (per unit fuel use) by the actual excess fuel consumption, and its application is carried out for VOCs emitted from mobiles on other roads (roads except the Main-Road) in seven counties with 34 districts over Jung-Rang stream, while the latter is calculated by multiplying the amounts emitted per unit mileage (unit distance covered) by the actual distance covered (mileage) and applied to the Eastern main-road emission from mobile sources. The EI techniques depend on the vehicle type [passenger car (PC), duty-car (DC), and duty-truck (DT)] related to fuel type (gasoline, diesel, and LPG). In the present study, PCs make up more than 80% of the total vehicles and the gasoline powered PCs are almost 70% of the total vehicles over the target area, while the diesel powered DVs comprise 20% of the total vehicles over the target area. The VOCs emissions by the gasoline powered PC are dominant. The total mobile emissions of the target area as obtained by the two methods indicate that mostly toluene, almost equivalent to xylene, followed by benzene, and styrene, in that order. The amounts emitted from other road sources are five times greater when compared to those from the Eastern main road mobile sources; the BTXS emission from the Eastern main road sources is approximately 14% of the total mobile sources, and about 86% of the total mobile sources are caused by other road emission sources. The present study estimates that mobile emission is mainly dependent on fuel consumption and the numbers of the vehicles.

This study shows that the mobile emission is clearly the major source of VOC in urban areas away from industrial sources and that the two methods applied in this study are possible for estimating mobile emissions from the line and/or area sources. For further study, real world vehicular estimation factors (EF) are required for the accurate vehicular emission estimation of non-methane hydrocarbons (NMHC) such as Na et al. [2002] in Seoul. When comparing the mobile emission and ambient concentrations obtained during the project period, the mobile emission does not explain the ambient concentration. In other words, the ambient concentration of toluene is significantly affected by other sources as well as the mobile sources. To explain the uncertainty or the accuracy of the VOCs emission inventory requires considering not only the mobile sources but also all other sources including point and non-point emission sources. It is important to estimate emissions accurately because it provides detailed data of source information, helps to understand the current situation of the air pollution, and reduces the uncertainty of model prediction.

NOMENCLATURE

d_i : the fuel density [kg L^{-1}]
DVN_i : the registered vehicles due to the vehicle model for each district in the target area
E : the VOC emission rate [kg/year]
EF_i : the VOC emission coefficient due to the vehicle model [g km^{-1}]
F_i : the annual fuel consumption [kl vehichle^{-1} yr^{-1}]
i : the vehicle model related to the fuel type and business use
R : the mass composition ratio of VOCs kg kg^{-1}
RL : the road extension [km vehicle^{-1}]
TV : the annual traffic volume [vehicles year^{-1}]
tVN_i(=tVN) : the total registered vehicles in the target area
VN_i(=VN) : the number of registered vehicles due to the vehicle model in the target area

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