Screening technique to estimate high benzo[a]pyrene concentrations near roads

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Abstract: The emission factors of benzo[a]pyrene for gasoline cars and for diesel cars were measured by directly collecting the exhaust gas using a portable sampling equipment and became 660 ng km-1 and 1700 ng km-1, respectively. The regression equations to easily evaluate the concentrations at the roadsides and at the crossroads were introduced as the functions of building height, road width, and wind speed from CFD simulations. The estimated concentrations showed that there were almost locations with the high concentrations exceeding the screening level to the probability of 10-5 that a person has a cancer during his life.

Keywords: benzo[a]pyrene; emission factor; screening level; regression equation; CFD; computational fluid dynamics.

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1 Introduction

Exhaust gas emitted from cars is one of the main causes of air pollution in urban environments. Until 1980, the target air pollutants of exhaust gas were nitrogen oxides, sulphur oxides, carbon monoxide and suspended particulate matter. In Japan, emission control of gasoline cars and diesel cars has been enforced every year. The emission factor for nitrogen dioxides in 2010 decreased to 1% of the 1970s emission factor (Ministry of Environment, 2008). Sulphur content in diesel fuel also decreased to less than 1% of the 1970s sulphur content (Ministry of Environment, 2008). After 1990, the target air pollutants of exhaust gas shifted to hasardous chemical substances, especially BTEX (benzene, toluene, ethyl benzene, and xylene) and PAHs (Polycyclic Aromatic Hydrocarbons) with carcinogenicity (Pope, 2000; Kan et al., 2007; Ravindra et al., 2008). In 1995, the atmospheric standard of benzene was determined in Japan (Ministry of Environment 2008). Countermeasures to reduce the amount of benzene added in gasoline were enforced, and benzene concentrations in a lot of observatories stopped exceeding atmospheric standards. However, there are very few countermeasures for benzo[a]pyrene, which is one of the PAHs, and is classified as Group 1 (The agent is carcinogenic to humans) by IARC (International Agency for Research on Cancer) (1989).

Researches on emissions of these substances from cars have been carried out worldwide. The main approaches to determine the emission factor are measurements in a tunnel (Geller et al., 2005; Phuleria et al., 2006) and experiments with a chassis dynamometer system (Zielinska et al., 2004; Riddle et al., 2007). However, it is known well that the emission factors from cars running on actual roads are different from these emission factors (Yang et al., 2005). Their emission factors strongly depend on fuel type, engine type, road conditions and driving conditions. Therefore, the measured emission factors of benzo[a]pyrene widely varied from a few tens of ng km^{-1} to a few thousands of ng km^{-1} (Bergvall and Westerholm, 2009; Karavalakis et al., 2009). In this study, the emission factors at actual conditions including sudden acceleration, quick stops and idling states were estimated by using portable sampling equipment, which directly collected the exhaust gas.

Many researches (Ravindra et al., 2006; Marr et al., 2006; Saarnio et al., 2008; Phuleriaa et al., 2007) on PAHs measurement reported that emissions from exhaust gas were the largest contribution in urban areas. The high air pollutant concentration locally occurs due to heavy traffic or the geometric structure of buildings and of roads. The limited air pollution monitoring network cannot perfectly cover all locations with high concentrations. To conserve the air quality in urban areas, it is necessary to easily identify the locations of high concentration. In this study, the regression equations to simply estimate the concentration at roadsides and crossroads were obtained from CFD simulations by varying the parameters of building height, road width, wind speed and wind direction. Using the emission factors obtained from the portable sampling equipment, the regression equations, and the traffic volume, benzo[a]pyrene concentration at the roadsides and at the crossroads in Osaka City was evaluated.

2 Measurement of emission factor

2.1 Sampling equipment

The outline of the portable sampling equipment (Shi et al., 2009) is shown in Figure 1. The sampling head is inserted into the muffler of a car and is fixed. The exhaust gas is sampled at a constant flow rate maintained by a pump installed in the car and is collected by the absorbers filling up the sampling tube. The absorbers used are TenaxTA20/35 for collecting PAHs. Each substance is analysed by the GC-MS (Simazu-QP2010) with Thermal Desorbor (Perkin Elmer-Turbo Matrix ATD). The emission factor EF [g km^{-1}] of each substance is determined by equation (1).

$$c = \frac{m}{Q}, \quad EF = \frac{cV\Omega t}{2\mathrm{L}}, \tag{1}$$

where c is a substance concentration into exhaust gas [gm^{-3}], m is a substance weight into sampling tube [g], Q is a sampling volume [m^3], V is a displacement volume [m^3], Ω is the number of rotations [min^{-1}], t is sampling time [min] and L is a mileage [km].

Figure 1 Outline of the portable sampling equipment (see online version for colours)

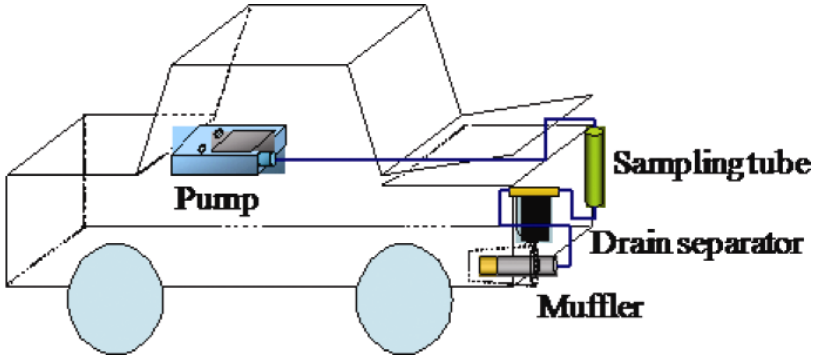


Table 1 Specification of cars

	Manufacture	*Age*	*Displacement* [L]	*Weight* [kg]	*Mileage* [km]
1 Diesel car	T	2001	4.61	2330	149,400
2 Diesel car	M	2004	1.99	2345	65,000
3 Diesel car	T	2005	4.00	4455	63,600
4 Diesel car	T	2007	4.00	4394	34,800
5 Diesel car	T	2008	2.98	1860	25,800
6 Gasoline car	M	2003	1.83	1585	87,900
7 Gasoline car	H	2006	1.49	1170	48,000
8 Gasoline car	N	2008	1.49	1395	28,800

Table 2 Emission factor of PAHs [ng/km]

	Diesel car				*Gasoline car*			
	1	*2*	*3*	*4*	*5*	*6*	*7*	*8*
Naphthalene	2.8e5	4.7e5	6.9e4	2.8e4	3.3e3	5.0e4	1.8e4	1.7e3
Acenaphthylene	8.0e3	1.8e3	4.5e2	3.4e2	3.4e2	3.2e2	2.3e2	1.5e1
Acenaphthene	3.2e3	1.5e3	5.4e2	4.5e3	2.8e2	1.5e2	9.6e1	6.7e1
Fluorene	1.1e4	1.7e3	1.5e3	5.3e3	6.5e1	1.8e2	1.9e2	2.1e1
Phenanthrene	1.3e4	1.3e3	3.7e3	4.1e3	5.4e2	2.6e2	1.3e2	1.4e2
Anthracene	2.0e3	3.2e2	4.9e2	2.1e2	8.5e1	2.2e1	2.0e2	1.2e2
Fluoranthene	6.2e3	5.6e3	2.3e2	5.5e3	3.8e2	3.8e1	6.1e1	8.0e1
Pyrene	1.2e4	1.1e4	3.2e2	9.8e3	5.5e2	1.4e2	9.5e1	8.9e1
Benzo[a]Anthracen	5.9e2	2.1e2	6.0	1.7e1	3.0	2.7e1	5.0e1	5.7
Chrysene	2.9e2	2.0e2	7.0	1.4e1	2.0	4.3	5.3	3.7
Benzo[b]Fluoranthene	2.9e3	4.1e2	2.3e1	1.5e2	1.5e1	3.2	3.9	2.7
Benzo[k]Fluoranthene	1.8e3	3.4e2	3.7e1	1.3e2	1.1e1	3.1e1	-	1.9e1
Benzo[a]pyrene	5.2e3	3.3e2	1.4e2	2.8e2	4.2e1	2.2e2	3.2e2	3.1e1
Indeno[1,2,3-d]pyrene	1.5e2	-	3.7e1	7.0	7.0	-	-	-
Dibenzo[a,h]Fnthracene	4.0e2	-	1.1e2	3.9e1	4.8e1	-	-	-
Benzo[g,h,i]Perylene	6.2e2	-	3.8e1	1.2e1	1.5e1	-	-	-

2.2 Emission factor

The samplings from three gasoline cars and five diesel cars were carried out in October and November 2009. The sampling frequencies were four times for a diesel car and 2 times for a gasoline car. Sixteen chemical species of PAHs were analysed and their emission factors were calculated from equation (1). Though the running condition strongly affected the emission factors, the average emission factors were adopted. The specification of the used diesel cars and the gasoline cars is shown in Table 1. The average emission factors of PAHs were summarised in Table 2.

It is well-known that gasoline cars emit plenty of hydrocarbons in cold starts (Devos et al., 2006). Assuming a cold start until the mileage of 4 km, the emission factor in hot start was estimated. Shi et al. (2009) reported the relation between emission factors in hot start and manufacture year and emissions in cold start. Based on this report, the average PAHs emission factor from the gasoline cars was corrected.

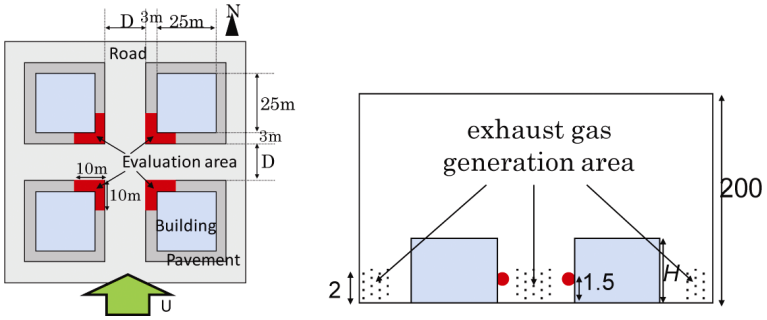
The emission factor of diesel cars hardly changed in cold start. Osaka City (the target area) has a regulation that diesel cars that do not satisfy hydrocarbon emission standards cannot enter the city. Therefore, the PAHs emission factor was determined by deriving the average for five diesel cars.

Finally, using the TEF (Toxicity Equivalent Factor) of PAHs against benzo[a]pyrene, the benzo[a]pyrene TEQ emission factor became 660 ng km^{-1} for gasoline car and 1700 ng km^{-1} for diesel car. These values were comparatively higher than the regulation of Euro 2. Since the number of cars tested in this study is not enough, an increase in the number of cars will be required to enhance the accuracy of the emission factor.

3 Numerical simulation model

The CFD software of Flow Designer (Advanced Knowledge Technology Limited.), in which the turbulent model is k-ε and the solution method is SIMPLE, was used in this study. The wind speed was specified at the height of 10 m and the logarithm rule was adopted for the vertical profile of the wind speed, which was set to the inflow boundary, The inflow boundary condition of the concentration was set to 0. The generation of turbulence in the street canyons induced by traffic was not considered.

Figure 2 Crossroad model simplified (see online version for colours)



3.1 Simulation of crossroads

The simplified crossroads model (Shi et al., 2006) is shown in Figure 2. Building width and pavement width are 25 m and 3 m respectively. Wind direction is south. Road width D [m], building height H [m], and wind speed U [ms^{-1}] are the parameters in these simulations. The periodic boundary conditions are set in the east-west lateral boundaries. Emissions are uniformly given in the range from road surface to the height of 2 m. The average concentration is evaluated at the range of 10 m in the crossroads, at the height of 1.5 m on the pavement.

The average concentrations were calculated against all combinations of H = 3, 4, 5, 6, 7, 8, 9, 12, 15, 18, 21, 24, 27, 30, 33, 36, 39, 42, 45, 48, 51, 54, 57, 60 m and D = 8, 12, 18 m and U =1, 2, 3, 4 ms^{-1}. The average concentrations with the change of H are shown in Figure 3. The average concentrations increased until the building height of 9 m and decreased above the building height of 9 m. Therefore, the following two regression equations were obtained by these simulations.

$$C = 3.31U^{-1.09}D^{-0.63}H^{0.02} \quad 3 \le H \le 9 \tag{2}$$

$$C = 5.43U^{-1.04}D^{-0.56}H^{-0.30} \quad 9 \le H \le 60. \tag{3}$$

The flow fields for the building heights of 8 m and 24 m are shown in Figure 4. In the case of low building height, the horizontal wind speed on parallel roads against wind direction was low. In the case of high building height, the horizontal wind speed on parallel roads against wind direction was high. These differences caused the differences of concentration variations accompanying with the building height.

Figure 3 Average concentrations against building height

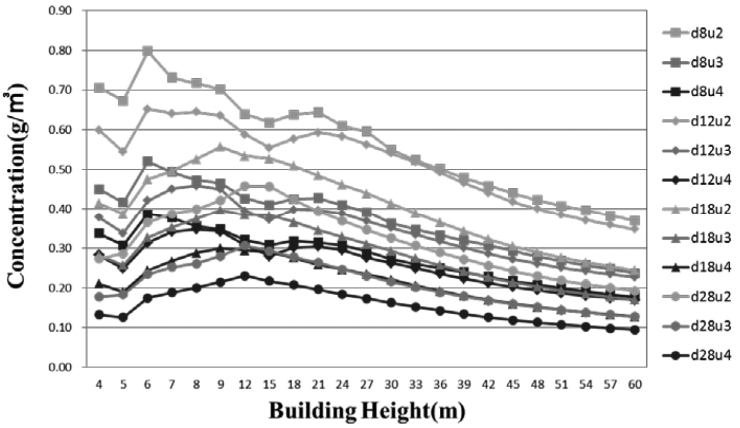
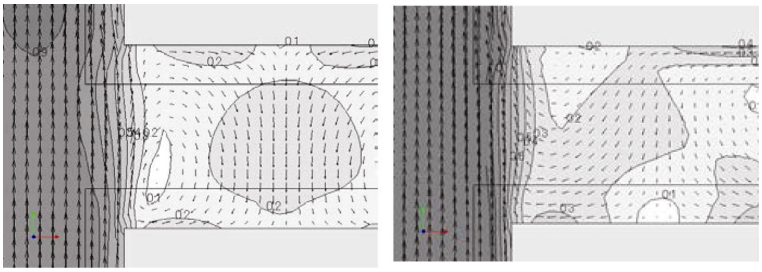


Figure 4 Flow fields against the building height of 8 m and 24 m



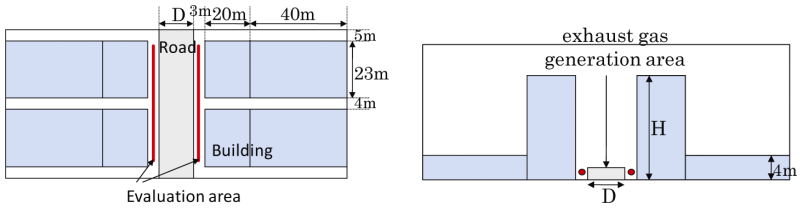
The same simulations were carried out against the crossroads model with the parallel overpass and the perpendicular overpass. The ratio of the average concentration without overpass to the average concentration with overpass was one in almost all cases. When the height of the perpendicular overpass was almost the same as the height of the building, the ratio of the average concentration without overpass to the average concentration with overpass was obviously less than one. From the viewpoint of safety, it was assumed that the overpass existed on no roads in the calculations in Osaka City.

3.2 Simulation of roadsides

The simplified roadsides model is shown in Figure 5. The size of the building facing the road is $23\ \text{m} \times 20\ \text{m} \times H$ [m], and the size of the building away from the road is $23\ \text{m} \times 40\ \text{m} \times 4$ [m]. Pavement width is 3 m. Two wind directions are considered: parallel and perpendicular to the road.

Road width D [m], building height H [m], and wind speed U [ms^{-1}] are the parameters in these simulations. The periodic boundary conditions are set in the parallel lateral boundaries against the wind direction. Emissions are uniformly emitted from the road surface to the height of 2 m. Though there was a great difference between the leeward concentration and the windward concentration, the average concentration is evaluated at the height of 1.5 m above the pavement.

Figure 5 Roadside model simplified (see online version for colours)

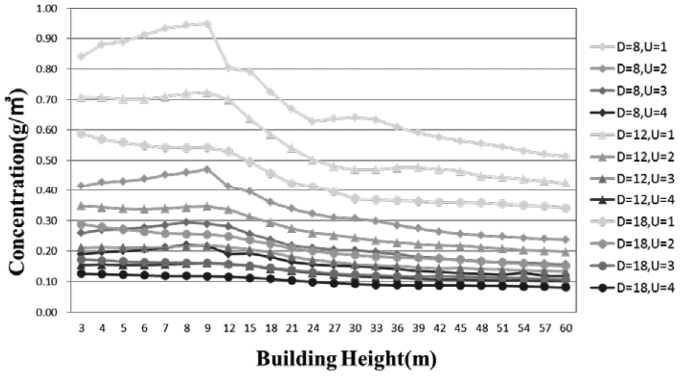


The average concentrations were calculated against all combinations of H = 3, 4, 5, 6, 7, 8, 9, 12, 15, 18, 21, 24, 27, 30, 33, 36, 39, 42, 45, 48, 51, 54, 57, 60 m and D = 8, 12, 18, 28 m, U = 1, 2, 3, 4 ms^{-1}, and wind direction = parallel, perpendicular. The average concentrations for the change of H are shown in Figure 6, when wind direction is perpendicular to the road. Except for the case of U = 1 m/s, the average concentrations showed almost the same tendency. In the cases of U = 1 m/s, a few simulations (H = 21 m to 36 m) did not converge. Therefore, except for the cases of U = 1 m/s, the two regression equations by these simulations were obtained as the index of D + 6, which represents the net distance of both buildings.

$$C = 3.13U^{-1.01}D^{-0.67}H^{0.40} \quad (D+6)/2 > H \tag{4}$$

$$C = 10.06U^{-0.99}D^{-0.41}H^{-0.41} \quad (D+6)/2 < H. \tag{5}$$

Figure 6 Average concentrations against building height (perpendicular)



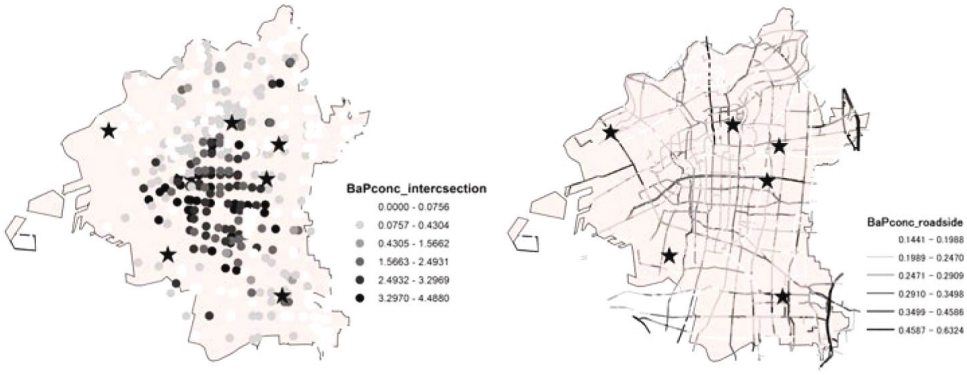
The same simulations were carried out for the parallel wind direction. The average concentration levels were one tenth of the concentrations for the perpendicular wind direction. From the viewpoint of safety, it was assumed that the wind direction was perpendicular to roads in the calculations in Osaka City.

4 Screening results

Benzo[a]pyrene_TEQ concentrations at 6,999 roadsides and 449 crossroads in Osaka City were calculated by using the traffic volume, the emission factor, and the regression equations. Benzo[a]pyrene TEQ concentrations at the crossroads and roadsides are shown in Figure 7 (left) and Figure 7 (right), respectively. The unit risk (http://www.epa.gov/

ncea/iris/index.html) of benzo[a]pyrene is 9×10^{-2} person/μg m^{-3}. Setting the screening level to 10^{-5} person, the concentration of benzo[a]pyrene should be regulated at less than 0.11 ng m^{-3}. In Japan, atmospheric environmental standard of benzo[a]pyrene is not yet regulated. The calculations at the roadsides gave an average benzo[a]pyrene TEQ concentration of 0.63 ng m^{-3}, and the concentration at the roadsides nearly exceeded 0.11 ng m^{-3}. The calculations at the crossroads gave an average benzo[a]pyrene TEQ concentration of 1.96 ng m^{-3}, which was three times higher than that of the roadsides. These results suggest that atmospheric environmental standard of benzo[a]pyrene should be regulated as soon as possible.

Figure 7 Benzo[a]pyrene concentrations at the roadsides and crossroad (see online version for colours)



5 Conclusions

To estimate benzo[a]pyrene concentrations at the roadsides and crossroads in Osaka City, the emission factors of benzo[a]pyrene for three gasoline cars and five diesel cars were measured by using portable sampling equipment that directly collected the exhaust gas. The emission factors of gasoline cars were corrected by considering the cold start and car life time. Using the Toxicity Equivalent Factor of PAHs against benzo[a]pyrene, the benzo[a]pyrene emission factor became 660 ng km^{-1} for gasoline car and 1700 ng km^{-1} for diesel car. To easily evaluate the concentrations at the roadsides and crossroads, regression equations were introduced as the functions of building height, road width, and wind speed from CFD simulations. Benzo[a]pyrene concentrations at the roadsides and crossroads in Osaka City were calculated by using the traffic volume, measured emission factors, and the regression equations. The average benzo[a]pyrene concentration calculated at the roadsides and crossroads was 0.63 ng m^{-3} and 1.96 ng m^{-3}, respectively, which exceeded the screening level for 10^{-5} person. These results suggest that the atmospheric environmental standard of benzo[a]pyrene in Osaka City should be regulated as soon as possible.

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