

Estimation of the emission factors of PAHs by the traffic activities with the model of atmospheric dispersion and deposition from heavy traffic road

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Abstract

In order to consider the total atmospheric loadings of the PAHs (polycyclic aromatic hydrocarbons) from traffic activities, the emission factors of PAHs were estimated and from the obtained emission factors and vehicle transportation statistics, total atmospheric loadings were integrated and the loadings into the water body were estimated on a regional scale. The atmospheric concentration of PAHs was measured at the roadside of a heavy traffic road in the Hiroshima area in Japan. The samplings were conducted in summer and winter. Atmospheric particulate matters (fine particle, 0.6~ 7 μm ; coarse particle, over 7 μm) and their PAH concentration were measured. Also, four major emission sources (gasoline and diesel vehicle emissions, tire and asphalt debris) were assumed for vehicle transportation activities, the chemical mass balance method was applied and the source partitioning at the roadside was estimated. Furthermore, the dispersion of atmospheric particles from the vehicles was modelled and the emission factors of the sources were determined by the comparison to the chemical mass balance results. Based on emission factors derived from the modelling, an atmospheric dispersion model of nationwide scale (National Institute of Advanced Industrial Science and Technology - Atmospheric Dispersion Model for Exposure and Risk assessment: AIST-ADMER) was applied, and the atmospheric concentration and loading to the ground were calculated for the Hiroshima bay watershed area.

Keywords

Polycyclic aromatic hydrocarbons; PAHs; particulate matter; vehicle emission factor

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds composed of two or more fused aromatic rings. Several compounds of this class are believed to be human carcinogens. PAHs are emitted into atmospheric environments (mainly due to incomplete combustion processes), deposited on ground surfaces, and discharged into aquatic environments. Many researchers have estimated PAH emissions on a nationwide scale, or the atmospheric concentrations and depositions have been measured (Baek et al. 1991; Fukushima et al. 2003; Ozaki et al. 2006). According to the results, vehicle emissions are the major sources of PAH emissions in urban areas. These relations, however, have not been scientifically determined yet. Also, inventory analysis based on the total loadings has not been well-established for PAHs. One major difficulty is the uncertainty of the emission factors of vehicles in the actual environment. In the present study, in order to consider the total atmospheric loadings of the PAHs from traffic activities, the emission factors of PAHs were estimated using an atmospheric dispersion model based on the PAHs concentration changes and vehicle transportation statistics at a heavy traffic road. Total atmospheric loadings results were then integrated and the loadings into the water body was estimated on a regional scale.

EXPERIMENTAL METHODS

Sampling campaigns

Roadside samplings were conducted at the roadside of a heavy traffic road (national road Route 2, Higashihiroshima, Japan). It was located in a rural area, and no other major source of PAHs emissions was found in the vicinity of the sampling area. The average traffic density was around 25,000 vehicles d^{-1} . Atmospheric particulate matter was collected at the roadside, 1 m away from the road. The sampling height was set at 1 m above the ground for the atmospheric measurements. Atmospheric particulate matter was collected using a high-volume air sampler (HVS-500-5; Shibata Kagaku Co., Ltd.) with an

impactor system followed by a glass fiber filter. The sampling was conducted 24 or 48 hours, and the sampling rate was 500 L min⁻¹. Using the impactor system, particles of > 7 µm (coarse particulate matter) were trapped on a stainless plate. After sieving, particles of 0.6 ~ 7 µm (fine particulate matter) were collected on a glass fiber filter (GB-100R, Advantec Co., Ltd.). Prior to the experiment, the stainless plate and glass fiber filter were precombusted at 110 °C for 24 hours. The first sampling session was performed in winter, from 19 to 20 December, 2001. The second sampling session was performed in summer (from 22 to 24 July, 2002). In order to measure the concentration changes within a day, each sampling lasted 4 hours and the sieve and filter were changed for each sampling. During the sampling sessions, air temperature, wind speed, and traffic density were measured. Air temperature was measured with an electric sensor (StowAway TidbiT temperature data logger -5°C to +37°C Model, Onset Computer Co., Ltd.). Wind speed was measured with a wind speed sampler (KADEC21-KAZE, Kona System Co., Ltd.). Traffic density was counted by VTR, classified into light-duty vehicles (LDV) and heavy-duty vehicles (HDV) which was separated by the gross weight limit of 3.5 t. The traffic of motorcycle was neglected because it was extremely lower (less than 200 vehicles d⁻¹). The atmospheric and traffic conditions were given in **Table 1**. Climate conditions were stable and mild, and no precipitation was observed during the samplings. The traffic density was higher during the winter sampling period, and for comparison of LDV and HDV, HDV traffic is higher for both sampling periods.

PAHs extraction and analysis

The details of the extraction and analysis were shown elsewhere (Ozaki et al. 2006). Sixteen unsubstituted PAHs were measured: acenaphthylene (Ace), acenaphthene (Act), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo(a)anthracene (B(a)A), chrysene (Chr), benzo(b)fluoranthene (B(b)F), benzo(k)fluoranthene (B(k)F), benzo(e)pyrene (B(e)P), benzo(a)pyrene (B(a)P), dibenzo(ah)anthracene (D(ah)A), benzo(ghi)perylene (B(ghi)P), and indeno(123-cd)pyrene (Ind(123)P). The detection limit was set at the level of 3 in the SN ratio. Detection limits ranged from 1 to 5 ng for individual PAHs. Within this level, the CV ratio of each of the compounds was less than 20 %. Quality of extraction was checked using dried marine sediments (HS-3B, National Research Council of Canada Institute for Marine Biosciences). The recovery averaged 50 ~ 80% for all PAHs, and the repetition error was 5 ~ 10%.

Chemical mass balance method

In considering the sources of PAHs in this study, four main emission sources were supposed, i. e., gasoline and diesel tail pipe exhaust, tire and asphalt debris, and the chemical mass balance was taken for ten PAHs (Pyr, B(a)A, Chr, B(b)F, B(k)F, B(e)P, B(a)P, D(ah)A, B(ghi)P, and Ind(123)P) (CMB method; Hayakari and Hanaishi, 2001). Light molecular portions (Ace, Act, Flu, Phe, Ant, Flt) were excluded from the CMB analysis for minimizing the volatilization effects. For B(b)F and B(k)F, the two values were added to each other because the summed values were used for many other referred source measurements, and nine variables were consequently applied for CMB analysis. PAHs concentration patterns for each source were summarized elsewhere (Ozaki et al. 2005).

Modelling of PM and PAHs dispersion at the roadside

PAH dispersion was described numerically on a 3-dimensional meshed type model. The basic equations were continuous equations and advection equations, and pollution dispersion was described using the plume puff model. The input data was traffic (density and velocity time series), and atmospheric data (air temperature, and wind speed and direction). The obtained wind data was at 1.5 m height, and the height profile of wind speed was described on the similarity law of the boundary layer. The turbulence arisen from vehicle movement was described by the wake theory developed by Eskridge and Hunt (1979). The spatial volume was set to 1,000 m (road direction) 10 m in height 50 m in width. Road was set to be at the center in the calculation area.

RESULTS

Atmospheric concentrations

The climatic conditions and atmospheric concentrations of PM and total PAHs in particulate matter are listed in **Table 1**. In the sampling periods, temperature and precipitation were moderate for this sampling site. For particulate matters, the atmospheric concentration of FPM (fine particulate matter) was 52.6 ~

136.3 $\mu\text{g m}^{-3}$, and that of CPM (coarse particulate matter) was 0.69 ~ 10.4 $\mu\text{g m}^{-3}$. For PAHs, the atmospheric concentration of total sixteen PAHs (16PAHs) in FPM was 0.26 ~ 4.41 ng m^{-3} , and that in CPM was 0.001 ~ 1.16 ng m^{-3} . For both FPM and CPM, the concentration is higher in winter than summer. This may be due to the seasonal fluctuation because this pattern is commonly observed in many atmospheric measurements (Fukushima et al. 2003; Ozaki et al. 2006; Wu and Fang 2001; Panther et al. 1999). But at the same time, the transportation density was lower in summer, and this would also be the reason for it.

Table 1. Atmospheric conditions and traffic density in the sampling events

	19 to 20 Dec., 2001	22 to 24 Jul., 2002
(Air temperature, wind speed: daily average; In the parenthesis is the min. to max.)		
Air temperature ($^{\circ}\text{C}$)	3.0 (-2.2~13.0)	28.2 (21.8~38.3)
Wind speed (m s^{-1})	0.50 (0.0~1.6)	0.91 (0.2~2.3)
LDV* (vehicles d^{-1})	22400	11700
HDV* (vehicles d^{-1})	24800	14300
PM concentration	n=6	n=12
FPM ($\mu\text{g m}^{-3}$)	76.6 (52.6~90.1)	89.3 (57.2~136.3)
CPM ($\mu\text{g m}^{-3}$)	3.0 (1.51~4.79)	5.2 (0.69~10.4)
Total 16PAHs		
in FPM (ng m^{-3})	3.02 (1.83~4.41)	0.70 (0.26~1.18)
in CPM (ng m^{-3})	0.098 (0.069~1.16)	0.018 (0.001~0.073)
10PAHs for CMB		
in FPM (ng m^{-3})	2.07 (1.25~3.51)	0.21 (0.18~0.93)
in CPM (ng m^{-3})	0.049 (0.034~0.067)	0.011 (0.000~0.051)

*LDV: Heavy-duty vehicle, LDV: Light-duty vehicle

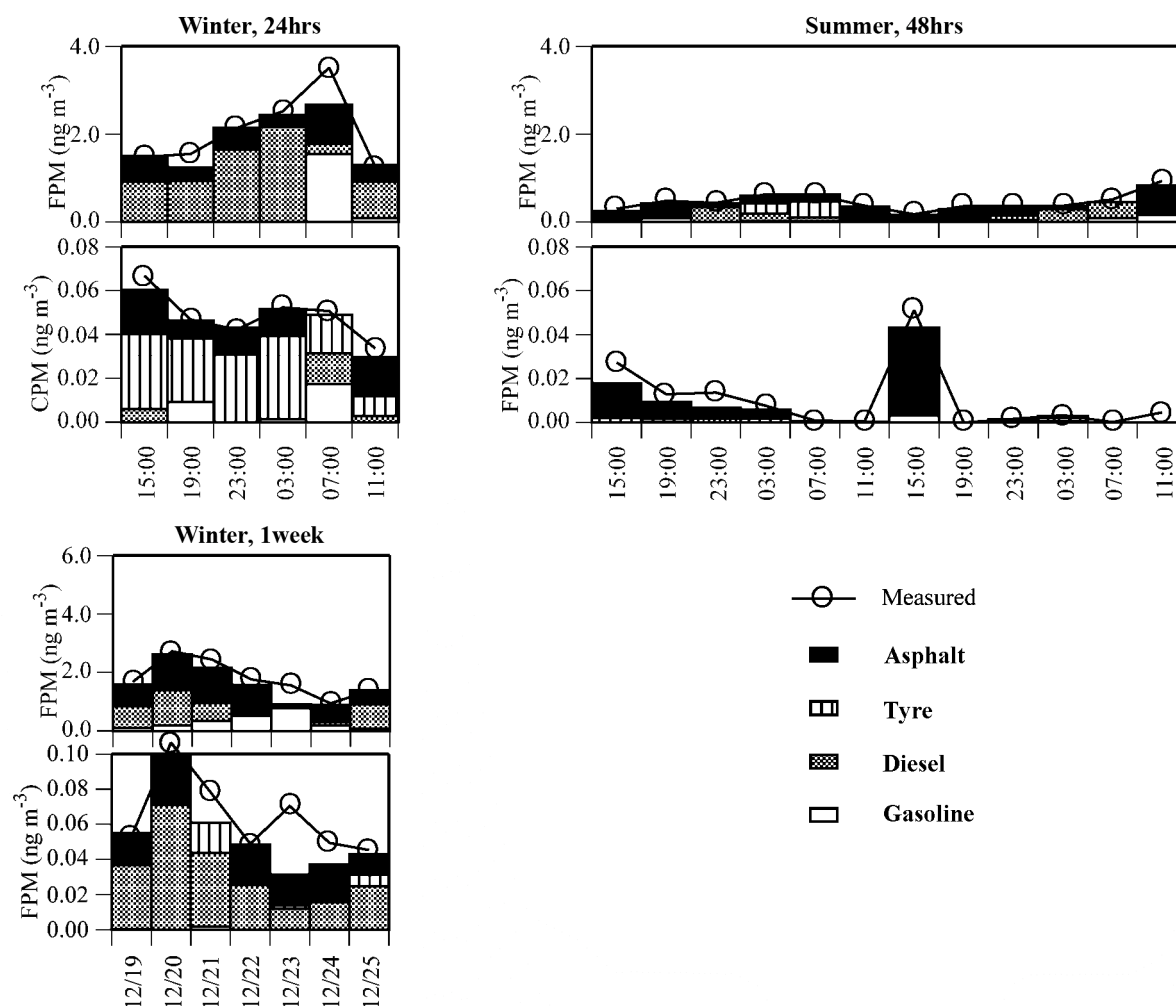


Fig. 1 Profile of atmospheric and deposition PMs and PAHs.

The temporal variations of 10PAHs concentration are shown in **Fig. 1**. The CMB source apportionments results are also shown. The concentration in summer is clearly lower. For each source, diesel tail pipe emission is higher in overall, and the gasoline tail pipe emission is lower than those of diesel. Tyre debris is higher in CPM in winter. In summer, on the other hand, the level of asphalt is relatively higher. In the winter, one week sampling was also carried out. The results of 2001/12/19 correspond to the 24-hour measurements in winter. For the averaged total level, the values are comparable to those of 2001/12/19. The source apportionment pattern is also similar. For CPMs, on the other hand, patterns were a bit different. In the 24-hour results, tyre debris is higher. On 2001/12/19 in the one week sampling, the diesel is higher. The reason for this discrepancy is not clear.

Modelling of source apportionment profile

Fitting the concentration profile onto the measured results by model, emission factors of vehicle transportation can be determined. Prior to the fitting, initial values for each vehicle classification were set from the literature. Vehicle classification was different for different analyses. The classification in CMB for this study was gasoline and diesel for tail pipe emissions; however, the classification by the other researchers (e.g. Namikawa et al., 2003) was a bit more detailed for diesel. Also, the classification for video observation by this study (LDV, HDV) may not correspond to gasoline and diesel vehicles because the category ‘passenger vehicles’ include diesel vehicles as well as gasoline vehicles. The correspondence of each classification is shown in **Table 2**. The ratio for each composition was obtained from the literature (Namikawa et al. 2003). The emission factors obtained from the literature were shown in **Table 3** for particulate matters. Asphalt emission factors could not be obtained. From the obtained values, initial values for modelling were set as shown in **Table 4**. From the PM emission factors, PAHs emission factors were determined using the following relation:

$$EF_{PAHs}(ngPAHs\ km^{-1}\ v\ h^{-1})=EF_{PM}(mgPAH\ km^{-1}\ v\ h^{-1})\cdot C_{PAH}(\mu gPAH\ gPM^{-1})$$

PAHs contents(C_{PAH}) was determined by the direct measurements (Ozaki et al., 2005). EF_{PM} varied with test conditions (driving mode, weight, etc.). For purposes of representation in our study, the averaged value of similar velocity ranges was used. Our measuremental averaged velocity was 55 ± 5 ($km\ hr^{-1}$; average \pm standard deviation). So, the literature values in the condition of $60\sim80$ ($km\ hr^{-1}$) driving speed were picked out and averaged for our study. (The number of data for $40\sim60$ ($km\ hr^{-1}$) was not enough for our study.) The summarized initial emission factors for the modelling are shown in **Table 4**. For tyre and asphalt, emission factors was supposed to be proportional to the number of tyres, and the tyre numbers of each class was supposed to be 4(GPV), 8(DPV), and 12(DFV). From the results, fitting parameters for the modelling were set, as shown in **Table 5**. The modelled data was fit for winter measurements because PAHs concentration is considered to be stable in winter.

Before fitting the data, a time-averaged concentration profile for along the roadside was compared with the measuremental data (**Fig. 2**). The emission factors were set at the fitted values shown hereafter (**Table 6**). Though the measuremental results were obtained in a different season (11/19~26), the decreasing concentration profile can still be usefully compared to the winter or summer data of our study. In both, the concentration comes near to the zero or background level at around 30 m. The model can be considered to describe the air dispersion. The background level was about half of the maximum at the roadside. In the fitting, the background level was not subtracted. As a result, our EF may be an overestimate and if this supposition is correct, the ratio of the overestimation would be twice the actual value. The reason for omitting the effect was that it was difficult to determine the background level at each time and source.

Table 6 shows the overall fitting results of emission factors, and **Fig. 3** shows the time profile of the fitting results of one-day measurement in winter (2001/12/19~20). Compared to the measuremental results (**Fig. 1**; Winter, 24hrs), the model described the time profile of PAHs well for both FPM and CPM. Compare to the initial values of emission factors determined from the literature, the model values were within a factor of $1/100\sim100$ (k values of **Table 6**). These discrepancies would mainly be due to the fluctuation of emission factors of PAH with driving test conditions or vehicle conditions. PAH emission factors generally vary by 100-fold in different studies (Ozaki et al. 2006). In this study, overall averaged emission factors were successfully obtained from the field observation and modelling. Our obtained emission factors were about 500, 1000~4000, 10~40, and 200~500 ($ng\ km^{-1}\ v\ h^{-1}$) for gasoline tail pipe, diesel tail pipe, tyre debris, and asphalt debris, respectively (PM(=FPM+CPM) results in **Table 6**). With

the detailed observation, gasoline FPM emissions ($k_{G,FPM}=73$) and tyre CPM emissions ($k_{T,CPM}=0.05$) deviate significantly from the literature values. For estimation of gasoline FPM, the one measuremental value was abnormally higher (7:00 measurement at winter, 24hrs in **Fig. 1**). When recalculation is made without this result, emission factors come near to the literature values ($k_{T,CPM}=5.0$), suggesting the abnormality of this gasoline FPM result. For tyre emissions, the literature value that we used for the study was the lowest, and still, our estimation is far lower than the literature. The reason for this was not clear. For asphalt, the obtained values were higher and comparable to that of the results for tail pipe emissions. We have found no other research that can compare to our study, though further study is needed to validate this.

Table 2. Vehicle classification

Classification for CMB		by the literatures	(Abbrev.)	Namikawa <i>et al.</i> (2003)	for video observation by this study
Gasoline		Gasoline passenger vehicle	GPV	83.5%	LDV (light duty vehicle)
		Diesel passenger vehicle	DPV	16.5%	
Diesel		Diesel freight vehicle (small, middle size)	DFV(S,M)	67%	HDV (heavy duty vehicle)
		Diesel freight vehicle (large size)	DFV(L)	33%	

Table 3. Emission factors of vehicles

		PM	Tyre	Asphalt
		Tail pipe		
		(mg km ⁻¹ vh ⁻¹)	(mg km ⁻¹ vh ⁻¹)	(mg km ⁻¹ vh ⁻¹)
PM	GPV	0.58~6.0		
	DPV	64~147	200 (JCAP, 2002)	
	DFV(S,M)	55~149	20 (Doki <i>et al.</i> 2006)	no data
	DFV(L)	(mg km ⁻¹ t ⁻¹) 36~61	5 (USEPA, 1995)	
References		Namikawa <i>et al.</i> (2003)		

Table 4. Initial emission factors for calculation

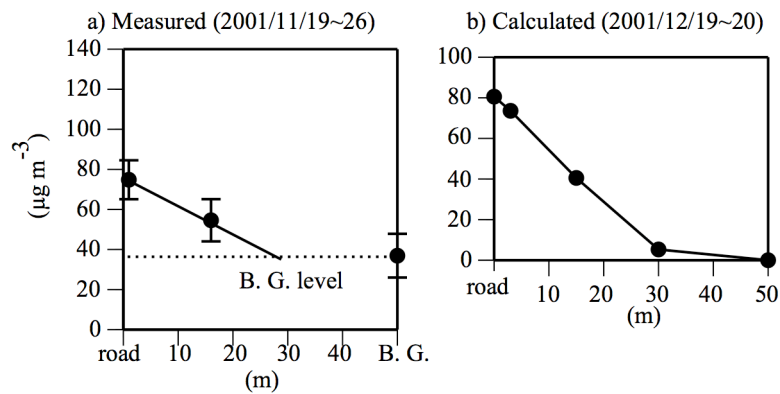
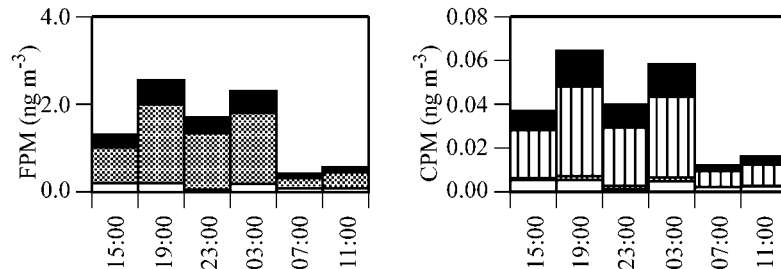
		PM			9PAHs		
		Tail pipe	Tyre	Asphalt	Tail pipe	Tyre	Asphalt
		(mg km ⁻¹ vh ⁻¹)	(mg km ⁻¹ vh ⁻¹)	(mg km ⁻¹ vh ⁻¹)	(ng km ⁻¹ vh ⁻¹)	(ng km ⁻¹ vh ⁻¹)	(ng km ⁻¹ vh ⁻¹)
FPM	GPV	0.83	0		6.60	0	
	DPV	70	0		1383	0	
	DFV(S,M)	53	0		1041	0	
	DFV(L)	264	0		5220	0	
CPM	GPV	0.02	5	no data	13.33	271	no data
	DPV	0.13	10		0.05	541	
	DFV(S,M)	0.10	10		0.04	541	
	DFV(L)	0.49	15		0.18	812	

Table 5. Calculation scheme of EFs

		9PAHs		
		Tail pipe	Tyre	Asphalt
		(ng km ⁻¹ vh ⁻¹)	(ng km ⁻¹ vh ⁻¹)	(ng km ⁻¹ vh ⁻¹)
FPM	GPV	6.6*k _{G,FPM}	0	4*EF _{A,FPM}
	DPV	1383*k _{D,FPM}	0	8*EF _{A,FPM}
	DFV(S,M)	1041*k _{D,FPM}	0	8*EF _{A,FPM}
	DFV(L)	5220*k _{D,FPM}	0	12*EF _{A,FPM}
CPM	GPV	13.33*k _{G,CPM}	271*k _{T,CPM}	4*EF _{A,CPM}
	DPV	0.05*k _{D,CPM}	541*k _{T,CPM}	8*EF _{A,CPM}
	DFV(S,M)	0.04*k _{D,CPM}	541*k _{T,CPM}	8*EF _{A,CPM}
	DFV(L)	0.18*k _{D,CPM}	812*k _{T,CPM}	12*EF _{A,CPM}
Free parameters		k _{G,FPM} , k _{D,FPM} , k _{G,CPM} , k _{D,CPM}		
Parameter Unit		k _{T,CPM}		
		(Calculated EF/Literatures EF)		
		(Calculated EF/Literatures EF)		
		EF _{A,FPM} , EF _{A,CPM}		
		(ng km ⁻¹ tyre ⁻¹)		

Table 6. Estimated EFs by calculation

		9PAHs		Tyre		Asphalt	
		Tail pipe					
		(ng km ⁻¹ vh ⁻¹)	(Est./Lit.)	(ng km ⁻¹ vh ⁻¹)	(Est./Lit.)	(ng km ⁻¹ vh ⁻¹)	(ng km ⁻¹ tyre ⁻¹)
FPM	GPV	481	$k_{G,FPM}=73$	0		183	
	DPV	940		0		366	
	DFV(S,M)	708	$k_{D,FPM}=0.68$	0	-	366	$EF_{A,FPM}=45.7$
	DFV(L)	3550		0		548	
CPM	GPV	13	$k_{G,CPM}=0.97$	13.6		5.4	
	DPV	1.04		27.1		10.9	
	DFV(S,M)	0.78	$k_{D,CPM}=21.2$	27.1	$k_{T,CPM}=0.05$	10.9	$EF_{A,CPM}=1.36$
	DFV(L)	3.9		40.7		16.3	
PM (=FPM +CPM)	GPV	494		13.6		188	
	DPV	941		27.1		377	
	DFV(S,M)	709		27.1		377	
	DFV(L)	3554		40.7		565	

**Fig. 2** Comparison of PM concentration profile from the roadside (PM: FPM+CPM).**Fig. 3** Measured and calculated PAHs concentration time profile.

Estimation of total vehicle transportation emissions in the Hiroshima bay watershed area

The total PAHs loading arisen from vehicle transportation activities was estimated using the regional vehicle transportation statistics in the Hiroshima bay watershed (the total population: 9.8×10^5 , area: 1,710 km²) and the obtained emission factor data. From the statistics for the year of 2001, the total emissions were summarized using a calculation process of a nationwide meshed-type atmospheric dispersion modelling program (National Institute of Advanced Industrial Science and Technology - Atmospheric Dispersion Model for Exposure and Risk assessment: AIST-ADMER). **Table 7** shows the summary of emissions. Along with the vehicle transportation, the estimate of the yearly atmospheric deposition is shown (the data was calculated similar to the calculation by Ozaki et al., 2006). Comparing to this, the vehicle transportation emission was calculated to 7.4% of the deposition. While the emission is overestimation because EFs were obtained using winter data (winter concentration level is higher than summer) and moreover, background level was not subtracted, the vehicle transportation emission is significantly lower than the atmospheric deposition. Baek et al. (1991) summarized various nationwide inventory analyses of PAHs in U.S.A, Sweden, and Norway, and the contribution of vehicle sources was 7~25%. While these analyses did not include tyres and asphalt as a source of vehicle activities, the order of the source contribution was similar to our results.

Table 7. Total vehicle transportation emission and atmospheric deposition loading in Hiroshima bay watershed area

(kg year ⁻¹)	Vehicle transportation emissions	Atmospheric deposition*
Gasoline tail pipe	2.0	-
Diesel tail pipe	2.3	-
Tyre	0.13	-
Asphalt	1.8	-
Total	6.2	90

*The value was calculated on the results obtained by Ozaki et al., (2006)

CONCLUSIONS

In this study, the atmospheric concentration of PAHs was measured and the source apportion was calculated using the CMB analysis at the roadside of a heavy traffic road in Hiroshima area in Japan. The samplings were conducted in summer and winter. From the results, the averaged emission factors in actual environments of gasoline and diesel tail pipe, tyre and asphalt debris were successfully determined by atmospheric dispersion modelling. Further, from the obtained emission factors, the vehicle transportation was summarized in a watershed area and compared to the atmospheric depositions. The contribution of transportation was estimated to around 7% of the total atmospheric loading.

This study focused on particle PAHs. The loads from the vapor phase and the dissolved phase were not estimated due to the lack of sampling of the vapor phase in the atmosphere. The vaporization or condensation can affect the PAHs behavior even for the higher molecular weight portions. Another limitation of our study is that we have just focused on the vehicle transport emissions. Other than by vehicle transport, PAHs can be emitted by industrial emissions, domestic heating, or petroleum leakage. The results of our study should be compared to these other source investigations.

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