# ESTIMATION OF ORIGINS OF POLYCYCLIC AROMATIC HYDROCARBONS IN SIZE-FRACTIONATED ROAD DUST IN TOKYO WITH MULTIVARIATE ANALYSIS

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## ABSTRACT

This study aimed to estimate the origins of polycyclic aromatic hydrocarbons (PAHs) in size-fractionated road dust in Tokyo. First of all, seven categories of PAHs sources were defined: diesel vehicle exhaust, gasoline vehicle exhaust, tire, pavement, asphalt or bitumen, petroleum products excluding tire and asphalt, and combustion products except for those in vehicle engines. The 189 source data of 12-PAHs profiles were classified into 11 groups (from S1 to S11) based on cluster analysis combined with principal component analysis. Next, 18 road dust samples were collected from eight streets in Tokyo and fractionated into four different particle-size-fractions: 0.1-45, 45-106, 106-250, and 250-2,000μm. In order to estimate the contributions of the classified source groups (S1-S11) to PAHs in the road dust, multiple regression analysis was performed with 12-PAH profile of the road dust as dependent variable and average 12-PAHs profiles of the 11 source groups as 11 explanatory variables. According to the results, diesel vehicle exhaust, tire and pavement were the major contributors of PAHs in the fractionated road dust. Although the estimated contributions of the 11 source groups varied among the particle-size-fractions, there was no clear and consistent relationship between particle size and the major PAH contributor.

**KEYWORDS : pavement, polycyclic aromatic hydrocarbons, road dust, size-fractionated sample, tire, vehicle exhaust**

## INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) , many of which are genotoxic that may cause mutations (Mirisola et al., 2001) and certain types of cancer (Baumann, 1998; Collins et al., 1998), have been detected widely in water environment. Effective control strategies for controlling PAHs release are required in order to *assure the safety* of humans and ecosystems.

Road dust has been recognized to bring a large amount of PAHs into water environment via road runoff (Brown et al., 1985; Maltby et al., 1995; Boxall and Maltby, 1995), but only few attempts have been done to quantitatively assess the comparative contribution of various PAHs sources to road dust. The possible PAHs sources in road dust are diesel vehicle exhaust, gasoline vehicle exhaust, tire, pavement (asphalt or bitumen), oil spill and so on. Based on the enrichment factor, Takada et al.(1990) indicated vehicle exhaust as a main PAHs contributor to the road dust collected from roads with heavy traffic while atmospheric fallout was more significant in residential area in Tokyo. Zakaria et al.(2002) suggested the used crankcase oil as a major PAHs contributor to road dust in Malaysia.

The objective of this study is to assign the comparative contribution of possible source to PAHs in fractionated road dust sample with a statistical approach. One hundred eighty nine PAHs data of possible sources were obtained from literature and from our experiment. Principal component analysis and cluster analysis were applied to classification of PAHs profiles of possible sources and multiple regression analysis were conducted to estimate the contribution of the classified sources to the PAHs in road dust collected in Tokyo.

## METHODS

### *Classification of PAHs profile of possible sources*

#### Source data

One hundred eighty nine PAHs data of possible sources were obtained from literature and from our experiment (see next section). The obtained data were categorized into seven possible sources in Table 1: diesel vehicle exhaust [D], gasoline vehicle exhaust [G], tire [T], asphalt-pavement [P], asphalt or bitumen [A], petroleum products excluding tire and asphalt [O], and combustion products except for those in vehicle engines [E].

#### Sampling and PAHs analysis

Two samples of diesel exhaust and four samples of gasoline exhaust were collected at 1cm apart from an exhaust pipe of an idling automobile operated in acceleration/deceleration cycles (1,000 rpm ~ 5,000 rpm ~ 1,000 rpm). The sampling equipment consisted of a glass fiber filter (Millipore, AP20) and an air pump (SKC, AirChek HV30). Eight tire samples were obtained by cutting used tires produced by five different tire companies. Eight asphalt-pavement samples were collected from four street reconstruction sites in Tokyo and were homogenized by grinding. Eight samples of asphalt were derived from four Japanese companies. All samples except those of asphalt were dried for 24 hours in a desiccator before PAHs extraction.

**Table1 Number of collected data as possible PAHs sources**

| Source | Category | number of PAHs data | |
|---|---|---|---|
| | | this study | literature * |
| diesel vehicle exhaust | [D] | 2 | 77 |
| gasoline vehicle exhaust | [G] | 4 | 49 |
| tire | [T] | 8 | - |
| asphalt-pavement | [P] | 8 | - |
| asphalt or bitumen | [A] | 8 | 3 |
| petroleum products excluding tire and asphalt | [O] | - | 10 |
| combustion products except for those in vehicle engines | [E] | - | 20 |
| Total | | 30 | 159 |

* refered literature was listed at the end of this paper

The analytical procedure for PAHs was adapted from EPA method 3550 (EPA.,1996) and method TO-13A (EPA.,1999). Firstly, samples were spiked with surrogates (Semivolatile internal standard mix, Sigma 4-8902) and were thawed at room temperature for half a day before extracted with 50 mL dichloromethane in ultrasonic bath (output 100 W, duty cycle 50 %, 30 minutes). The extracts were concentrated to 1 mL in an evaporator at 40 °C and –70.6 kPa. After adding internal standard (Semivolatile base/neutrals surrogate spike mix, Sigma 4-8935), the concentrated extracts were then analyzed for PAHs by GC/MS (Hewlett Packard HP6890, MS5973) with HP5-TA column (30 m × 0.25 μm id) using selected ion monitoring (SIM) mode. In this system, helium gas was used as the carrier gas with the flow rate of 1 mL/min. The oven temperature program started at 50°C with 3 minutes hold, and covered a range from 50°C to 250°C at 20°C/min. The temperature was then held for 12 minutes before heated to 300°C at 22°C/min and held on finally for 4 minutes.

**Classification of PAHs profiles**

Data analysis of this study are mainly based on the content of the following 12 individual PAHs: phenanthrene (Ph), anthracene (An), fluoranthene (Fr), pyrene (Py), benzo(a)anthracene (Ba), chrysene (Ch), benzo(k)fluoranthene +benzo(b)fluoranthene (Bf), benzo(a)pyrene (Bpy), indeno(1,2,3-cd)pyrene (In), dibenz(a,h)anthracene (Db) and benzo(ghi)perylene (Bpe). The ratios of the 12 PAHs concentration to the sum of those were calculated and the composition was defined as PAHs profile in this study.

Since PAHs profile in the possible sources varied even in same category, a simple averaged profile could not represent the source category. Then, it was necessary to conduct a different classification in terms of PAHs profile similarity. In this study, principal component analysis (Varimax rotated) followed by cluster analysis (Ward method) was applied to classify the 189 profiles. Through the principal component analysis, the PAHs profiles were transformed into factor scores. Based on the factor scores, the 189 source data were then classified by cluster analysis. Because the clusters will be explanatory variables of the following source analysis for PAHs in road dust, the cluster should be ideally independent to each other. To reduce the dependency among the clusters, similar clusters were merged under the criteria: two clusters should be merged when the determination coefficient was greater than 0.70 for explaining the PAHs profiles in one cluster by average PAHs profile in the other cluster.

***Estimation of source contribution to PAHs in size-fractionated road dust***

Sampling, size fractionation and PAHs analysis

Eighteen road dust samples were collected at the gutters of eight streets in Tokyo: Sakurada, Kannana, Keiyo, Aratamagawasuido, Kanpachi, Showa, Oume and Kiyosumi. Considering the road runoff process, the sampling was done in wet condition with a carpet vacuum cleaner (Puzzi100, Kärcher) operated with 1.6 L/min of water spray (Milli-Q water). Road dust accumulating on 2.3 m length of the gutter was gathered as one sample. The recovered liquid was filtered through four stainless-steel mesh (2,000, 250, 106 and 45μm) and a membrane filter (pore size 0.1 μm; Omnipore, Millipore). The fractionated road dust (0.1-45μm, 45-106μm, 106-250μm and 250-2,000μm) was applied PAHs analysis in the same way with source.

Estimation of source **contribution to** PAHs in road dust

Multiple regression analysis was performed in order to estimate the contributions of possible sources to PAHs in 72 road dust samples (18 roads X 4 fractions). The PAH profile of each size-fractionated road dust sample was used as dependent variable and the average PAH profiles of the classified source groups were applied as explanatory variables. To find out the major PAHs contributor, a variable forward selection method was applied. A regression model explaining with the least numbers of independent variables at the determination coefficient of greater than 0.80 was adopted for result interpretation. The ratio of each regression coefficient to the sum of them was defined as a comparative contribution (%) of each source group to road dust. For further discussion, comparative source contributions for non-fractionated samples were also calculated.

## RESULTS AND DISCUSSIONS

***Classification of PAHs profiles in possible sources***

The 189 PAHs profiles of possible sources were classified into 11 groups (S1- S11) (Table 2). It was found that S4 and S5 consisted of the profile data only from [G], and that S8 and S9 were composed of only [E], while the other seven groups included several categories of sources. However, it could be interpreted that the 12-PAH profiles of samples in S1 indicated [T], that those of S2 and S3 implied [D] and that those of S7 represented [P] and [A]. S6 and S10 were minor groups having only three data each. S11 included a large numbers source data in various categories and was difficult to be translated.

**Table2 Classification of PAHs profiles in 189 source data**

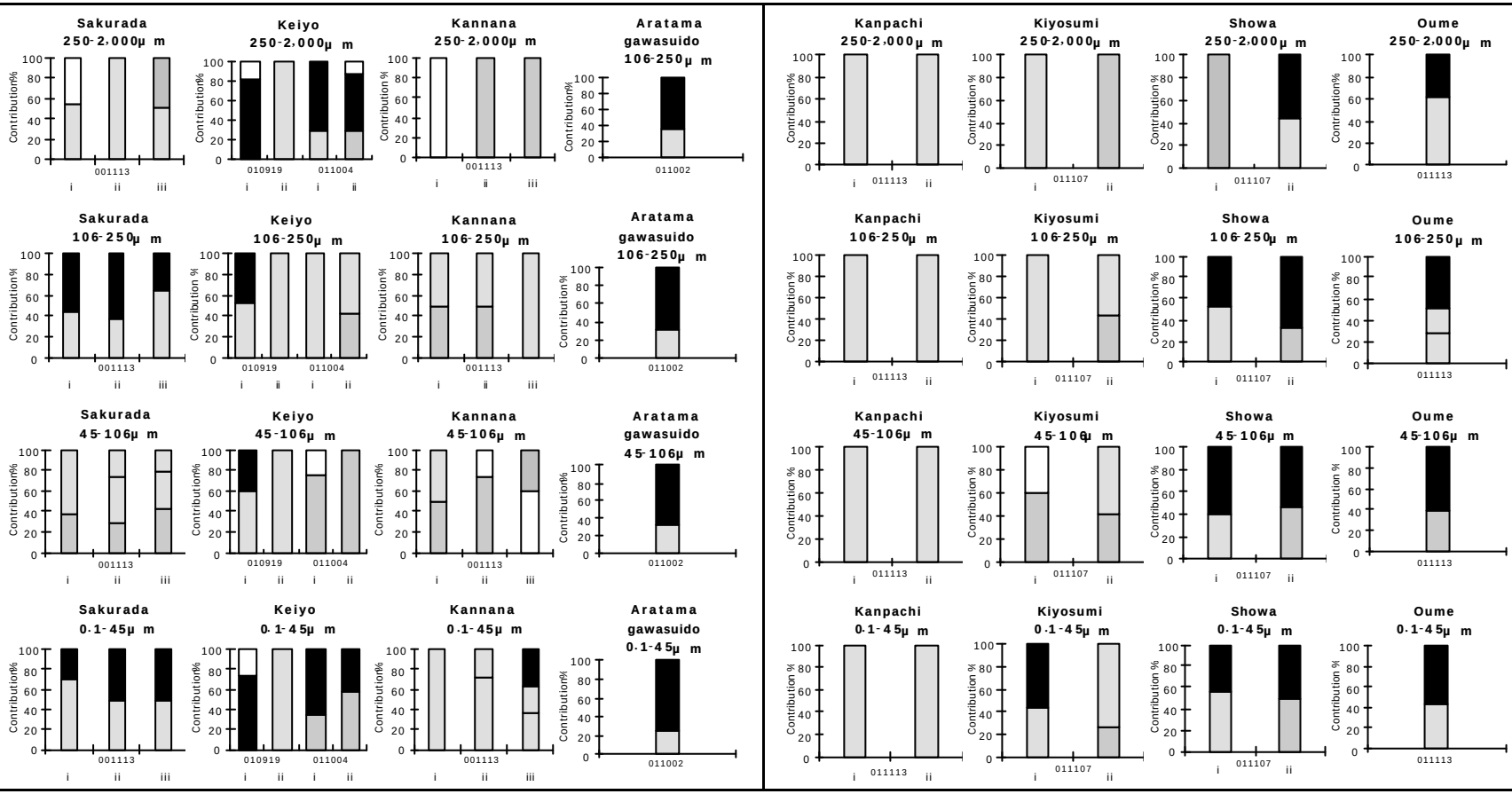
| | S1 | S2 | S3 | S4 | S5 | S6 | S7 | S8 | S9 | S10 | S11 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| ***Average PAHs profiles*** | | | | | | | | | | | |
| phenanthrene (Ph) | 6 | 26 | 7 | 3 | 4 | 31 | 16 | 17 | 11 | 1 | 48 |
| anthracene (An) | 1 | 4 | 43 | 27 | 7 | 8 | 3 | 2 | 3 | 2 | 15 |
| fluoranthene (Fr) | 13 | 22 | 9 | 15 | 3 | 1 | 6 | 6 | 51 | 0 | 8 |
| pyrene (Py) | 43 | 31 | 8 | 13 | 14 | 2 | 11 | 19 | 6 | 0 | 10 |
| benzo(a)anthracene (Ba) | 0 | 4 | 3 | 26 | 11 | 7 | 5 | 2 | 4 | 1 | 5 |
| chrysene (Ch) | 3 | 4 | 3 | 1 | 10 | 10 | 12 | 2 | 4 | 1 | 2 |
| benzo(k)fluoranthene + benzo(b)fluoranthene (Bf) | 3 | 2 | 8 | 4 | 28 | 27 | 15 | 10 | 7 | 6 | 3 |
| benzo(a)pyrene (Bpy) | 4 | 2 | 4 | 3 | 0 | 0 | 8 | 31 | 5 | 11 | 3 |
| indeno(1,2,3-cd)pyrene (In) | 6 | 2 | 7 | 2 | 1 | 2 | 6 | 1 | 3 | 23 | 1 |
| dibenz(a,h)anthracene (Db) | 1 | 1 | 4 | 4 | 21 | 7 | 3 | 8 | 0 | 46 | 2 |
| benzo(ghi)perylene (Bpe) | 19 | 2 | 4 | 2 | 1 | 5 | 15 | 1 | 5 | 9 | 2 |
| Total % | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| ***Sample number belonging to each group*** | | | | | | | | | | | |
| diesel vehicle exhaust [D] | 1 | 49 | 5 | - | - | 2 | 2 | - | - | - | 20 |
| gasoline vehicle exhaust [G] | - | 8 | 2 | 10 | 8 | 1 | 2 | - | - | 1 | 21 |
| tire [T] | 8 | - | - | - | - | - | - | - | - | - | - |
| asphalt-pavement [P] | - | 2 | - | - | - | - | 6 | - | - | - | - |
| asphalt or bitumen [A] | - | - | - | - | - | - | 9 | - | - | - | 2 |
| petroleum products excluding tire and asphalt [O] | - | 1 | - | - | - | - | - | - | - | 2 | 7 |
| combustion products except for those in vehicle engines [E] | - | 1 | 1 | - | - | - | - | 7 | 5 | - | 6 |
| Total number of sample | 9 | 61 | 8 | 10 | 8 | 3 | 19 | 7 | 5 | 3 | 56 |

*shaded cells are characteristic PAHs species/source categories in each group

***Source contributions to PAHs in road dust***

The estimated source contributions to PAHs in size-fractionated road dust were shown in Figure 1. Total vehicle volumes per day (VV) and ratios of *passenger* car volume to *freight* car volume (P/F) for each street (Ministry of land, infrastructure and transport Japan, 1999) were mentioned at the bottom of the figure. The result indicated that the predominant PAHs contributor to road dust varied with time, with particle size or with sampling location in the same road, except for the samples collected at the avenues Aratamagawasuido and Kanpachi. In the case of of Keiyo avenue, for example, PAHs in 010919 i and 010919 ii samples (meaning 2 different samples to be collected in 19 September, 2001) derived from diesel exaust (S2) and pavement (S7) while the 011004 samples collected two weeks later were significantly influenced by tire debris (S1). It is interesting that different particle size fractions has different major PAHs contributors, for example in Sakurada avenue samples, S2 (diesel exaust) contributed to all the size fractions, S7 (pavement) appeared in the finest and 106-250 μm fractions and S1 (tire) showed a significant contribution in 45-106μm fraction. This result suggested the importance of estimating PAHs source contribution to each particle size separately. Coupled with the percentage of total PAHs in each road dust fraction, the comprehension of major PAHs source in each particle size can provide an effective option for control strategies of PAHs in road runoff.

Comparative source contributions for whole (non-fractionated) road dust were also calculated, based on the particle size distribution of road dust and the source contribution to each fraction (Figure 2). For PAHs in the road dust of Kannana and Kiyosumi avenues, more than 50% was assigned to diesel vehicle




| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| VV | 34,718 | 146,905 | 75,060 | 4,931 | 76,928 | 24,523 | 61,054 | 44,920 |
| P/F | 1/4.9 | 1/2.3 | 1/1.9 | 1/1.9 | 1/1.61 | 1/1.73 | 1/1.70 | 1/1.73 |

: S1, S2, S3, : S4 : S5, : S7, : S9, S11

*Figure 1 Source contribution to PAHs in size-fractionated road dust*

exhaust and tire wear as major sources. While in case of the avenues of Sakurada, Aratamagawasuido, Oume and Showa, greater than 55% of PAHs in road dust was apportioned to diesel vehicle exhaust and asphalt-pavement debris as main contributors. This result supported the hypothesis that tire and pavement were worn away into particles during vehicle traveling, especially in case of diesel engine vehicles such as heavy-duty trucks. Overall, despite of the difference in vehicle volume or the ratio of passenger car to freight car, it can be said that PAHs in road dust samples originated from any of 3 sources: diesel vehicle exhaust, tire and asphalt-pavement.

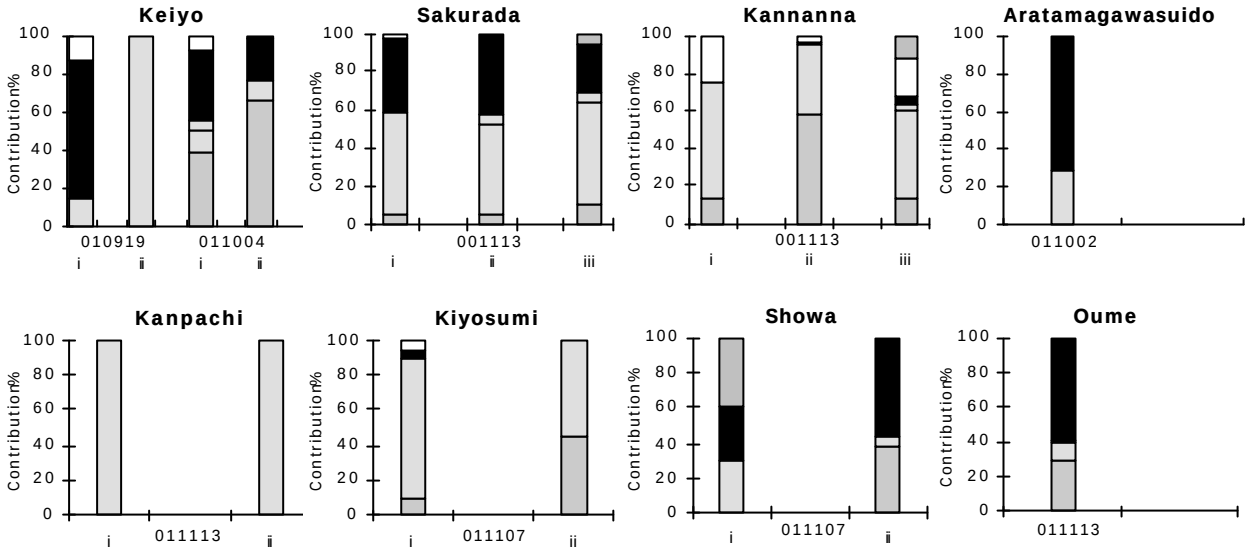



*Figure 2 Comparative source contribution to PAHs, calculated from particle size distribution of road dust and the result of Figure 1 (Legend is same with Figure 1)*

This study proposed one approach using PAHs profile data for source distribution assessment. Similar approach can be applied for relevant further studies, such as the evaluation of road dust contribution to water environment or the source assessment of other pollutants in road dust. However, these results call for further investigation. The variation of source type in each classified source group leads to the necessity of involving other significant substances in the contribution evaluation. Moreover, since numbers of PAHs profiles for source classification used in this study were taken from various journal papers and reports adopting a variety of sampling and analytical methods, the inclusion of some errors in the result remains as a matter to be discussed further.

## CONCLUSIONS

This study aimed to estimate the origins of polycyclic aromatic hydrocarbons (PAHs) in size-fractionated road dust in Tokyo. The 189 possible source data of 12-PAHs profiles were classified into 11 groups (from S1 to S11) based on cluster analysis combined with principal component analysis . In order to estimate the contributions of the classified source groups (S1-S11) to PAHs in the road dust, multiple regression analysis was performed.

The result demonstrated that abrasion of tire and asphalt-pavement provided a certain amount of PAHs to road dust, in addition to diesel vehicle exhaust, which has been recognized as the main source of PAHs in road dust. This information will be useful for establishing the control strategies of PAHs in road runoff.

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