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Size- and density-distributions and sources of polycyclic aromatic hydrocarbons in urban road dust

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) present in size- and density-fractionated road dust were measured to identify the important fractions in urban runoff and to analyse their sources. Road dust was collected from a residential area (Shakujii) and a heavy traffic area (Hongo Street). The sampling of road dust from the residential area was conducted twice in different seasons (autumn and winter). The collected road dust was separated into three or four size-fractions and further fractionated into light ($<1.7\ g/cm^3$) and heavy ($>1.7\ g/cm^3$) fractions by using cesium chloride solution. Light particles constituted only $4.0 \pm 1.4\%$, $0.69 \pm 0.03\%$ and $3.4 \pm 1.0\%$ of the road dust by weight for Shakujii (November), Shakujii (February) and Hongo Street, respectively but contained $28 \pm 10\%$, $33 \pm 3\%$ and $44 \pm 8\%$ of the total PAHs, respectively. The PAH contents in the light fractions were 1–2 orders of magnitude higher than those in the heavy fractions. In the light fractions, the 12PAH contents in February were significantly higher than the 12PAH contents in November ($P < 0.01$), whereas in the heavy fractions, no significant difference was found ($P > 0.05$). Cluster analysis revealed that there was a significant difference in the PAH profiles between locations rather than between size-fractions, density-fractions and sampling times. Multiple regression analysis indicated that asphalt/pavement was the major source of Shakujii road dust, and that tyre and diesel vehicle exhaust were the major sources of finer and coarser fractions collected from Hongo Street road dust, respectively.

Keywords: PAHs; Source analysis; Size- and density-fraction; Urban runoff; Time variation

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are diffusely deposited onto impervious areas and may have adverse effects on the aquatic ecosystem through urban stormwater runoff (Maltby et al., 1995a,b). Since urban runoff behaviour of particles and the PAH contents in aerosol particles, road dust and runoff are known to depend on the particle size-distribution (Furumai et al., 2002; Murakami et al., 2003, 2004; Fang et al., 2004), several studies have focused on runoff behaviour of PAHs in relation to the size-fractions. However, runoff behaviour of particles depends on the particle density-distribution as well as the size-distribution. Since the density of particles differs according to their sources, the size and density of particles are the key factors to

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control PAH wash-off to an aquatic environment. PAH distribution, by size and density, in harbour sediment fractions has been reported (Ghosh et al., 2000; Rockne et al., 2002; Ghosh et al., 2003). Rockne et al. (2002) revealed that 85% of the total PAHs in Piles Creek sediment were found in the light fractions (<1.7 g/cm^3), despite the fact that the light density components comprised only 4% of the total sediment mass. In addition, they suggested that the preferential sequestration in the Piles Creek sediment was likely due to the presence of detrital plant debris. Ghosh et al. (2000) showed that the coal/wood-derived particles (specific gravity < 1.8) constituted only 5% of Milwaukee Harbour sediment by weight but contained 62% of the total PAHs. However, little information is available on PAH distribution in size- and density-fractionated road dust and its sources, although urban runoff has been recognized as an important PAH pathway to sedimentation at harbours (e.g. Hoffman et al., 1984; Brenner et al., 2002).

Table 1 shows the density of particles from sources such as tyre, pavement, asphalt, diesel exhaust particles as well as particles from the earth's crustal erosion, which are considered to contribute PAHs in road dust (Hänel and Thudium, 1977; National Astronomical Observatory, 1994; Park et al., 2003; Hearings from tyre and construction companies and tyre association: Dunlop. The Japan Automobile Tyre Manufacturers Association, Nippon Road, Taiheiyo Cement and Watanabe-Gumi, 2003). Since the PAH profiles (fractional patterns of individual PAH content to total PAH content) in urban dust depend on the contributions from sources such as tyre, pavements, asphalt and vehicle exhaust (Pengchai, 2002; Murakami et al., 2003; Pengchai et al., 2004, 2005), the PAH profiles and the contents are believed to differ among fractions differing in size and density. In this study, PAHs were measured in size- and density-fractionated road dust. The objectives of this study are (1) to find the important fractions in urban runoff for reducing pollutant load of particle-bound PAH, (2) to examine the sampling time variation of PAHs in each fraction and (3) to analyse the sources of the PAHs by statistical methods.

2. Materials and methods

2.1. Road dust samples

Road dust was collected under wet conditions with a vacuum cleaner (Puzzi100, Kärcher) from Shakujii, Nerima Ward (a residential area) and from Hongo Street, Bunkyo Ward (a heavy traffic area) in Tokyo, Japan. In the residential area, infiltration facilities such as permeable pavement, infiltration inlets and infiltration trenches have been placed since 1980s. Two litres of deionised water was sprayed and vacuumed with the vacuum cleaner to collect road dust from a road gutter surface of 1250 cm^2. Deionised water was passed through the device to prevent cross-contamination after sampling. The sampling of road dust at the residential area was conducted twice in different seasons (autumn and winter). The sampling conditions in this study are summarised in Table 2. Fig. 1 shows the sampling timing with rainfall data from Shakujii. Significant differences in surface loads of road dust were observed between the two samplings at Shakujii, possibly due to the rainfall history before samplings.

2.2. Size- and density-fractionation

The collected road dust was size-fractionated by wet sieving using four stainless-steel sieves (2000, 250, 106, 63 μm), resulting in the following fractions: <63, 63–106, 106–250 and 250–2000 μm. The size-fractionated road dust was further fractionated into light (<1.7 g/cm^3) and heavy (>1.7 g/cm^3) fractions by cesium chloride solution by modifying the methods descried by Ghosh et al. (2000) and Rockne et al. (2002). As the first step, the floating particles in water were separated by centrifuging the size-fractionated sample at 2200*g* for

Table 1
Density of particles

Source	Density (g/cm^3)
Tyre (Hearings from tyre companies and tyre association, 2003)	1.2–1.3 (as density of rubber material)
Pavement (Hearing from construction companies, 2003)	2.0–2.5 (void fraction: 20–0%)
Asphalt (National Astronomical Observatory, 1994)	1.04–1.40
Diesel exhaust particle (Park et al., 2003)	0.55–1.20[a] (engine running: 10%, mobility size: 0.22–0.05 μm) 0.32–0.95[a] (engine running: 50%, mobility size: 0.30–0.05 μm)
Particle from earth's crustal erosion (Hänel and Thudium, 1977)	2.6–2.7 for desert dust

[a] Effective density: $\rho_{eff} = \rho_{true} \times (d_{ve}/d_{me})^3$ (ρ_{eff}: effective density, ρ_{true}: inherent density, d_{ve}: volume equivalent diameter, d_{me}: mobility equivalent diameter).